Doctoral theses at NTNU, 2020:210

Fabrice Abunde Neba

Integrated Model-Based Frameworks for Synthesis of Anaerobic Treatment Process: Optimizing Operation Using Reactor Networks

Attainable Regions for Waste-to-Energy Optimization

Doctoral thesis

Fabrice Abunde Neba

ISBN 978-82-326-4768-2 (printed ver.)
ISBN 978-82-326-4769-9 (electronic ver.)
ISSN 1503-8181

Doctoral theses at NTNU, 2020:210

NTNU
Norwegian University of Science and Technology
Thesis for the Degree of Philosophiae Doctor
Faculty of Engineering
Department of Civil and Environmental Engineering

NTNU
Norwegian University of Science and Technology

NTNU
Norwegian University of Science and Technology

NTNU

Fabrice Abunde Neba

Integrated Model-Based Frameworks for Synthesis of Anaerobic Treatment Process: Optimizing Operation Using Reactor Networks

Thesis for the Degree of Philosophiae Doctor

Trondheim, June 2020

Norwegian University of Science and Technology
Faculty of Engineering
Department of Civil and Environmental Engineering

NTNU
Norwegian University of Science and Technology

NTNU
Norwegian University of Science and Technology

Thesis for the Degree of Philosophiae Doctor

Faculty of Engineering
Department of Civil and Environmental Engineering

ISBN 978-82-326-4768-2 (printed ver.
ISBN 978-82-326-4769-9 (electronic ver.
ISSN 1503-8181

Doctoral theses at NTNU, 2020:210

Printed by NTNU Grafisk senter

Abstract

Anaerobic treatment technology offers great potential towards achieving Sustainable Development Goals due to its ability to simultaneously breakdown pollutants, generate renewable bioenergy and recycle valuable nutrients from organic waste streams. However, the successful operation of anaerobic digestion (AD) process requires design of optimal process configurations that are well adapted to the characteristics of feedstock available.

Due to the highly complex nature of AD, involving multiple reactions with each catalysed by specific groups of microorganisms, multistage anaerobic digestion, in which multiple digesters are operated in a network configuration becomes highly invaluable. This is because such network configurations can optimize overall performance of the AD process by ensuring that the specific conditions under which each reaction step takes place is optimized. In addition, each digester has unique characteristics often making them more adapted to treat waste of specific characteristics than others, and thus utilizing one digester in one configuration may limit the possible combination of pathways, hence limiting overall performance.

Model-based design of anaerobic digesters is particularly important as the kinetics captured by AD models can predict operating conditions, volumetric gas production, process stability as well as effluent quality. In addition, systematic model-based approaches for design of anaerobic digestion systems significantly reduce the number of expensive prototype systems and time-consuming studies usually required to obtain and optimal configuration of anaerobic digesters.

Even though there exist several studies that use kinetic models to guide design of anaerobic digesters, published literature has primarily been geared towards describing the process of developing a given model to guide design of single stage digester configurations. Remarkably, little research has been carried out on model reliability analysis, especially for the synthesis of

multistage digester configurations. This thesis therefore provides both the theoretical background and illustrations (with practical application cases) for development and use of systematic model-based frameworks to guide design and operation of multistage anaerobic digesters irrespective of the information available to the designer. The study uses a methodological approach that develops synergy by systematically coupling model-based techniques (multicriteria decision making tools, practical identifiability, Monte Carlo simulation, adjoint based gradient optimization and attainable region theory) in an optimal framework for synthesis and optimization of anaerobic digester networks. The result of the approach is an optimal framework (decision support system) for synthesis and optimisation of anaerobic digester networks under four practical scenarios: (a) Synthesis based on model requirements or characteristics whereby the study considered the case of no model availability, one-stage kinetic models, two-stage kinetic models, kinetic uncertainty as well as changes in kinetic model structure; (b) Synthesis based on operational/ process objectives, whereby the study considered process stability and process performance (measured in terms of biogas production and organic matter reduction) as design objectives; (c) Synthesis based on economic objectives whereby the study developed digester economic evaluation models based on known economic feasibility indicators as well as macroeconomic parameters; and (d) Synthesis based on feedstock characteristics whereby the study considered two classes of organic substrates (industrial wastewater and animal manure) and analysed the effect of substrate characteristics on the performance targets and optimal configuration of anaerobic digester networks. The results have been captured in five journal publications with the contribution from each paper summarized as follows:

Paper 1 presents a framework that uses two-stage kinetic models and process objectives for digester synthesis (mainly methane productivity and volatile solids reduction) while considering the effects of substrate characteristics, using five types of animal manure. The results illustrate that a change in digested substrate significantly influences the operating limits (defined by the attainable region), optimized parameter, as well as the design configuration of the optimal digester structure. This observed substrate effect on attainable regions shows great promises as it paves the way for other substrates such as blackwater, food waste, lignocellulosic waste, as well as co-digested feeds to be considered.

Paper 2 presents a framework that uses two-stage kinetic models and stability objectives for digester synthesis (considering inoculum to substrate ratio and instantaneous methanogenic yield) while considering the effects of model structure and sources of inoculum used to start-up digester operation. The findings illustrate that the inoculum characteristics influences the structure of the kinetic model used to describe the growth of anaerobic microorganisms and hence the performance targets and digester configurations obtained.

Paper 3 presents a framework that uses one-stage kinetic models and economic design objectives for digester network synthesis (developing economic evaluation models based on known economic feasibility indicators as well as macroeconomic parameters), with industrial wastewater as feedstocks. The results illustrate that synthesis of the anaerobic digesters can be tackled using both technical and economic parameters such as payback period as well as country-specific macroeconomic parameters such as interest rate and renewable energy feed-in tariff rate. A change in the value of any of these parameters affects the optimal digester configuration.

Paper 4 presents a framework that requires no kinetic model for digester synthesis and couples fuzzy multicriteria decision tools with attainable regions for simultaneous synthesis of digester structures and selection of digester subunits considering both techno-economic and environmental aspects. This implies that for the same digester structure, defined in terms of plug flow and continuous stirred tank reactors, the subunits (mainly type of plug flow digester) will differ based on the practical considerations for operating the digester system.

Paper 5 presents one of the more significant findings of the study by introducing a framework that simultaneously analyse model reliability, quantifies uncertainty in model states and construct attainable regions that are self-optimizing. Hence, when using attainable regions for performance targeting and digester network synthesis, the results indicate that it is possible to propagate uncertainty of model prediction onto the attainable regions to obtain self-optimizing attainable regions, which is generally smaller than the attainable region but has an advantage of increased robustness.

Summarily, the study indicates that using digester networks as opposed to single digesters is able to bypass regions of lower reactivity and improve performance of the anaerobic treatment process. The decision support system should be considered the first point of contact, and used to compliment experiments during planning, design, scale-up and installation of anaerobic digestion plants involving multistage digesters. This will significantly reduce the number of expensive prototype systems and time-consuming studies usually required to obtain an optimal configuration of anaerobic digesters. It is also worth mentioning that even though the study is based on the anaerobic treatment process, the developed frameworks can be applied for synthesis and optimization of other biochemical processes.

Dedication

This PhD thesis is dedicated to the wholeheartedly committed team of Abunde Sustainable Engineering Group (AbundeSEG), a tech start-up leveraging on model-based techniques, artificial intelligence and digital technology to unlock Africa’s potentials in the Food-Water-Energy-Health nexus.

Acknowledgements

The most striking aspect of this PhD study is NOT that I have learned technical and scientific skills, but that throughout the PhD study, I have learned the ability to learn ANYTHING faster and more efficiently. The PhD journey has been a truly life-changing experience for me, and it would not have been possible without the support and guidance of many individuals and institutions.

I would like to first say a very big thank you to my main supervisor, Prof. Razak Seidu for all the support and encouragement he gave me, during both the long months I spent undertaking my field work in Ghana as well as the time I spent at NTNU Ålesund. Without his guidance, constant feedback and unwavering ability to drive success, this PhD would not have been achievable. More importantly, because of his enthusiasm for international development, he provided the funding opportunity for this thesis, through his project 'Upgrading Education and Research Capacity in Renewable Energy Technologies', funded by NORAD-EnPe. Permit me also seek this opportunity to express my sincere gratitude to the management of NORAD-EnPe.

Many thanks also go to Prof. Ahmad Addo who believed in me during my masters studies at Kwame Nkrumah University of Science and Technology (KNUST) and acted as the main catalyst that made it possible for me to obtain this PhD fellowship under Prof. Razak Seidu. I lack adequate words to express my profound gratitude to Prof. Ahmad for without him, this PhD wouldn't have been possible.

I am also grateful to Dr. Nana Asiedu for his invaluable technical support, encouragements and assistance throughout the challenging times of my PhD journey right from masters studies to completion of this PhD study. My experience with Dr. Nana has been very exciting, especially when some of the things I thought were possible only in my wildest dreams were in fact attainable, at least theoretically. Special appreciation also goes to Prof. Stein Wold Østerhus for his direction and guidance during my stay in Trondheim for PhD course work, and to Prof. John Morken for his support. I deeply appreciate their amazing dedication and genuine advice that culminated in the success of this PhD. Indeed, I deem it an honor to have tapped from their knowledge and expertise.

Special thanks also go to Mr. Joseph Aveyire, Mr. Ohene Asamoah and Mr. Bediako Alexander for their invaluable support during design and construction of the AR-inspired prototype of a novel anaerobic digester.

My sincere thanks also go to the Water and Environmental Engineering Group of the Department of Ocean operations and Civil Engineering in NTNU Ålesund. I would like to mention Kristian Fjørtoft, Hadi Mohammed and Hoese Michel Tornyeviadzi for their invaluable assistance of varying forms. The wonderful experiences I had with them in the course of my study would be forever etched on my memory. I am grateful to Hadi and Kristian for the warmth they extended to me during my time in Norway and for always making me feel so welcomed. The technical assistance of Michel for some of the complex mathematical and computational tools is highly commendable.

I also extend special thanks to The Brew-Hammond Energy Centre of Kwame Nkrumah University of Science and Technology, Kumasi, Ghana, for the warm hosting during my stay in Ghana for field work.

I also wish to thank my family and friends in Cameroon, Ghana and all over the globe for the constant love, emotional support, well wishes, inspiration, and prayers they offered me in pursuit of this dream. I am indeed proud of them all.

Finally, and most importantly, I wish to express my sincere gratitude to God almighty for the success of this PhD has not been by my strength, but by his will for my life.

List of publications (05)

The thesis is grounded on the following publications as numbered from one to five:

1. **ABUNDE NEBA, F**., ASIEDU, N. Y., ADDO, A., MORKEN, J., ØSTERHUS, S. W. & SEIDU, R. 2019d. Use of attainable regions for synthesis and optimization of multistage anaerobic digesters. ***Applied Energy***, 242, 334-350.
2. **ABUNDE NEBA, F.,** ASIEDU, N. Y., ADDO, A., MORKEN, J., ØSTERHUS, S. W. & SEIDU, R. 2019c. Simulation of two-dimensional attainable regions and its application to model digester structures for maximum stability of anaerobic treatment process. ***Water Research***, 114891.
3. **ABUNDE NEBA, F.,** ASIEDU, N. Y., ADDO, A., MORKEN, J., ØSTERHUS, S. W. & SEIDU, R. 2020. Biodigester rapid analysis and design system (B-RADeS): A candidate attainable region-based simulator for the synthesis of biogas reactor structures. ***Computers & Chemical Engineering***, 132, 106607.
4. **ABUNDE NEBA, F.,** ASIEDU, N. Y., ADDO, A. & SEIDU, R. 2020. Attainable regions and fuzzy multi-criteria decisions: Modeling a novel configuration of methane bioreactor using experimental limits of operation. ***Bioresource Technology***, 295, 122273.
5. **ABUNDE NEBA, F.,** TORNYEVIADZI, H. M., ØSTERHUS, S. W. & SEIDU, R. 2020b. Self-optimizing attainable regions of the anaerobic treatment process: Modeling performance targets under kinetic uncertainty. ***Water Research,*** 171, 115377.

Other related publications by the author during the PhD (06)

This section presents other publications written by the author (not included in the PhD thesis), during the period of his PhD studies, applying model-based techniques for synthesis and optimization of the anaerobic treatment process.

1. **ABUNDE NEBA** F., TORNYEVIADZI, M., ASIEDU N.Y., AHMAD A., MORKEN, J. ØSTERHUS, S. W., SEIDU, R. (2019). "Geometry, kinetic analysis and reactor network synthesis: Attainable limits for minimizing residence time of the anaerobic treatment process "***Computers & Chemical Engineering***
2. **ABUNDE NEBA, F.,** ASIEDU, N. Y., ADDO, A., MORKEN, J., ØSTERHUS, S. W. & SEIDU, R. 2019b. A coupled modeling of design and investment parameters for optimal operation of methane bioreactors: Attainable region concept approach. ***Renewable Energy.***
3. **ABUNDE NEBA, F.,** ASIEDU, N. Y., MORKEN, J., ADDO, A. & SEIDU, R. 2020b. A novel simulation model, BK_BiogaSim for design of onsite anaerobic digesters using two-stage biochemical kinetics: Codigestion of blackwater and organic waste. ***Scientific African***, 7, e00233.
4. **ABUNDE NEBA F.,** ASIEDU N.Y., AHMAD A., MORKEN, J. ØSTERHUS, S. W., SEIDU, R. (2019). " Attainable regions generated by mixing and biodegradation: A new strategy to analyze biomethane potential of complex organic substrates" ***under review in Biochemical Engineering Journal (BEJ-D-19-01158)***
5. **ABUNDE NEBA F.,** TORNYEVIADZI, M., ASIEDU N.Y., AHMAD A., SEIDU, R. (2019). " Can the operating limits of biogas plants operated under non-isothermal conditions be defined with certainty? Modeling self-optimizing attainable regions " ***revision submitted in Computers & Chemical Engineering (CACE_2019_993)***

6. ABUNDE NEBA F., TORNYEVIADZI, M., AHMAD A., ØSTERHUS, S. W., SEIDU, R. (2019). “Modeling attainable regions in a three-dimensional space: Synthesizing configurations of anaerobic digesters that minimize overall residence time" ***under review in Biochemical engineering (BEJ-D-19-01215)***

Other publications by the author during the PhD. Studies (10)

This section presents other publications by the author, during the period of his PhD studies applying model-based techniques for synthesis and optimization of processes other than anaerobic digestion.

1. ASIEDU, N. Y., **ABUNDE NEBA, F**., ADDO, A., 2019. Modeling the attainable regions for catalytic oxidation of renewable biomass to specialty chemicals: Waste biomass to carboxylic acids. ***South African Journal of Chemical Engineering***, 30:1-14.
2. **ABUNDE, N. F.,** ASIEDU, N. Y. & ADDO, A. 2019. Modeling, simulation and optimal control strategy for batch fermentation processes. ***International Journal of Industrial Chemistry***, 10, 67-76.
3. MENSAH, M., ASIEDU, N. Y., **NEBA, F**. A., AMANIAMPONG, P. N., BOAKYE, P. & ADDO, A. 2020. Modeling, optimization and kinetic analysis of the hydrolysis process of waste cocoa pod husk to reducing sugars. ***SN Applied Sciences***, 2, 1160.
4. BAMAALABONG, P. P., ASIEDU, N. Y., **NEBA, F. A**. & ADDO, A. 2020. Dynamic Behavior, Simulations, and Kinetic Analysis of Two-Dimensional Substrate–Product Inhibitions in Batch Fermentation Processes. ***Industrial & Engineering Chemistry Research***, 59, 9797-9807.
5. P. AGYEMANG; **F. ABUNDE NEBA;** P. P BAMAALABONG; NANA Y ASIEDU; A. ADDO; RAZAK SEIDU. Modelling the operating limits of selected feedstocks for bioethanol production: Product inhibition kinetics and fermenter network synthesis, ***under review in Chemical Engineering Research and Design (CHERD-D-19-01711)***
6. **F. ABUNDE NEBA;** P. AGYEMANG; P. P BAMAALABONG; NANA Y ASIEDU; A. ADDO; RAZAK SEIDU. Modeling maximum fermentation and reactor design

targets for bioethanol production: Use of economic objective functions with attainable regions. ***Submitted to Biomass and Bioenergy***

7. **F. ABUNDE NEBA;** P. AGYEMANG; P. P BAMAALABONG; NANA Y ASIEDU; A. ADDO; RAZAK SEIDU. Dynamic model-based simulation and optimization of submerged ethanol fermentation under conditions of substrate and product inhibitions.
8. P. P BAMAALABONG; **F. ABUNDE NEBA;** NANA Y ASIEDU; A. ADDO; Modeling and simulation of one-dimensional inhibitions of ethanologenic microorganisms in batch fermenters: mimicking the effects of wort characteristics, ***under review in industrial biotechnology (IND-2019-0042)***
9. E. ENDENE, L. L. YONG, S. M. DASSANAYAKE, GIDIGASU S.S.R, V. ANGGRAINI, **F. ABUNDE NEBA**, A. ASADI. Optimizing the contents of calcined seashell powder and treated coir fiber for improving the mechanical behavior of a tropical residual soil: A response surface methodology-based approach, ***submitted to Sustainable Materials and Technologies (SUSMAT_2020_168)***
10. AMO-AIDOO, A., HENSEL, O. KORESE, JK., **ABUNDE NEBA,** STURM, B. A framework for simultaneous optimization of energy efficiency and integration of hybridized solar energy in industrial plants: Bioethanol production from cassava in Ghana, ***submitted to Journal of cleaner production***

Conference participations: Oral (01)

1. **ABUNDE NEBA F**., ASIEDU N.Y., AHMAD A., MORKEN, J. ØSTERHUS, S. W., SEIDU, R. (2019). " A novel approach for simultaneous modeling of process configurations and self-optimizing operating targets for biogas plants" ***Accepted at 4th International Conference on Fossil and Renewable Energy***" February 2020 at Houston, TX, USA.

Book contributions (01)

1. **ABUNDE NEBA** F., AGYEMANG P., YAHAYA D. N., ENDENE E., EYONG G. RAZAK SEIDU (2020). Leveraging model-based approaches to unlock bioenergy potentials in enhancing green energy and environment, chapter of book Green Energy and Environment, ***under review in IntechOpen,*** the world's leading publisher of Open Access books

Author's contribution to the list of publications captured in the thesis

Paper 1

The author proposed and carried out all theoretical developments and numerical computations. The results were interpreted together with the co-authors and the author wrote the paper.

Paper 2

The author proposed and carried out all theoretical developments and numerical computations. The results were interpreted together with the co-authors and the author wrote the paper.

Paper 3

The author proposed and carried out all theoretical developments and numerical computations. The results were interpreted together with the co-authors and the author wrote the paper.

Paper 4

The author proposed the concept, carried out all theoretical developments and numerical computations and designed the experiments. The results were interpreted together with the co-authors and the author wrote the paper.

Paper 5

The author proposed the conceptual framework. The identifiability analysis, model parameter estimation and uncertainty quantification were performed by Hoese M. Tornyeviadzi while the attainable region analysis was performed by the author. The results were interpreted together with the co-authors and the author wrote the paper.

Table of Contents

List of Figures

Nomenclature

ABR	Anaerobic Baffled Reactor
ACR	Anaerobic Contact Reactor
AD	Anaerobic Digestion
AF	Anaerobic Filter
AFBR	Anaerobic Fluidized Bed Reactor
AHP	Analytical Hierarchy Process
AR	Attainable Region
ASBR	Anaerobic Sequencing Batch Reactor
COD	Chemical Oxygen Demand
CSTR	Continuous Stirred Tank Reactor
DAE	Differential algebraic equations
DSR	Differential Side-Stream Reactor
DSS	Decision Support System
EGSB	Expanded Granular Sludge Blanket
FAST	Functional Analysis System Technique
GHC	Green House Gas
ICR	Internal Circulation Reactor
IDEAS	Infinite DimEnsionAl State-space
ILP	Infinite Linear Programming
LP	Linear Program
MCDM	Multicriteria Decision Making
PFR	Plug Flow Reactor
RCC	Recursive constant control
SSE	Sum of squared error
TOPSIS	Technique of Order of Preference by Similarity to Ideal Solution
UASB	Upflow Anaerobic Sludge Blanket
UASSR	Upflow Anaerobic Solid-State Reactor
C	State concentration vector
K	Collinearity index
$r(C)$	Reaction rate vector
β	Vector of model parameters
τ	Residence time
C_f	Feed concentration vector
δ_k^{msqr}	sensitivity measure

Chapter 1: General introduction

1.1 Context, background and importance

1.1.1 Biomethane from anaerobic digestion: A source of sustainable energy

Energy poverty has been a major challenge to sustainable development, particularly in the developing world and remains an increasingly important topic in many discourses on international development. To put this into perspective, approximately two-thirds of Africa's population rely on wood and other biomass residues as source of energy and predictions show that about 650 million people will still lack access to electricity by 2030, 90% being in Sub-Saharan Africa (IEA et al., 2019). In addition to the unprecedented challenges facing societies as a result of energy poverty, the lack of reliable and clean energy is a social and economic tragedy, which results in other challenges including greenhouse gas (GHC) emissions, environmental deterioration and climate change. There becomes a need to devise technological solutions that ensure assess to energy is not only reliable and affordable, but also environmentally sustainable.

Taking all these into consideration, the anaerobic treatment process has become very popular due to its ability to simultaneously stabilize waste and reduce GHG emissions, generate renewable bioenergy as well as recycle valuable nutrients from waste streams (Jørgensen, 2009, Henze et al., 2008, Wang et al., 2007). In fact, recent studies have confirmed that anaerobic digestion is a robust technology that offers a great potential to reduce energy poverty in developing countries (Andersson et al., 2016). However, the successful operation of the anaerobic digestion process is only possible if the following two prerequisite factors are met: (1) Availability of a sustainable supply of organic feedstock and (2) design of optimal process configurations that are well adapted to the characteristics of available feedstock. Concerning the former, huge tones of organic waste (such as livestock manure, food waste and faecal sludge) are produced annually from various anthropogenic activities, which can serve as feedstock for the anaerobic treatment process. Surprisingly, disposal of these wastes still comprises a significant economic burden to developing countries in addition to the social and environmental challenges associated to poor waste management (Andersson et al., 2016). Concerning the latter (design of optimal process configurations), a wide variety of anaerobic digester systems have been developed (see Figure 1 for some examples), which can be classified in to three groups: conventional digesters (e.g. ASBR, CSTR, and PFR), sludge retention digesters (e.g. ACR, UASB, UASSR, ABR and ICR) and membrane digesters (e.g. AF, EGSB and AFBR) (Mao et al., 2015).

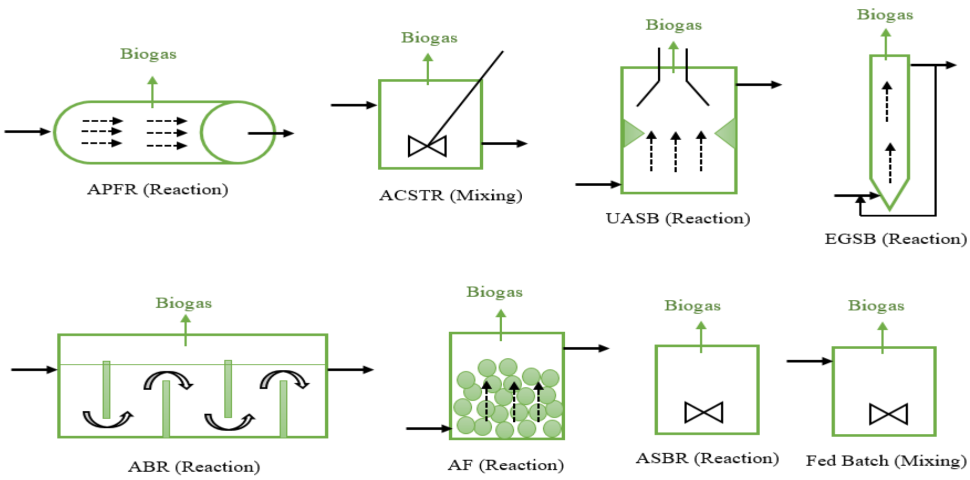

Figure 1: Schematic representation of selected anaerobic digestion systems

Recent studies continue to develop new digesters, which either modify the principle of an existing digester technology or present novel features, all geared towards improving process performance. However, the different anaerobic digesters have different physical and hydrodynamic characteristics making them more adequate to treat waste of specific characteristics than others. Understanding the individual characteristics of each digester is a starting point towards obtaining optimal process configuration for a generating biomethane from a given feedstock.

1.1.2 Anaerobic digestion process and reactor networks

Although various digester systems exist, each with unique physical and geometric characteristics, the hydrodynamic configurations of all digesters can be derived from different combinations of three fundamental regimes: flow regime, mixing regime and sludge retention regime, as illustrated in Figure 2.

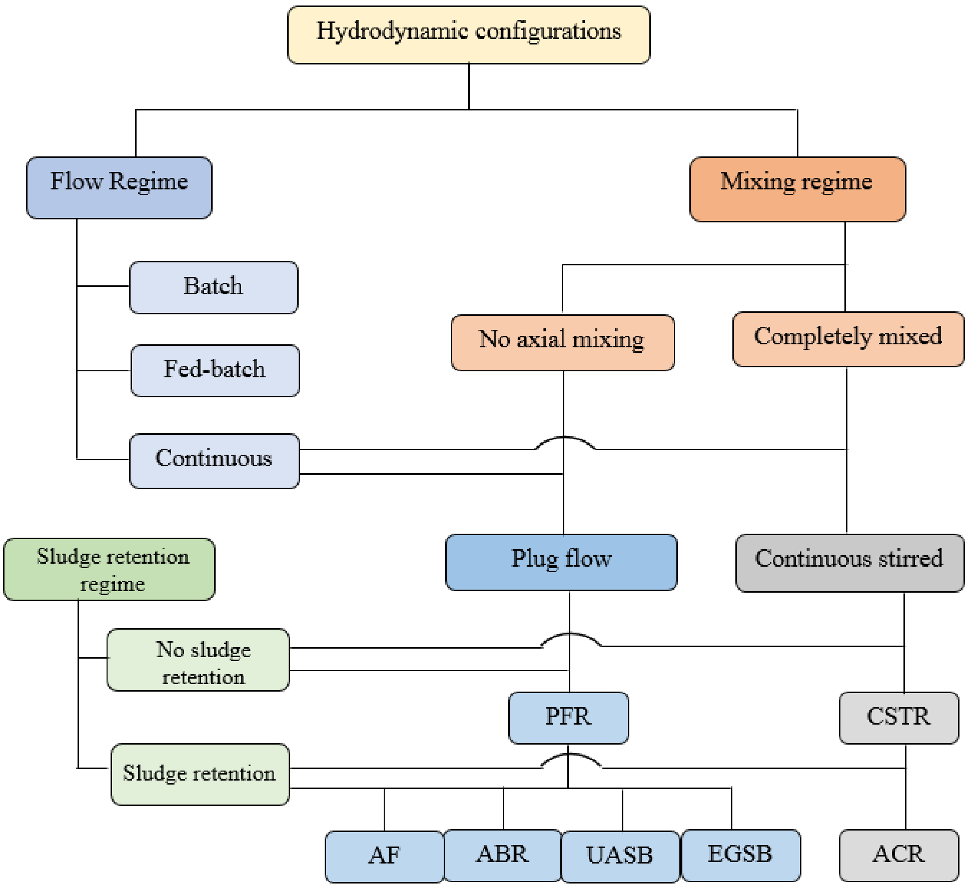

Figure 2: Classification of hydrodynamic configurations of anaerobic digester systems

Under flow regime, anaerobic digesters can be operated as batch, fed-batch or continuous; under mixing regime, they can be operated as completely mixed or with no axial mixing and under sludge retention regime, the operation can be with or without sludge retention. For example, a continuous flow regime operated with no axial mixing and no sludge retention gives a plug flow anaerobic digester (PFR) and when operated with sludge retention can result in either AF, ABR, UASB or EGSB. Other digesters can be derived by different combinations of the three fundamental regimes: flow, mixing and sludge retention regime. (Ming et al., 2016) illuminated critical aspects concerning a plug flow and continuous stirred tank reactors focusing on mixing and reaction. By their analogy of a plug flow reactor (reactor with no axial mixing along the length) as a series of batch reactors (reacting vessels) travelling on a conveyor belt, the Plug flow reactor can be considered a reaction reactor. The authors also illustrated that CSTR operates directly opposite to the PFR with respect to mixing due to its perfect mixing assumption where conversion of reactants into product is assumed to occur as a result of mixing and dilution rather than from reaction alone. The PFR and CSTR are therefore at the extremes of mixing and reaction and different combinations of these digesters will provide different extents of mixing and reaction in the entire system. In essence, all high rate digesters (sludge retention and membrane reactors) provide separation in addition to mixing and/or reaction. For example the anaerobic contact reactor provides mixing and separation while the anaerobic baffled reactor provide reaction and separation (Abunde Neba et al., 2019). What differentiates the high rate digesters is the mechanism in which separation is performed. High rate systems perform separation using two main mechanisms: (1) fixed microbial films on solid surfaces (membrane digesters); or (2) suspended microbial mass where retention is achieved through external (e.g. contact reactor) or internal (e.g. UASB) settling (Mes et al., 2003). As expected, the different mechanisms result in different extents of separation, each of which is more suited for different substrates characteristics or digestion objectives (nutrient recycling, energy generation or waste treatment) than others. Therefore, irrespective of the type of digester technology, the performance of the anaerobic treatment process depends on three fundamental processes, mixing (performed by CSTR) reaction (performed by PFR) and separation (performed by high rate systems). Therefore, the performance targets or limits of achievability of the anaerobic treatment process depends on the combination of reaction, mixing and separation only. What this means is that instead of focusing attention to devise new or perhaps novel digesters with the aim of improving the systems performance, it would be more important to focus attention on optimally arranging combinations of PFR, CSTR and/or high rate systems, or integrating more fundamental processes to the anaerobic treatment process (e.g. reversed osmosis + anaerobic digestion). More interestingly, the anaerobic digestion process involves multiple reactions and when operated as a single stage, the process conditions are only suitable for all the reactions with no particular reaction being optimized (EPA, 2006)**.** As illustrated in section 1.1.1, each digester has unique characteristics often making them more adapted to treat waste of specific characteristics than others, and thus utilizing one digester in one configuration may limit the possible combination of pathways, hence limiting overall performance. Multistage anaerobic digestion, in which multiple digesters are operated in a network are designed to optimize each biochemical step of the AD process by ensuring that the specific conditions under which each reaction step takes place is optimized (Zhang et al., 2017). Multistage systems also allow for operational flexibility, by easily adapting specific system configurations to meet different operational targets (EPA, 2006). For instance, if a multistage anaerobic digestion facility wants to produce more of hydrogen as opposed to methane gas, it might require a thermophilic stage; however, if organic matter reduction or methane gas production is its primary objectives, mesophilic stages may be prioritized. The plant might also be configured in a manner that optimizes production of both biohydrogen and methane gases. Summarily, because of the multiple biological steps involved in anaerobic digestion, using multi-stage systems as opposed to single stage systems can lead to significant improvements in quality of treated effluent as well as methane gas productivity. Therefore, if designers begin to think about anaerobic digesters in terms of digester networks as opposed to single digesters then more effective designs may be achieved overall.

1.1.3 Premise for model-based synthesis of anaerobic digester networks

The modeling of anaerobic treatment process is a key instrument to improve process performance and over the years, various models of varying complexity have been developed and applied in three main aspects: operational analysis, technology development, as well as digester design. Model development for digester design is particularly important as the technical and economic parameters of anaerobic digesters determined from the design process is a key motivation to guide investment decisions required to deploy biogas facilities into production. In addition, the kinetics captured by anaerobic digestion (AD) models is highly important for an optimal digester design since operating conditions, volumetric gas production, process stability as well as effluent quality can be predicted (Batstone, 2006, Haugen et al., 2013, Kythreotou et al., 2014, Yu et al., 2013). The modeling of anaerobic treatment process is a mature research area, now with a strong shift from model development towards model reliability analysis aimed at using kinetic models to solve various design and operational challenges. Even though there exist several studies that use kinetic models to guide design of anaerobic digesters, published literature has primarily been geared towards single stage digesters. As mentioned in section 1.1.2, single stage systems are limited to specific waste streams (due to their unique characteristics) and limit overall combination of biochemical pathways involved in the anaerobic treatment process. On the other hand, there still exist a high degree of empiricism in the design of multistage digester networks, often requiring construction of expensive prototype systems and time-consuming studies. For instance, during design of multistage systems, it is conventional for studies to predefine several candidate network configurations (referred to as dimensional space, example in Figure 3), from which the best configuration is chosen after experimental evaluation.

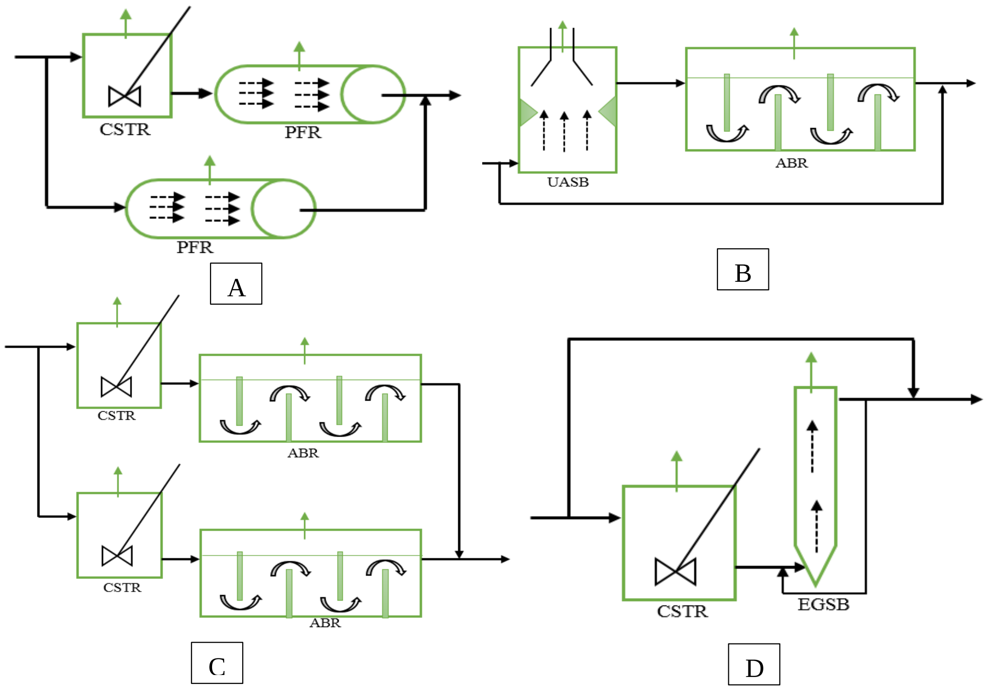

Figure 3: Conceptual configurations of anaerobic digester structures

This empirical approach to design of multistage digesters most often results in local minimum, with respect to process performance due to the relatively small dimensional space covered by the experiments. Most at times, due to economic constraints, studies do not even define a small dimensional space but just assume a given network configuration and only perform experimental evaluation of its performance. The optimal design of multistage digester systems requires a systematic approach to answer the following three questions: (1) How many individual digesters should be included in an optimal network (2) what type of digesters should be considered (PFR, CSTR, UASB, etc.) (3) Do we include recycle and bypass streams? If so, where are they placed within the structure? Very few writers have drawn on any systematic model-based procedure for design of anaerobic digester networks, and systematic procedures can further increase the ability of multistage digesters to improve process stability and operation as well as process economics. Model-based techniques for process synthesis are well-established for conventional chemical processes but limited in anaerobic digestion process and if such tools are used to compliment experiments during synthesis of anaerobic digester structures, it will present a breakthrough in expanding the application of anaerobic digestion technology to tackle energy poverty.

1.1.4 Statement of the Problem

In conventional process engineering, there are two major approaches for synthesis of reactor networks: superstructure optimization and attainable region targeting (Lakshmanan and Biegler, 1996). The superstructure optimization involves constructing a very large reactor superstructure, which usually suffers from the problem of local optimum (or multiple solutions). On the other hand, the AR targeting approach, which solves reactor network problem from a geometric point of view, presents a global optimization approach, which uses kinetic models to interpret chemical processes as geometric objects. This is followed by defining a region of achievability, which contains information about the performance targets and reactor structures required to achieve defined targets (Hildebrandt and Glasser, 1990, Horn, 1964, Ming et al., 2016). However, the use of AR in anaerobic digestion is greatly hampered by a couple of limitations, which can be categorized under three main groups: Reactor-based limitations, model-based limitations, and feedstock-based limitations. Concerning the reactor-based limitations, the attainable region targeting approach only yields results for continuous flow plug reactors, continuous flow stirred tank reactors or differential side-stream reactors. Recall from Figure 2 that there are several anaerobic digesters that can be considered to operate in plug flow mode, posing a challenge on what type of plug flow digester to use for a given optimal digester structure. Concerning the feedstock-based limitations, the anaerobic treatment process presents significant variability in terms of feedstock characteristics resulting in kinetic uncertainty as to what settings of kinetic parameters accurately describe the anaerobic degradation of a given substrate. The AR technique is unique for a given kinetics and feed composition, hence uncertainty in kinetics will result in digester structure uncertainty and possible failure during operation. Concerning the model-based limitations, there exist several types of kinetic models (with different degrees of complexity) describing the same steps of anaerobic digestion, making it a challenging task to know, which model is reliable for use in AR analysis for a given digestion process. These limitations explain why up until date, despite the strength of the AR technique, no study has been presented using the tool for synthesis and optimization of anaerobic digester systems.

A systematic approach for handling the above challenges and laying down theoretical framework for using attainable regions in optimizing the anaerobic treatment process will be breakthrough in advancing model-based design and operation of multistage anaerobic digesters. In fact, the retrofitting of multi-stage systems into facilities where single stage systems already exist is an important topic of discussion in most research concerning design and operation of anaerobic digestion facilities.

1.1.5 Brief synopsis of relevant literature

Much of the existing literature on modeling of anaerobic digestion pays attention to model development, describing the process of developing a given kinetic model but less effort geared towards assessing the performance or reliability of the model for design of anaerobic digesters (Batstone et al., 2002, Faisal and Unno, 2001, Zinatizadeh et al., 2006, Linke, 2006, Martinez et al., 2012, Borisov et al., 2016, Yang et al., 2016, Ware and Power, 2017, Esposito et al., 2011, Zaher et al., 2009). In addition, what is known about model-based design of anaerobic digesters is focused on using kinetic models to guide decision on engineering/operational characteristics such as: hydraulic retention time, organic loading rate, solids retention time, volumetric gas productivity, effect of temperature and pH, etc. (Lawrence, 1971, Henze et al., 2008, Wang et al., 2007, Linke, 2006, Yu et al., 2013, Yang et al., 2016, Lohani et al., 2018, López and Borzacconi, 2009, Kythreotou et al., 2014). Very few writers have drawn on any systematic studies for using models to guide synthesis of anaerobic digester structures, which is why the current study is designed to develop an integrated model-based framework, which includes the concept of attainable regions, for synthesis of anaerobic digester networks. The attainable region concept, a technique that incorporates elements of geometry to understand how networks of chemical reactors can be designed and improved was first introduced by the work on Horn (Horn, 1964). Following this initial work, many other researchers advanced AR research. Glasser et al. (Glasser et al., 1987) proposed a geometric approach that identified candidate AR's satisfying several necessary conditions that the AR must possess. Burri et al (Burri et al., 2002) demonstrated that, within the Infinite DimEnsionAl State-space (IDEAS) conceptual framework, construction of the true AR, and increasingly accurate AR approximants, can be carried out through Infinite Linear Programming (ILP), and a sequence of approximating finite Linear Programs (LP) respectively. Subsequently, Manousiouthakis et al (Manousiouthakis et al., 2004) developed, within IDEAS, necessary and sufficient conditions that the true AR must satisfy, proposed the Shrink–Wrap algorithm for AR construction, and established, this algorithm's equivalence to the aforementioned LP based AR construction methods. They also demonstrated that the true AR can be potentially larger than the candidate AR's identified by geometric methods. Zhou and Manousiouthakis (Manousiouthakis et al., 2004, Zhou and Manousiouthakis, 2006) demonstrated that the true AR for reactor networks involving only reaction and mixing may be smaller than the true AR for reactor networks also incorporating diffusion effects (e.g. by considering non-ideal dispersion reactor models). Zhou and Manousiouthakis carried out pollution prevention studies using the AR approach (Zhou and Manousiouthakis, 2007a), extended the AR approach to reactor networks involving variable density fluids (Zhou and Manousiouthakis, 2007b), discussed the dimensionality of the space in which AR construction can be pursued (Zhou and Manousiouthakis, 2008), and extended the AR approach to non-isothermal reactor networks

(Zhou and Manousiouthakis, 2009). Around the same time, Posada and Manousiouthakis (Posada and Manousiouthakis, 2008), proposed AR construction methods for reactor networks with multiple feeds, while Davis et al. (Davis et al., 2008) extended the AR approach to batch reactor networks. Seodigeng proposed an automated technique for attainable region construction using recursive constant control (RCC) policy algorithm (Seodigeng et al., 2009) More recently, Ming et al. (Ming et al., 2013) showed that by use of appropriate transformations, results developed from a continuous AR may be used to form a related batch structure, Conner and Manousiouthakis extended the AR approach to general process networks (Conner and Manousiouthakis, 2014), while Ming et al. (2016) summarized many of the theoretical and applied literature results on AR. As mentioned in section 1.1.4, published studies with regards to the use of AR for process synthesis and optimization have been well directed to other chemical processes but limited in anaerobic digestion. The AR technique has been applied to optimize various systems and processes, which include esterification process (Asiedu et al., 2015), production planning and scheduling (Sung and Maravelias, 2006, Sung and Maravelias, 2007), comminution process (Metzger et al., 2009), distillation process (Agarwal et al., 2008a, Agarwal et al., 2008b), separation and recycle process (Mcgregor et al., 1998), as well as polymerization process (Smith and Malone, 1997). For synthesis problems involving bioreactors, the few studies recorded include that of Scott et al. (Scott et al., 2013), who addressed the search of bioreactor configurations with improved residence times for continuous enzymatic saccharification and fermentation operations for bioethanol production. The only recorded study related to anaerobic digestion is that of Muvhiiwa et al. (Muvhiiwa et al., 2015) who used theoretical predictions of biogas production limits based on thermodynamics and attainable regions. However, these theoretical predictions based on elemental substrate analysis presume complete breakdown of organic matter (actual breakdown is usually between 27-76%) (Jingura and Kamusoko, 2017) as the effect of inhibitions, which normally occur during anaerobic digestion is not accounted for. In addition, no information is provided on the reactor designs required to achieve the defined limits. For this reason, it can mainly be used for comparing the biomethane potential of different organic substrates and cannot provide reliable economic estimates for real-time operation. A couple of reasons can be stated to justify why the concept has not been applied for synthesis and optimization of anerobic digester configurations. (1) Unlike conventional chemical processes, which receive influent of somewhat known composition, the anaerobic digestion process, normally applied to waste/wastewater treatment poses as main drawback uncertainty in feed characteristics, very common in AD plants. (2) Unlike conventional chemical reactors, which are classified as plug flow, CSTR or DSR, a wide variety of anaerobic digester systems have been developed, which are rather classified in to three groups: conventional digesters (e.g. ASBR, CSTR, and PFR), sludge retention digesters (e.g. ACR, UASB, UASSR, ABR and ICR) and membrane digesters (e.g. AF, EGSB and AFBR) (Mao et al., 2015). Unlike conventional chemical reactions where the kinetic constants (mostly order of the reaction) are relatively more defined due to less variability in feedstock characteristics and well-defined reaction invariants, the biodegradation reactions of anaerobic digestion present kinetic uncertainty as to what settings of kinetic parameters accurately describe the anaerobic degradation of a given substrate.

Owing to the complex and interactive nature of the challenges, the use of integrated model-based techniques can be advantageous in supporting decision making and making trade-offs within the significant and overlapping variabilities that exist amongst the feedstocks, digesters and kinetic models. In addition, coupling model-based techniques in a methodological framework eliminates the weaknesses of the individual techniques and makes it possible for applying an AR-based framework for synthesis of multistage anaerobic digester structures. However, due to the unfamiliarity of many researchers to the range of mathematical, analytical and computational techniques that could be applied in the field of anaerobic digestion, the advantage of model-based design is not being exercised in research. This thesis is therefore designed to fill this gap, by providing both the theoretical background and illustrations with practical application cases where some of the relevant systematic model-based techniques can be used to guide design and operation of anaerobic digesters. More importantly, some of the techniques will be modified or improved in order to enhance their suitability to the anerobic treatment process. Even though the presented methods have a wide range of other areas of application, the discussions and case studies presented in this thesis will be focused on anaerobic digestion. However, studies on other areas of application, conducted by the author of this thesis has been cited (see Table of Contents) for readers who are interested in getting more insight into the application of model-based techniques to unlock bioenergy potentials. The techniques presented in the thesis include:

- Multicriteria decision-making techniques to support policy formulation in the selection of digester subunits for use in synthesis of digester structures, considering both techno-economic and environmental characteristics
- Attainable region technique for performance targeting, synthesis as well as analysis of process configurations required to operate anaerobic digesters
- Monte Carlo simulation method for modeling uncertainty in model states resulting from variability in feedstock and kinetic characteristics.
- Practical/sensitivity-based identifiability for assessing the reliability of kinetic models by determining parameter subsets that can be accurately estimated to ensure model remains reliable for synthesis of anaerobic digester structures.
- Adjoint-based gradient optimization for estimating and identifying kinetic coefficients required to completely define a model before it can be used for digester synthesis.

A more detailed overview on how the different model-based tools are used to guide synthesis of multistage anaerobic digesters will be presented in section 2.1 (Chapter 2).

1.1.6 Scope and focus of the study

This thesis was designed to lay down theoretical framework and adapt/improve upon the use of the attainable regions for synthesis of anaerobic digester networks. The study used a conceptual approach whereby systematic theoretical frameworks are developed for digester synthesis based on different practical scenarios, which include:

a) Synthesis based on model requirements/characteristics whereby the study considered the following cases: no model availability, one-stage kinetic models, two-stage kinetic models, kinetic uncertainty as well as changes in kinetic model structure.

b) Synthesis based on operational/ process objectives, whereby the study considered process stability and process performance (measured in terms of biogas production and organic matter reduction) as design objectives.

c) Synthesis based on economic objectives whereby the study developed digester economic evaluation models based on known economic feasibility indicators as well as macroeconomic parameters.

d) Synthesis based on feedstock characteristics whereby the study considered two classes of organic substrates (industrial wastewater and animal manure) and analysed the effect of substrate characteristics on the performance targets and optimal configuration of anaerobic digester networks

1.2 Research goal and objectives

1.2.1 Research goal

The goal of this study is to develop a set of integrated model-inspired frameworks, which couples model development/configuration, practical identifiability, uncertainty quantification and elements of geometry for optimal synthesis of methane bioreactor structures, considering both technical and economic design objectives. This would lay down the theoretical and practical framework required for using attainable regions for synthesis and optimization of multistage anaerobic digesters, adaptable to various practical constraints, which can be model-based, reactor based, or feedstock based.

To realize this, it is required that the framework built can be used in the following three scenarios: when only simplified or semi-simplified models are available, when there exists uncertainty in kinetic models or other operational parameters and when there exist no reliable kinetic models to describe the process. In addition, the framework should be able to reveal the effect of substrate characteristics and model structure on the optimal network configuration of the multistage anaerobic digesters.

1.2.2 Specific objectives

In achieving the research goal, the study defined the following five specific objectives to be attained.

a) To analyze the physical and hydrodynamic characteristics of anaerobic digester systems, devise a generalized digester classification as well as introduce the use of fuzzy multicriteria decision tools in selecting digester subunits.
b) To develop and/or configure (model-dimensionality reduction, model identification, etc.) both one- and two-stage microbial kinetic models for use in synthesis of digester structures
c) To design both technical and economic scenarios for digester synthesis and develop objective functions, which can be used for model-based synthesis of multi-stage anaerobic digesters
d) To develop systematic methodological frameworks for synthesis of anerobic digester structures that integrates the effect of model structure, kinetic uncertainty, substrate characteristics and can be used even when no kinetic model is available

e) To design (from an engineering point of view) a novel prototype of an AR-inspired digester superstructure easily adaptable to different scenarios presented in (c) and (d)

1.3 Organization of the thesis

The overall structure of the study takes the form of four chapters, including this introductory chapter. Chapter 2 presents a synopsis of the research methods/techniques followed by a detailed conceptual synthesis of the research framework. The chapter summarizes how the research goal, the specific objectives, the research techniques are logically connected to one another and how the publications appended in the thesis are derived from the main objective of the study. Chapter 3 presents a summary of the main findings in the form of a decision support system and discusses the scientific and practical significance of the results. The final chapter draws upon the summary and conclusion of the entire thesis, tying up the various theoretical and empirical strands and presenting both industrial recommendations as well as perspectives for further research.

Chapter 2: Research methods and conceptual framework

2.1 Synopsis of research methods and data sources

The approach taken in this study is a mixed methodology based on five main mathematical methods/techniques: Attainable regions for performance targeting and synthesis of anaerobic digester structures, multicriteria decision making tools (particularly the AHP-fuzzy TOPSIS method) to support selection of digester subunits, practical identifiability for model analysis and determination of model parameter subsets that can be reliably estimated, Adjoint-based gradient optimization for parameter estimation and the Monte Carlo simulation procedure for model uncertainty quantification. The research data for the study is drawn from two main sources: (1) primary data from experiments conducted at the Department of Agricultural and Biosystems Engineering, Kwame Nkrumah University of Science and Technology, Kumasi Ghana (2) secondary data obtained from the literature. The next section presents an overview of the of each of the methods.

2.1.1 Attainable Regions

The AR theory is a technique for process synthesis and optimization, which incorporates elements of geometry to understand how networks of chemical reactors can be designed and improved. The attainable region is defined as the set of all possible output for all possible reactor designs that can be achieved by using the fundamental processes occurring within the system and that satisfies all the constraints placed by the system. Geometrically, the attainable region represents the region bounded by the convex hull for the set of points achievable by the fundamental processes occurring in the system. Once the AR has been determined, the limits of achievability by the system for the given kinetics and feed point is known and the boundary of the AR can then be used to answer different design or optimization questions related to the system.

The following section outlines the methodological flow for AR construction and application for process synthesis and optimization. The framework involves five main steps (Ming et al., 2016):

Step 1: Preparation

This involves definition of the reaction kinetics, AR dimension, state variables (those used to represent the AR) as well as the feed point used to generate the AR. The feed point defines the initial value, or concentration of states fed into the reactor. Defining the reaction kinetics entails determining the specific type of kinetic models as well as the settings for the kinetic coefficients. The dimension of the AR is determined from the number of independent reactions occurring in the reactor system, which defines the dimension of the stoichiometric subspace (the rank of the stoichiometric coefficient matrix A), in which the AR must reside. The number of independent rows or columns of the stoichiometric coefficient matrix A (rank of A) gives the number of independent reactions in the system. A key criterion for choosing variables for representation of the attainable region is that the variables must follow the linear mixing law.

Step 2: AR construction

This step generates the AR using a combination of fundamental reactor types, which includes: PFR, CSTR and mixing for two-dimensional ARs or a combination of PFR, CSTR, DSR (Differential side-stream reactor) and mixing for higher dimensional constructions. This is the most difficult and time-consuming step but also provides the most valuable information about the operating limits of the system. AR construction typically begins by determining the PFR trajectory and CSTR locus from the feed. The PFR trajectory is the set of points generated by solving the steady state model of a PFR reactor (a set of ordinary differential equations) while the CSTR locus is the set of points generated by solving the CSTR model (a set of nonlinear equations). In AR theory, mixing is performed by a continuous stirred tank reactor (CSTR) while reaction (biodegradation) is achieved in a plug flow reactor (PFR), since the operation of both reactors respectively mimic the two fundamental processes. At steady state operation, the general mathematical representation of a CSTR and PFR are given by Eqs. (1) and (2) respectively.

$$C = C_f + \tau r(C) \quad (1)$$

$$\frac{dC}{d\tau} = r(C) \quad (2)$$

C is the state vector while $r(C)$ is the reaction rate vector.

Solving the CSTR system to obtain the roots at a given feed point (C_f) and for different residence times ($\tau_i \ for \ i = 1 \ to \ n$) results in a set of points referred to as a CSTR locus. In the same way, integrating the PFR system for a given feed point and residence time results in a set of points referred to as PFR trajectory.

Step 3: Boundary Interpretation

This step involves interpretation of the AR boundary into reactor structures, based on the fundamental characteristics of the AR boundary. The boundary of the AR is composed of reaction and mixing surfaces only. Reaction surfaces are always convex and the points that form convex sections of the AR boundary arise from effluent concentrations specifically from PFR trajectories. For a two-dimensional system, points on the AR boundary that initiate these convex PFR trajectories arise from specialized CSTRs while for a three-dimensional system, they arise from DSRs. The convex hull of the set of points generated by all possible combinations of fundamental reactor types and mixing defines the attainable region.

Step 4: Overlay objective function

The objective function is modeled in terms of the variables used to represent the AR and then overlaid onto the AR. For this study, two types of objective functions were used: (a) operational/ process objectives, whereby the study considered objectives based on process stability and process performance (measured in terms of biogas production and organic matter reduction). (b) Economic objectives whereby the study developed digester economic evaluation models based on known economic feasibility indicators as well as macroeconomic parameters. The points of intersection between the objective function and the AR boundary represent the optimal points of operation.

Step 5: Optimize

Since the entire boundary of the AR has been interpreted in terms of reactor structures (step 3), the reactor structure(s) required to achieve the optimal operating points (point of intersection) is known. One interesting aspect about the attainable region approach is that it contains solutions to all design and optimization problems. What this means is that there is no need to reoptimize when the objective function changes, all that is required is to overlay the new objective function on to the AR boundary and determine the points of intersection.

Summarily, starting from the feed point, the procedure entails finding all possible achievable outputs for the system under consideration, from the trajectory of the states of interest describing the system operation. These trajectories are convexified to obtain candidate attainable regions, which are tested against the necessary conditions and recursively updated so that any violated necessary conditions is eliminated. The process continues until no other necessary conditions are violated otherwise, a candidate AR (subset of the true AR) is obtained, which can still provide better understanding of the achievable limits of the system. Some necessary conditions for AR can be summarized as follows (Glasser et al., 1987, Glasser et al., 1993, Hildebrandt and Glasser, 1990, Hildebrandt et al., 1990):

- The AR includes all feed points to the system.
- The AR is convex.

- No rate vectors point out of the AR boundary.
- Backward extension of rate vectors in the complement region do not intersect the AR

It is not the intention of this section to present a detailed explanation of the AR theory. Interested readers can consult Ming et al. (Ming et al., 2016) for a more in-depth understanding. In addition, Chapters II to VI will provide more explanations by presenting specific application cases of the AR concept

2.1.2 Multicriteria decision making tools

The high level of variability in characteristics of the different anaerobic plug flow digesters makes selection of digester subunits a complex problem. Multi-criteria decision-making (MCDM) tools, which makes preference decisions over the available alternatives characterized by multiple (usually conflicting) attributes becomes indispensable in this case. Amongst the different MCDM based tools (Ibáñez-Forés et al., 2014, Kumar et al., 2017), the thesis integrates the Analytical Hierarchy Process (AHP) into the Technique for Order of Preference by Similarity to Ideal Solution (TOPSIS) technique in order to take advantage of the weighting strength of AHP and the ranking strength of TOPSIS. However, the use of ordinary Multicriteria decision making (MCDM) tools for ranking of alternatives requires that the performance score of the alternatives with respect to each criterion is quantitative in nature (i.e. can be measured and attributed a crisp numerical value). Some examples of quantitative attributes include: greenhouse gas emitted, recovered energy, recovered nutrients, operating cost, percentage COD removal, etc. For selection of anaerobic digester technologies, the performance score of the alternatives with respect to each criterion does not always have crisp numerical values and the ordinary MCDM cannot therefore be applied. The strength of this study is illustrated by extending the decision-making process to include fuzziness, where by ratings of alternatives versus criteria is done using linguistic variables represented as fuzzy triangular numbers (Kaya et al., 2019). This provides an opportunity of the decision-making process to be performed even in cases where crisp numerical ratings of the alternatives with respect to the criteria is not available or even in cases of uncertainty. The selection of the appropriate anaerobic plug flow digester was done using a hybrid of the Fuzzy Technique for Order Preference by Similarity to Ideal Solution (FTOPSIS) and the Analytical Hierarchy Process (**Paper 4**). At first, AHP is used to compute the criteria weights, which show the relative importance of the different attributes used for digester selection. Afterwards, the FTOPSIS method is applied to prioritize the different alternatives (plug flow digesters) based on the computed criteria weights. The algorithm for the integrated AHP-Fuzzy TOPSIS method utilized in this study is summarized in Figure 4.

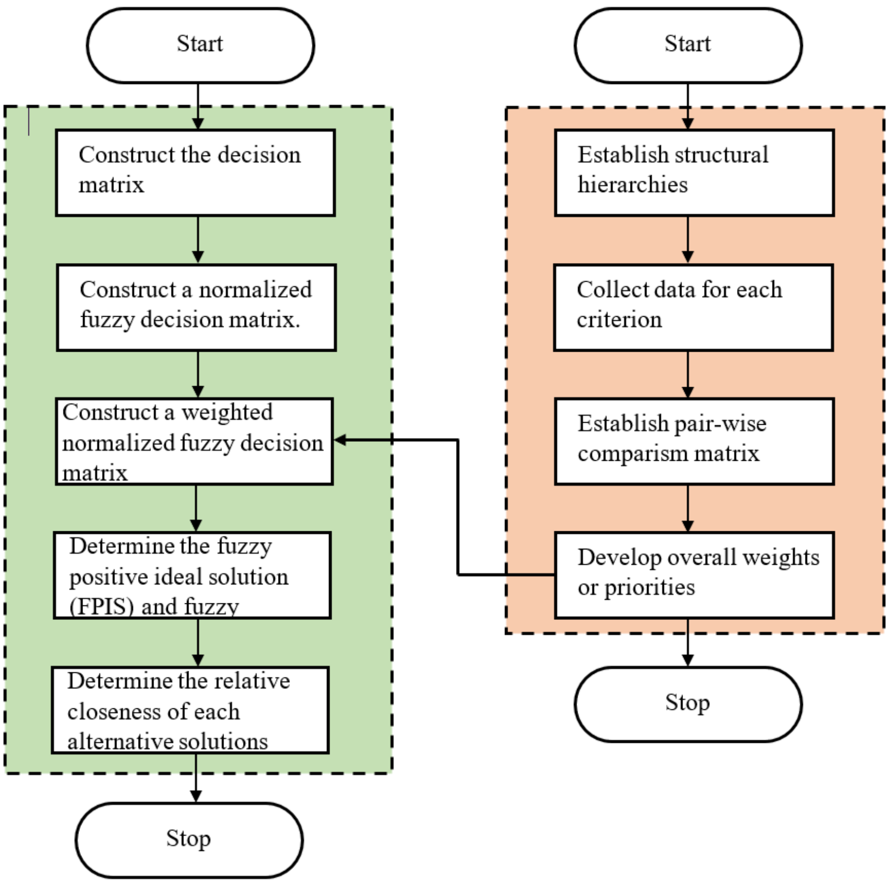

Figure 4: Integrated AHP-FTOPSIS model for multi-criterial decision analysis

Therefore, by coupling attainable region theory (for reactor network synthesis) and the fuzzy AHP-TOPSIS (for selection of digester subunits based on multiple attributes), a reliable tool is obtained, which does not only synthesize digester networks in a holistic way but is also able to select digester subunits within the network to perfectly match operational constraints.

2.1.3 Adjoint-based gradient optimization

Synthesis of anaerobic digesters using the attainable region technique requires well defined kinetic models, with optimal settings of kinetic coefficients. Some of the kinetic models used to describe the anaerobic treatment process are highly non-linear, involving species in the inoculum, liquid and gaseous phases. This results in a system of differential algebraic equations (DAEs), which need to be solved to completely identify the model parameters through the minimization of an objective function, which represents the error between the model solution and the experimental data. Most local optimization methods require the gradient of both the const function and the constraints and this is usually computed by numerical perturbations using the finite difference scheme. This study makes use of a two-step adjoint-based gradient algorithm (Figure 5), which computes gradients more accurately and efficiently (Benítez et al., 2017). Firstly, the gradient algorithm fits the whole set of model parameters and assesses the variability of the fit using marginal and joint confidence regions of the model parameters. Secondly, for parameters that show a high correlation, one of them is kept constant and a readjustment of the uncorrelated set of parameters is performed using algorithm. This allows a more accurate adjustment of the whole set of model parameters to experimental data.

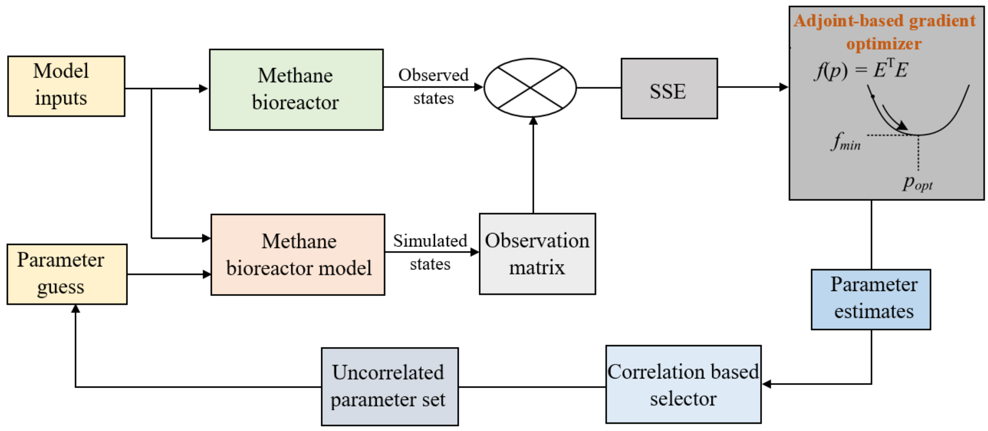

Figure 5: Model identification framework using the adjoint-based gradient optimizer

The advantage of this procedure is that parameter estimation and variability assessment is performed simultaneously, which allows the user to better understand the model's sensitivity to different influences and obtain reliable estimates

2.1.4 Practical/sensitivity-based identifiability

Even though the adjoint method presents a reliable method to estimate the kinetic constants of AD model, knowing the exact set of parameters that can be estimated to accurately and uniquely describe the mechanisms occurring in the digester is highly important. For a given kinetic model, there exist a maximum number of kinetic parameters that should be estimated (referred to as identifiable subset) to maintain the model's reliability. The characteristics of the identifiable subset and hence the reliability of a mechanistic model is influenced by three main factors: (1) the mathematical structure of the model, (2) the nature of the experimental data used for identification, and (3) the size of the identifiable subset. Attempting to estimate a higher than possible number of parameters (overparameterization) results in a model that can accurately describe the experimental data but loses is predictive or exploratory capabilities

(Donoso-Bravo et al., 2011). The techniques in practical identifiability analysis therefore serve to increase the reliability of the model for synthesis of anaerobic digesters.

Practical identifiability consists of analyzing the model's sensitivity to provide dynamic information on how the states (or model outputs) vary with changes in the settings of kinetic constants. This information is useful in identifying time intervals where the AD process is most sensitive to such fluctuations and experimental data points carry more importance for the parameter estimation process. The sensitivities are then used to screen for parameter significance ranking by calculating a sensitivity measure, δ_k^{msqr} and for analyzing the near-linear dependency between parameters by a measure called the collinearity index, K.

Given a model for a process, the following five key steps needs to be performed in order to completely assess the reliability and usage of the model (Brun et al., 2002):

Step 1: Compute the absolute sensitivity

Due to the nonlinearity of the AD models, an explicit solution to the differential equation model is normally not possible and the absolute sensitivities must be computed using the sensitivity equations. The sensitivity equations are coupled with the original model differential equations and solved numerically to obtain the parameter sensitivities for the necessary time points.

Step 2: Compute the non-dimensional sensitivity

The observable states have different physical meanings and different domains of variation. In order to ensure that the sensitivities are comparable, they are scaled using a weighting matrix, resulting in scaled sensitivities.

Step 3: Compute the sensitivity measure

From the matrix of non-dimensional sensitivities, an overall scoring for each parameter, called root mean squared sensitivity, δ_k^{msqr} is computed to consider changes in time or across experiments. The sensitivity measure (δ^{msqr}) measures the relative importance of the parameters with respect to how they influence the model outputs (states). The higher the magnitude of the sensitivity measure the more important the influence of the parameter on the states.

Step 4: Compute the normalized sensitivity

From the matrix of non-dimensional sensitivities, the normalized sensitivity for each parameter is computed.

Step 5: Compute the collinearity index

The final step consists of computing the collinearity index γ_K where K stands for the index of the parameter subset, which is a combinatorial function of the parameter vector β. If the sensitivity functions of two or more parameters are orthogonal (implying parameters are independent), the index of that parameter subset (K) is equal to unity, but if the parameters are linearly dependent, the index approaches infinity. In order to find an identifiable parameter subset, a threshold value (1-15) is usually used where by any parameter subset having index (K) greater than the threshold is said to be unidentifiable (Sin and Vanrolleghem, 2007, Brun et al., 2002).

2.1.5 Monte Carlo simulation procedure

As stated in section 2.1.4, using an an identifiable parameter subset reduces uncertainty in the parameter settings obtained from the parameter identification process. However, another serious challenge, which often arise when using an identifiable parameter subset is that of uncertainty in model states. This is because those parameters that are found not identifiable need to be kept constant (probably using values estimated from previous studies or independent experiments), which influences the reliability of the model. Since the geometric optimization technique of attainable regions is based on achievability of states by the system, uncertainty in model states results in uncertainty in performance targets and hence unreliable digester systems, which can easily lead to operational failure. In order to therefore use the model to define reliable performance target, one needs to quantify the model prediction uncertainty onto the attainable regions, resulting in the so-called self-optimizing attainable regions. The quantification of the model prediction uncertainty is done using the Monte Carlo simulation procedure, which is performed using four main steps (Sin et al., 2009, Morales-Rodriguez et al., 2012):

Step 1: Define the input/parameter uncertainty range or sample set

Input-output uncertainty analysis is highly dependent on the input uncertainty range (confidence bounds) as well as correlation coefficients. The variance metrics and correlation coefficients of the unidentifiable set of model parameters is obtained by estimating the complete set of parameters (identifiable and unidentifiable) using the estimation procedure presented in section 2.1.3.

Step 2: Determine the sampling method from the input set

This step consists of specifying the procedure or procedural rules that produces a pseudo-sample, which is used as inputs to the kinetic model of the anaerobic digestion process. The sampling rules must correspond to the characteristics of the sample set defined in step 1.

Step 3: Perform Monte-Carlo simulations

This step consists of simulating the AD process several defined numbers of times, each time using a different set of inputs obtained from the sample set by applying the sampling procedure presented in step 2.

Step 4: Compute the output uncertainty.

Calculate the probabilities or statistical coefficients of interest using the tabulation of outputs obtained from the Monte-Carlo simulations.

2.2 A conceptual synthesis of the thesis structure

After having stated the problem, defined the objectives as well as the methods to be utilized in the study, a systematic approach, the Functional Analysis System Technique (FAST) was used to obtain a work breakdown structure and arrive at the elemental components required to achieve the overall goal of the thesis. The advantage of this approach is that the FAST aids in thinking about the research problems systematically, clearly illustrating the publications where the different components are located and showing the logical relationships between goal of the thesis, specific objectives, research edge as well as the research methods. This enables the determination of all the necessary publication areas and a clear illustration of how all the papers complement one another to achieve the overall thesis goal of the thesis. Put in another way, the FAST representation moves the focus away from the interesting technical and scientific concepts presented in the manuscripts and places emphasis on how each manuscript contributes in solving the problems. Figure 6 presents a FAST-based representation of the thesis.

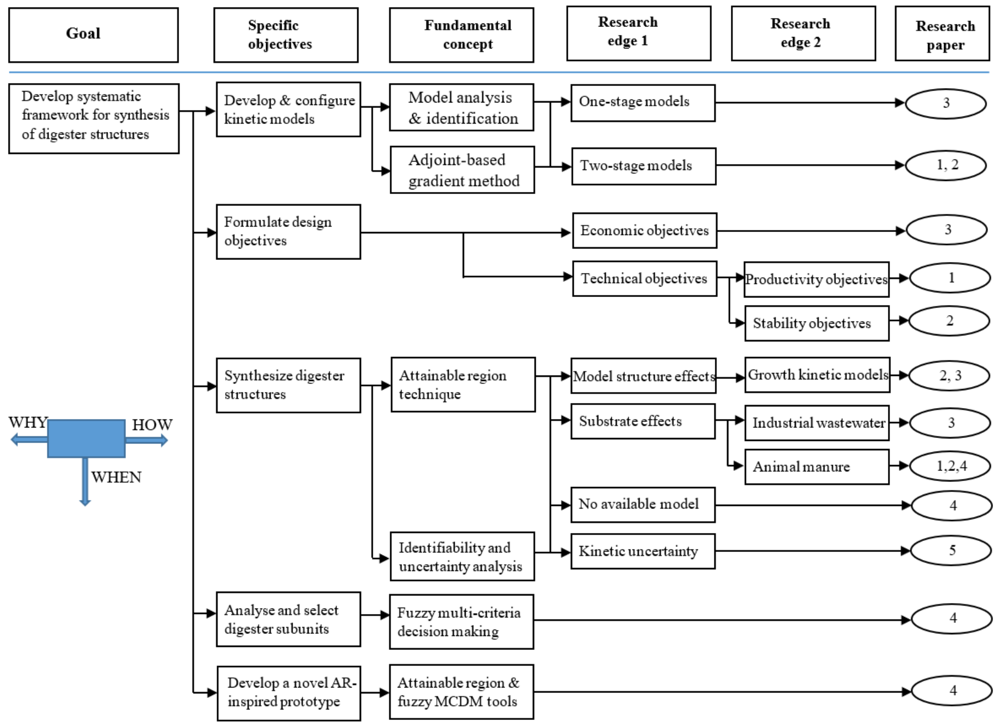

Figure 6: Conceptual framework and hierarchical breakdown of the study

When reading from left to right, the diagram answers the question "HOW", when reading from right to left, it answers the question "WHY" and when reading from top to bottom, it answers the question "WHEN". Overall, the diagram presents a conceptual framework, which summarizes how the research goal, the specific objectives, the model-based tools/methodologies are logically connected to one another.

Chapter 3: Summary and significance of findings

3.1 Framework for synthesis of anaerobic digester networks

There are several important areas where this study makes an original contribution to the field of anaerobic digestion and the findings from this study can be summarized in a decision support system (DSS) consisting of five main modules for synthesis of multistage anaerobic digesters, as shown in Figure 7.

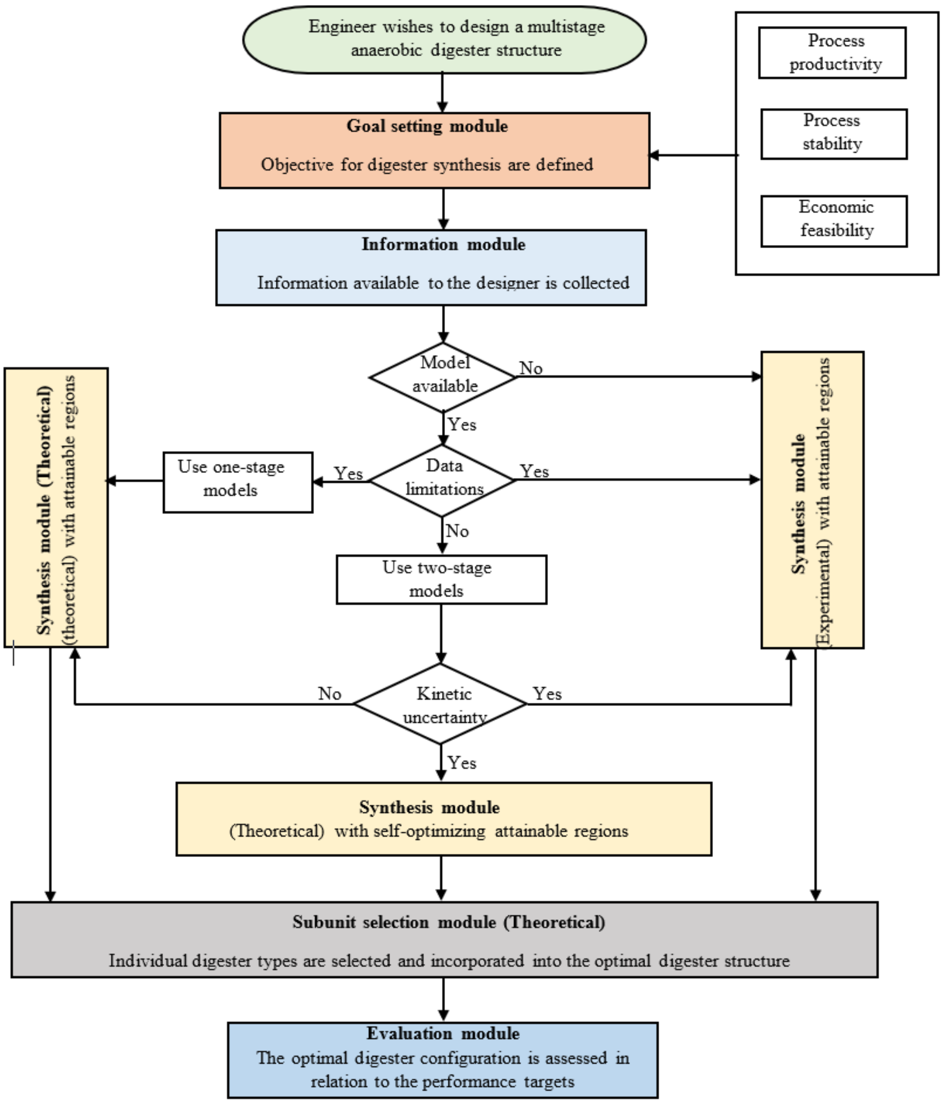

Figure 7: Decision support system for synthesis of anaerobic digester networks

3.1.1 Defining performance criteria for synthesis of digester networks

In the first module of Figure 7, goal setting module, a decision is made on performance objective or criteria to be used for synthesis of the anaerobic digester. Synthesis can be based on operational/ process objectives, whereby the study considered process stability **(Paper 2)** and process performance (measured in terms of biogas production and organic matter reduction) **(Paper 1).** Synthesis can also be based on economic objectives whereby the study developed digester economic evaluation models based on known economic feasibility indicators as well as macroeconomic parameters **(Paper 3).**

3.1.2 Information framework for synthesis of digester networks

In the second module of Figure 7, information available to the designer is collected and a decision is made on the synthesis approach based on the available information. Synthesis can be based on model requirements/characteristics whereby the study considered the following cases: no model availability **(Paper 4),** one-stage kinetic models **(Paper 3),** two-stage kinetic models **(Papers 1 & 2)**, kinetic uncertainty **(Paper 5)** as well as changes in kinetic model structure **(Papers 2 & 3)**.

3.1.3 Synthesis techniques of anaerobic digester networks

In the third module of Figure 7, synthesis module, the actual synthesis of the digester is performed at this stage. There are three synthesis modules, two theoretical and one experimental of which the choice of synthesis module depends on the information available to the designer. The experimental synthesis module is used when no model is available to describe the anaerobic treatment process, but data is available from experimental studies (**Paper 4**). The theoretical synthesis module based on attainable regions is used when there is data limitation (only data available is biogas yield measurements) and the designer is constraint to use simplified kinetic models **(Paper 3).** It is noteworthy that the first theoretical synthesis module can also be used when there is data availability, but the assumption is that the kinetic models and parameters are reliable (**Papers 1 & 2**). The second theoretical synthesis module (based on self-optimizing attainable regions) is used when data is available and the designer can use semi-simplified models, but there exists uncertainty in the kinetic coefficients (**Paper 5**). It is important for readers to note that even though kinetic data already contains information captured by kinetic models, the experimental synthesis module is limited to only graphical AR constructions, hence the number of design objectives or operational challenges that can be considered becomes limiting (especially for higher dimensional constructions).

3.1.4 Selection of digester subunits for synthesis of digester networks

After the synthesis module, the designer now has an optimal configuration of an anaerobic digester structure in terms of plug flow and continuous stirred tank anaerobic digesters. The next step is to decide on which plug flow digester to include in the network configuration considering there exist several types of plug flow digesters. This is done in the fourth module, the subunit selection module (**Paper 4**). After this stage, the designer now contains a complete network configuration, defined in terms of specific anaerobic digester system but before the system can be recommended for industrial use, it must be evaluated with respect to the performance criteria defined in the goal setting module.

3.2 Effects of feedstock and inoculum characteristics on digester network configuration

In addition, to the findings presented in the DSS, the study makes several noteworthy contributions of practical relevance to the scientific community. The study also analysed the effects of feedstock characteristics on the performance target and optimal configuration of anaerobic digester networks, considering three classes of organic substrates: industrial wastewater (**Paper 3**), animal manure (**Paper 1**) and inoculum from different sources (**Paper 2**). The results from both studies illustrated that a change in digested substrate and/or source of inoculum used to start-up the digester significantly influences the operating limits (defined by the attainable region), optimized parameter, as well as the design configuration of the optimal digester structure. This observed substrate effect on attainable regions shows great promises as it paves the way for other substrates such as blackwater, food waste, lignocellulosic waste, as well as co-digested feeds to be considered.

3.3 Brief discussion on the findings

Summarily, the contribution from all the five papers presented in the thesis can be stated as follows:

Paper 1: Presents a framework that uses two-stage kinetic models, productivity objectives for digester synthesis (mainly methane productivity and volatile solids reduction) and considers the effects of substrate characteristics, using five types of animal manure

Paper 2: Presents a framework that uses two-stage kinetic models, stability objectives for digester synthesis (considering inoculum to substrate ratio and instantaneous methanogenic yield) as well as effects of model structure and sources of inoculum used to start-up digester operation.

Paper 3: Presents a framework that uses one-stage kinetic models, economic design objectives (developing economic evaluation models based on known economic feasibility indicators as well as macroeconomic parameters) and industrial wastewater as feedstocks

Paper 4: Presents a framework that requires no kinetic model for digester synthesis, introduces fuzzy multicriteria decision making tools for selection of digester subunits, uses animal manure as substrate, and models a novel digester prototype for practical application

Paper 5: Presents a framework that simultaneously analyse model reliability, quantifies uncertainty in model states and construct attainable regions that are self-optimizing.

Considering all the five papers put together, the results are highly applicable (summarized in the DSS) for synthesis and optimization of multistage anaerobic digesters under different practical scenarios and varying degrees of information available to the designer.

It is important to bear in mind that since the CSTR model consist of a system of nonlinear algebraic equations, the existence of multiple steady states is often possible for a given feed point and residence time. This presents a considerable challenge when constructing and visualizing the AR as some aspects of the AR might change for different steady state values. In addition, the roots of the CSTR equation depend on the initial guess used to compute the numerical solution. In some cases, the roots give negative values and the author therefore limited the computation of CSTR roots to residence time values that give positive roots. It is therefore important to be cautious when interpreting the results presented in papers 1, 2, 4 & 5.

It is difficult to compare the results obtained in this study with those presented by other researchers as this study presents first of its kind, laying down such a comprehensive and systematic model-based framework for synthesis and optimization of anaerobic digester networks. So only a qualitative comparison with other model-based frameworks presented in the literature for bioprocess design could be conducted. Some of the published studies include: a model-based framework for optimization of lignocellulosic ethanol production under uncertainty (Morales-Rodriguez et al., 2012). The framework predefines candidate process configurations, identifies sources of process uncertainties, quantifies their impact on performance evaluation metrics of each process configuration, followed by selection of the best configuration. The candidates are predefined based on a set of subjective rules, which most often results in a local optimum in the final configuration obtained. The second example is a model-based framework for optimal synthesis of methane bioreactors using superstructure optimization (Pontes and Pinto, 2009). This involves construction of a very large reactor superstructure, which often suffers from multiple solutions. The last example includes the work of Alvarado-Morales *et al.* (Alvarado-Morales et al., 2010), who presented a model-based methodology for simultaneous design and control of a bioethanol production process. This framework involves the selection of reaction-process and separation-groups, which are then combined according to a set of connectivity rules and specifications to generate candidate flowsheet structures that are ranked through simulations. The connectivity rules still involve the problem of multiple solutions, as other connections can always be derived with similar or improved performance that those defined in the flowsheet candidates. As can be noticed, one common feature in the cited approaches is that a number of process configurations, referred to as flowsheet candidates are first defined before optimization/selection is performed to obtain the best configuration. However, an important question raised is that "What if better flowsheet configurations exists that are not predefined amongst the flowsheet candidates? On the other hand, the power of the framework presented in this study is that the limits of achievability for all possible reactor configurations (even in cases of kinetic uncertainties), even those that have not yet been devised, are obtained by incorporating attainable region analysis in the process. The approach presented in this study synthesizes a reactor configuration as part of the design process and connects the evaluation process to technical and economic objectives, which is the key interest of investors and engineers.

Chapter 4: Conclusion, recommendation and perspectives

4.1 Conclusion

Returning to the goal posed at the beginning of this study, it is now possible to state that the current study has successfully developed a set of integrated model-inspired frameworks for optimal synthesis of methane bioreactor structures, highly adaptable to different design objectives and degrees of data or model availability.

This study has laid down the theoretical framework and illustrated that it is possible to apply attainable region technique for synthesis and optimization of multistage anaerobic digesters under the following modeling scenarios for anaerobic digestion: one-stage kinetic models, two-stage kinetic models, and even in cases where there exists no kinetic model. This suggests that irrespective of the information available to the designer, the model-based framework can still be applied for synthesis of multistage anaerobic digesters. The findings illustrated that the structure of the kinetic model used to describe the growth of anaerobic microorganisms influences the performance targets and digester configurations obtained. This implies cautious selection of kinetic models for describing the anaerobic treatment process is invaluable for a reliable use of attainable regions for digester synthesis. The second major finding from the study is that synthesis of the anaerobic digesters can be tackled from both technical and economic perspectives making it highly attractive not only to engineers but also to investors. The study developed and illustrated the use objective functions that include economic feasibility indicators, operational indicators (such as volumetric methane production rate and volatile solids reduction) as well as stability indicators (inoculum to substrate ratio and instantaneous methanogenic yield) of methanogenic microorganisms.

Another interesting finding to emerge from the study is that a change in the type of digested substrate and/or source of inoculum used to start-up the digester significantly influences the operating limits (defined by the attainable region), optimized parameter, as well as the design configuration of the optimal digester structure. This finding paves the way for other substrates such as blackwater, food waste, lignocellulosic waste, as well as co-digested feeds to be considered. Owing to the wide variety of digester systems that exist, the study also introduced a framework that couples fuzzy multicriteria decision tools with attainable regions for simultaneous synthesis of digester structures and selection of digester subunits considering both techno-economic and environmental aspects. What this implies is that for the same digester structure, defined in terms of plug flow and continuous stirred tank reactors, the subunits (mainly type of plug flow digester) will be differ based on the practical considerations for operating the digester system. One of the more significant findings to emerge from this study is how the author has been able to propagate kinetic uncertainty on to the attainable regions using an integrated framework that couples practical identifiability and uncertainty quantification with attainable regions. Hence, when using attainable regions for performance targeting and digester network synthesis, the study indicates that it is possible to incorporate uncertainty of model prediction during construction of the attainable regions. The attainable region obtained in such cases is referred to as a self-optimizing attainable region, which is generally smaller than the attainable region but has an advantage of increased robustness.

Finally, the study has indicated that using digester networks as opposed to single digesters is able to bypass regions of lower reactivity and improve performance of the anaerobic treatment process. This is illustrated in the following specific predictions obtained from the results: (1) Methanogenic microorganisms have been shown to be more viable in digester structures as opposed to single digesters, (2) shorter payback periods (higher returns on investment) are achievable with digester structures, (3) volumetric methane productivity and improved effluent quality is observed with digester structures as opposed to single digesters. It is also worth mentioning that even though the study is based on the anaerobic treatment process, the framework for self-optimizing attainable regions can be applied to other chemical processes, where significant uncertainty exists.

4.2 Recommendation to industry

The findings, while preliminary, suggests that model-based techniques present an invaluable tool for design and operation of multistage anaerobic digesters, which if implemented by industry will significantly improve the efficiency of anaerobic digestion, both for small- and large-scale systems. The decision support system should be considered the first point of contact and used to compliment experiments during planning, design, scale-up and installation of anaerobic digestion plants involving multistage digesters. This will significantly reduce the number of expensive prototype systems and time-consuming studies usually required to obtain and optimal configuration of anaerobic digesters.

In addition, the novel AR-inspired prototype (developed in Paper 4), though still under experimental evaluation, will be highly recommended for use as mobile onsite biogas tank, for codigestion of blackwater and kitchen waste, where the gas produced will be used for cooking or heating. The AR-inspired prototype offers the following advantages: simple design (relative to other multistage systems), no moving parts, stability to organic shocks, low sludge generation (as much of the sludge is retained in the system), no requirement of biomass with special settling properties, no requirement of a special gas or sludge separation system, etc. More detailed performance results from field and experimental studies will be communicated in subsequent publications.

4.3 Perspectives for further research

The major contribution of the study has been the development and/or configuration of a simplified model (one- and two-stage kinetic models) and theoretical frameworks for use of attainable regions in digester network synthesis. AR is suitable for use not because of multiple reactors (or multistage digesters) but because of multiple reactions, such as the biological reactions in anaerobic digestion involving complex metabolic pathways. However, for practicality, the author has applied one- and two-stage lumped reaction models focusing on acid producing bacteria and methanogenic archaea to make the problem more tractable. It will be very interesting to consider a more complex (series and parallel) reaction set to align more closely to the biochemical pathways, i.e. a series of rate equations for hydrolysis, acidogenesis, acetogenesis and parallel reactions for acetoclastic and hydrogenotrophic methanogenesis in future work. The ultimate goal will be to consider the state-of-the-art anaerobic digestion model no. 1 (ADM1) for synthesis of anaerobic digesters using attainable regions.

More research is needed to expand the concept of self-optimizing attainable regions in the field of anaerobic digestion. This study has focused on kinetic uncertainty and it would be interesting to assess the effects of other potential sources of uncertainty (such as substrate characteristics, presence of inhibitions or temperature variations) on the performance targets of the anaerobic treatment process. More importantly, it would be very interesting to understand the geometric characteristics of the self-optimizing attainable regions with respect to the necessary conditions for attainable regions. Automated approaches for construction of self-optimizing attainable regions based on modifying the existing AR construction approaches should be researched.

Due to the author’s enthusiasm for digital sustainopreneurship, there is great interest for further progress in developing a comprehensive web-based expert system, Anaerobic Digestion Expert (including all the model-based techniques presented in the decision support system) to serve industry practitioners and researchers involved in design of anaerobic digesters. The system can also be expanded to include other bioenergy systems as this will be highly relevant for engineers, policy makers, and other stakeholders involved in sustainable waste management and renewable bioenergy systems.

References

ABUNDE NEBA, F., ASIEDU, N. Y., ADDO, A., MORKEN, J., ØSTERHUS, S. W. & SEIDU, R. 2019. Use of attainable regions for synthesis and optimization of multistage anaerobic digesters. *Applied Energy,* 242**,** 334-350.

AGARWAL, V., THOTLA, S., KAUR, R. & MAHAJANI, S. M. 2008a. Attainable regions of reactive distillation. Part II: Single reactant azeotropic systems. *Chemical Engineering Science,* 63**,** 2928-2945.

AGARWAL, V., THOTLA, S. & MAHAJANI, S. M. 2008b. Attainable regions of reactive distillation—Part I. Single reactant non-azeotropic systems. *Chemical Engineering Science,* 63**,** 2946-2965.

ALVARADO-MORALES, M., HAMID, M. K. A., SIN, G., GERNAEY, K. V., WOODLEY, J. M. & GANI, R. 2010. A model-based methodology for simultaneous design and control of a bioethanol production process. *Computers & Chemical Engineering,* 34**,** 2043-2061.

ANDERSSON, K., ROSEMARIN, A., LAMIZANA, B., KVARNSTRÖM, E., MCCONVILLE, J., SEIDU, R., DICKIN, S. & TRIMMER, C. 2016. *Sanitation, Wastewater Management and Sustainability: From Waste Disposal to Resource Recovery.*, UN Environment Programme and Stockholm Environment Institute.

ASIEDU, N., HILDEBRANDT, D. & GLASSER, D. 2015. Experimental Simulation of Three-Dimensional Attainable Region for the Synthesis of Exothermic Reversible Reaction: Ethyl Acetate Synthesis Case Study. *Industrial & Engineering Chemistry Research,* 54**,** 2619-2626.

BATSTONE, D. J. 2006. Mathematical Modelling of Anaerobic Reactors Treating Domestic Wastewater: Rational Criteria for Model Use. *Reviews in Environmental Science and Bio/Technology,* 5**,** 57-71.

BATSTONE, J. D., KELLER, J., ANGELIDAKI, I., KALYUZHNYI, S. V., PAVLOSTATHIS, S. G., ROZZI, A., SANDERS, W. T. M., SIEGRIST, H. & VAVILIN, V. A. 2002. The IWA anaerobic digestion model no 1 (ADM1). *Water Sci. Technol.,* 45**,** 65–73.

BENÍTEZ, M., BERMÚDEZ, A. & RODRÍGUEZ-CALO, J. F. 2017. Adjoint method for parameter identification problems in models of stirred tank chemical reactors. *Chemical Engineering Research and Design,* 123**,** 214-229.

BORISOV, M., DIMITROVA, N. & SIMEONOV, I. 2016. Mathematical Modelling of Anaerobic Digestion with Hydrogen and Methane Production**This research has been partially supported by the Bulgarian Science Fund under contract No. DFNI–E02/13. The work of the second author has been partially supported by Sofia University "St Kl. Ohridski" under contract No. 58/06.04.2016. *IFAC-PapersOnLine,* 49**,** 231-238.

BRUN, R., KÜHNI, M., SIEGRIST, H., GUJER, W. & REICHERT, P. 2002. Practical identifiability of ASM2d parameters—systematic selection and tuning of parameter subsets. *Water Research,* 36**,** 4113-4127.

BURRI, J. F., WILSON, S. D. & MANOUSIOUTHAKIS, V. I. 2002. Infinite DimEnsionAl State-space approach to reactor network synthesis: application to attainable region construction. *Computers & Chemical Engineering,* 26**,** 849-862.

CONNER, J. A. & MANOUSIOUTHAKIS, V. I. 2014. On the attainable region for process networks. *AIChE Journal,* 60**,** 193-212.

DAVIS, B. J., TAYLOR, L. A. & MANOUSIOUTHAKIS, V. I. 2008. Identification of the Attainable Region for Batch Reactor Networks. *Industrial & Engineering Chemistry Research,* 47**,** 3388-3400.

DONOSO-BRAVO, A., MAILIER, J., MARTIN, C., RODRÍGUEZ, J., ACEVES-LARA, C. A. & WOUWER, A. V. 2011. Model selection, identification and validation in anaerobic digestion: A review. *Water Research,* 45**,** 5347-5364.

EPA, U. S. E. P. A. 2006. Biosolids technology fact sheet, multi-stage anaerobic digestion. National Service Center or Environmental Publications (NSCEP).

ESPOSITO, G., FRUNZO, L., PANICO, A. & PIROZZI, F. 2011. Modelling the effect of the OLR and OFMSW particle size on the performances of an anaerobic co-digestion reactor. *Process Biochemistry,* 46**,** 557-565.

FAISAL, M. & UNNO, H. 2001. Kinetic analysis of palm oil mill wastewater treatment by a modified anaerobic baffled reactor. *Biochemical Engineering Journal,* 9**,** 25-31.

GLASSER, D., CROWE, C. & HILDEBRANDT, D. 1987. A geometric approach to steady flow reactors: the attainable region and optimization in concentration space. *Industrial & Engineering Chemistry Research,* 26**,** 1803-1810.

GLASSER, D., HILDEBRANDT, D., GODORR, S. & JOBSON, M. 1993. A Geometric Approach to Variational Optimization: Finding the Attainable Region. *IFAC Proceedings Volumes,* 26**,** 451-457.

HAUGEN, F., BAKKE, R. & LIE, B. 2013. Adapting Dynamic Mathematical Models to a Pilot Anaerobic Digestion Reactor. *Modeling, Identification and Control,* 34**,** 35-54.

HENZE, M., VAN LOOSDRECHT, M. C. M., EKAMA, G. A. & BRDJANOVIC, D. 2008. *Biological wastewater treatment*, IWA Publishing.

HILDEBRANDT, D. & GLASSER, D. 1990. The attainable region and optimal reactor structures. *Chemical Engineering Science,* 45**,** 2161-2168.

HILDEBRANDT, D., GLASSER, D. & CROWE, C. M. 1990. Geometry of the attainable region generated by reaction and mixing: with and without constraints. *Industrial & Engineering Chemistry Research,* 29**,** 49-58.

HORN, F. Attainable and non-attainable regions in chemical reactor technique. . Third European Symposium on Chemical Reaction Engineering, 1964. London: Pergamon Press, 123–138.

IBÁÑEZ-FORÉS, V., BOVEA, M. D. & PÉREZ-BELIS, V. 2014. A holistic review of applied methodologies for assessing and selecting the optimal technological alternative from a sustainability perspective. *Journal of Cleaner Production,* 70**,** 259-281.

IEA, IRENA, UNSD, WB & WHO 2019. Tracking SDG 7: The Energy Progress Report 2019. Washington DC.

JINGURA, R. M. & KAMUSOKO, R. 2017. Methods for determination of biomethane potential of feedstocks: a review %J Biofuel Research Journal. 4**,** 573-586.

JØRGENSEN, P., J 2009. *Biogas – green energy,* Faculty of Agricultural Sciences, Aarhus University.

KAYA, İ., ÇOLAK, M. & TERZI, F. 2019. A comprehensive review of fuzzy multi criteria decision making methodologies for energy policy making. *Energy Strategy Reviews,* 24**,** 207-228.

KUMAR, A., SAH, B., SINGH, A. R., DENG, Y., HE, X., KUMAR, P. & BANSAL, R. C. 2017. A review of multi criteria decision making (MCDM) towards sustainable renewable energy development. *Renewable and Sustainable Energy Reviews,* 69**,** 596-609.

KYTHREOTOU, N., FLORIDES, G. & TASSOU, S. A. 2014. A review of simple to scientific models for anaerobic digestion. *Renewable Energy,* 71**,** 701-714.

LAKSHMANAN, A. & BIEGLER, L. T. 1996. Synthesis of Optimal Chemical Reactor Networks. *Industrial & Engineering Chemistry Research,* 35**,** 1344-1353.

LAWRENCE, A. W. 1971. Application of Process Kinetics to Design of Anaerobic Processes. *Anaerobic Biological Treatment Processes.* AMERICAN CHEMICAL SOCIETY.

LINKE, B. 2006. Kinetic study of thermophilic anaerobic digestion of solid wastes from potato processing. *Biomass and Bioenergy,* 30**,** 892-896.

LOHANI, S. P., WANG, S., BERGLAND, W. H., KHANAL, S. N. & BAKKE, R. 2018. Modeling temperature effects in anaerobic digestion of domestic wastewater. *Water-Energy Nexus,* 1**,** 56-60.

LÓPEZ, I. & BORZACCONI, L. 2009. Modelling a full scale UASB reactor using a COD global balance approach and state observers. *Chemical Engineering Journal,* 146**,** 1-5.

MANOUSIOUTHAKIS, V. I., JUSTANIEAH, A. M. & TAYLOR, L. A. 2004. The Shrink–Wrap algorithm for the construction of the attainable region: an application of the IDEAS framework. *Computers & Chemical Engineering,* 28**,** 1563-1575.

MAO, C., FENG, Y., WANG, X. & REN, G. 2015. Review on research achievements of biogas from anaerobic digestion. *Renewable and Sustainable Energy Reviews,* 45**,** 540-555.

MARTINEZ, E., MARCOS, A., AL-KASSIR, A., JARAMILLO, M. A. & MOHAMAD, A. A. 2012. Mathematical model of a laboratory-scale plant for slaughterhouse effluents biodigestion for biogas production. *Applied Energy,* 95**,** 210-219.

MCGREGOR, C., HILDEBRANDT, D. & GLASSER, D. 1998. Process Synthesis for a Reactor-Separator-Recycle System using the Attainable Region Approach. *Developments in Chemical Engineering and Mineral Processing,* 6**,** 21-39.

MES, T. Z. D. D., STAMS, A. J. M., REITH, J. H. & ZEEMAN, G. 2003. Methane production by anaerobic digestion of wastewater and solid wastes. *Bio-methane and bio-hydrogen : status and perspectives of biological methane and hydrogen production.* The Hague, The Netherlands: Dutch Biological Hydrogen Foundation - NOVEM.

METZGER, M. J., GLASSER, D., HAUSBERGER, B., HILDEBRANDT, D. & GLASSER, B. J. 2009. Use of the attainable region analysis to optimize particle breakage in a ball mill. *Chemical Engineering Science,* 64**,** 3766-3777.

MING, D., GLASSER, D. & HILDEBRANDT, D. 2013. Application of attainable region theory to batch reactors. *Chemical Engineering Science,* 99**,** 203-214.

MING, D., GLASSER, D., HILDEBRANDT, D., GLASSER, B. & METZER, M. 2016. *Attainable Region Theory: An Introduction to Choosing an Optimal Reactor,* Hoboken, New Jersey, John Wiley & Sons, Inc.

MORALES-RODRIGUEZ, R., MEYER, A. S., GERNAEY, K. V. & SIN, G. 2012. A framework for model-based optimization of bioprocesses under uncertainty: Lignocellulosic ethanol production case. *Computers & Chemical Engineering,* 42**,** 115-129.

MUVHIIWA, R. F., HILDEBRANDT, D., GLASSER, D., MATAMBO, T. & SHERIDAN, C. 2015. A thermodynamic approach toward defining the limits of biogas production. *AIChE Journal,* 61**,** 4270-4276.

PONTES, R. F. F. & PINTO, J. M. 2009. Optimal synthesis of anaerobic digester networks. *Chemical Engineering Journal,* 149**,** 389-405.

POSADA, A. & MANOUSIOUTHAKIS, V. 2008. Multi-feed attainable region construction using the Shrink–Wrap algorithm. *Chemical Engineering Science,* 63**,** 5571-5592.

SCOTT, F., CONEJEROS, R. & AROCA, G. 2013. Attainable region analysis for continuous production of second generation bioethanol. *Biotechnology for Biofuels,* 6**,** 171.

SEODIGENG, T., HAUSBERGER, B., HILDEBRANDT, D. & GLASSER, D. 2009. Recursive constant control policy algorithm for attainable regions analysis. *Computers & Chemical Engineering,* 33**,** 309-320.

SIN, G., GERNAEY, K. V. & LANTZ, A. E. 2009. Good modeling practice for PAT applications: Propagation of input uncertainty and sensitivity analysis. 25**,** 1043-1053.

SIN, G. & VANROLLEGHEM, P. A. 2007. Extensions to modeling aerobic carbon degradation using combined respirometric–titrimetric measurements in view of activated sludge model calibration. *Water Research,* 41**,** 3345-3358.

SMITH, R. L. & MALONE, M. F. 1997. Attainable Regions for Polymerization Reaction Systems. *Industrial & Engineering Chemistry Research,* 36**,** 1076-1084.

SUNG, C. & MARAVELIAS, C. T. 2006. An attainable region approach for effective production planning. *In:* MARQUARDT, W. & PANTELIDES, C. (eds.) *Computer Aided Chemical Engineering.* Elsevier.

SUNG, C. & MARAVELIAS, C. T. 2007. An attainable region approach for production planning of multiproduct processes. *AIChE Journal,* 53**,** 1298-1315.

WANG, L. K., SHAMMAS, N. K. & HUNG, Y.-T. 2007. *Biosolids Treatment Processes,* New Jersey, Humana Press Inc.

WARE, A. & POWER, N. 2017. Modelling methane production kinetics of complex poultry slaughterhouse wastes using sigmoidal growth functions. *Renewable Energy,* 104**,** 50-59.

YANG, H., DENG, L., LIU, G., YANG, D., LIU, Y. & CHEN, Z. 2016. A model for methane production in anaerobic digestion of swine wastewater. *Water Research,* 102**,** 464-474.

YU, L., WENSEL, P. C., MA, J. & CHEN, S. 2013. Mathematical Modeling in Anaerobic Digestion (AD). *Bioremediation & Biodegradation***,** 2155-6199.

ZAHER, U., PANDEY, P. & CHEN, S. 2009. A simple elemental continuity based model application to study the anaerobic microbial activity for the treatment of dairy manure. *Applied Mathematical Modelling,* 33**,** 3553-3564.

ZHANG, J., LOH, K.-C., LI, W., LIM, J. W., DAI, Y. & TONG, Y. W. 2017. Three-stage anaerobic digester for food waste. *Applied Energy,* 194**,** 287-295.

ZHOU, W. & MANOUSIOUTHAKIS, V. I. 2006. Non-ideal reactor network synthesis through IDEAS: Attainable region construction. *Chemical Engineering Science,* 61**,** 6936-6945.

ZHOU, W. & MANOUSIOUTHAKIS, V. I. 2007a. Pollution prevention through reactor network synthesis: the IDEAS approach. *International Journal of Environment and Pollution,* 29**,** 206-231.

ZHOU, W. & MANOUSIOUTHAKIS, V. I. 2007b. Variable density fluid reactor network synthesis—Construction of the attainable region through the IDEAS approach. *Chemical Engineering Journal,* 129**,** 91-103.

ZHOU, W. & MANOUSIOUTHAKIS, V. I. 2008. On dimensionality of attainable region construction for isothermal reactor networks. *Computers & Chemical Engineering,* 32**,** 439-450.

ZHOU, W. & MANOUSIOUTHAKIS, V. I. 2009. Automating the AR construction for non-isothermal reactor networks. *Computers & Chemical Engineering,* 33**,** 176-180.

ZINATIZADEH, A. A. L., MOHAMED, A. R., NAJAFPOUR, G. D., HASNAIN ISA, M. & NASROLLAHZADEH, H. 2006. Kinetic evaluation of palm oil mill effluent digestion in a high rate up-flow anaerobic sludge fixed film bioreactor. *Process Biochemistry,* 41**,** 1038-1046.

Appended Papers

Paper 1:

Use of attainable regions for synthesis and optimization of multistage anaerobic digesters

A paper published in ***Applied Energy***

Citation:

ABUNDE NEBA, F., ASIEDU, N. Y., ADDO, A., MORKEN, J., ØSTERHUS, S. W. & SEIDU, R. 2019d. Use of attainable regions for synthesis and optimization of multistage anaerobic digesters. ***Applied Energy***, 242, 334-350.

Highlights:

- A theoretical framework for use of attainable regions to model performance targets and anaerobic digester networks is presented
- A physical and geometric classification of methane bioreactor types considering both low- and high-rate digesters is proposed
- Two-stage kinetic models are used to interpret anaerobic digestion process as geometric objects
- Volumetric methane productivity and percentage volatile solids reduction are used as design objectives
- Five substrates: pig manure, goat manure, swine manure, horse manure and chicken manure) are considered as feedstocks
- Attainable regions and optimized parameters differ for each digested substrate
- Digester networks as opposed to single digesters improves anaerobic treatment

Applied Energy 242 (2019) 334–350

Contents lists available at ScienceDirect

Applied Energy

journal homepage: www.elsevier.com/locate/apenergy

Use of attainable regions for synthesis and optimization of multistage anaerobic digesters

F. Abunde Neba[a,d,e,*], Nana Y. Asiedu[a,c], Ahmad Addo[a,b], John Morken[f], Stein W. Østerhus[d], Razak Seidu[e]

[a] *Abunde Sustainable Engineering Group (AbundeSEG), Ghana*
[b] *Department of Agricultural and Biosystems Engineering, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana*
[c] *Department of Chemical Engineering, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana*
[d] *Department of Civil and Environmental Engineering, Norwegian University of Science and Technology, Trondheim, Norway*
[e] *Institute for Marine Operations and Civil Engineering, Norwegian University of Science and Technology, Ålesund, Norway*
[f] *Faculty of Science and Technology, Drobakveien 31, 1432 Aas, Norwegian University of Life Sciences, Ås, Norway*

HIGHLIGHTS

- Digester networks as opposed to single digesters improves anaerobic treatment.
- We introduced the use of attainable regions to model anaerobic digester networks.
- A physical and geometric classification of methane bioreactor types are presented.
- Technique uses process kinetics to define performance targets and digester networks.
- Attainable regions and optimized parameters differ for each digested substrate.

ARTICLE INFO

Keywords:
Attainable regions
Biodigester networks
Multistage anaerobic digestion
Biogas production

ABSTRACT

Anaerobic digestion involves multiple reactions, and when operated as a single stage, the process conditions are only suitable for all the reactions with no particular reaction being optimized, hence limiting overall performance. Multistage anaerobic digestion, in which multiple digesters are operated in a network are designed to optimize each process reaction, but very few writers have drawn on any systematic procedure for the design of digester networks. This study is about multistage digester networks, but contrary to traditional multistage digestion articles that focus on the experimental evaluation of a predefined network configuration, this study develops a systematic methodological framework based on the concept of attainable regions for optimal synthesis of digester networks. Within the framework, a simplified model is developed, which accounts for the geometric characteristics of fundamental anaerobic digester types. The model is validated with experimental data of diary, horse, goat, chicken and swine manure, and shows good agreement (model errors between 0.01 and 0.06). The attainable regions and their optimized parameters differ for each digested substrate, and the optimal networks are made of different combinations of digesters operated in a continuous (axial mixing) and/or plug flow (no axial mixing) mode. This substrate effect on attainable regions shows great promises as it paves the way for other substrates such as food waste, lignocellulosic waste, co-digested feeds, etc. This study though preliminary presents a breakthrough in extending the use of digester networks to solve more operational challenges as well as support retrofitting multi-stage systems into facilities where single-stage digesters already exist.

1. Introduction

In the new global economy, bioenergy conversion processes have become a central component in sustainable development due to their ability to minimize depletion of natural energy resources as well as climate and environmental deterioration. Amongst the existing bioenergy conversion processes, the anaerobic treatment process has become very popular due to its ability to simultaneously stabilize waste,

* Corresponding author at: Department of Civil and Environmental Engineering, Norwegian University of Science and Technology, Trondheim, Norway.
E-mail address: fabricen@stud.ntnu.no (F. Abunde Neba).

https://doi.org/10.1016/j.apenergy.2019.03.095
Received 15 November 2018; Received in revised form 21 February 2019; Accepted 8 March 2019
0306-2619/

Nomenclature

$(VS)_0$	initial concentration of volatile solids (gVS/L)
A_f	acidity factor (gVFA/L)/(gBVS/L)
B_0	biodegradability constant (gBVS/L)/(gVS/L)
K_{ime}	VFA inhibition constant for methanogenic archae (gVFA/L)
$K_{s_{ac}}$	monod half-saturation constant for acidogenic bacteria (gBVS/L)
$K_{s_{me}}$	monod half-saturation constant for acidogenic bacteria (gVFA/L)
S_{BVS_0}	initial concentration of biodegradable volatile solids (gBVS/L)
S_{BVS}	concentration of biodegradable volatile solids (gBVS/L)
S_{VFA_0}	initial concentration of volatile fatty acids (gVFA/L)
S_{VFA}	concentration of volatile fatty acids in bioreactor (gVFA/L)
$T_{\max}$	maximum temperature at which growth rate is zero (°C)
$T_{\min}$	minimum temperature at which growth rate is zero (°C)
X_0	initial concentration of biomass in reactor (g/L)
X_{ac_0}	initial concentration of acidogenic bacteria (gac. /L)
X_{ac}	concentration of acidogenic bacteria in bioreactor (gac. /L)
X_{me_0}	initial concentration of methanogenic archae (gme. /L)
X_{me}	concentration of methanogenic archae in bioreactor (gme. /L)
k_1	yield constant (gBVS/gac. /L)
k_2	yield constant (gVFA/gac. /L)
k_3	yield constant (gVFA/gme. /L)
$r_{S_{BVS}}$	reaction rate for biodegradable volatile solids (gBVS/L/d)
$r_{S_{VFA}}$	reaction rate for volatile fatty acids (gVFA/L/d)
$r_{X_{ac}}$	reaction rate for acidogenic bacteria (gac. /L/d)
$r_{X_{me}}$	reaction rate for methanogenic archae (gme. /L/d)
$t_{\upsilon,\alpha/2}$	student t-distribution parameter
$\hat{\beta}$	vector of estimated model parameters
γ_{CH_4}	volumetric methane productivity ($LCH_4/m^3/d$)
γ_s	methane yield
$\mu_{m_{ac}}$	maximum specific growth rate of acidogenic bacteria (d^{-1})
$\mu_{m_{me}}$	maximum specific growth rate of methanogenic archae (d^{-1})
μ_{ac}	specific growth rate of methanogenic archae (d^{-1})
μ_{me}	specific growth rate of methanogenic archae (d^{-1})
σ^2	standard error
B	Ratkowsky parameter ($°C^{-1}$ $h^{-0.5}$)
C	Ratkowsky parameter C ($°C^{-1}$)
T	reactor temperature (°C)
EMY_{90}	90% experimental methane yield ($mLCH_4/gVS$)
HRT	hydraulic retention time (d)
J	Jacobian matrix evaluated at parameter estimates
VSR	volatile solids reduction (%)
n	number of experimental data points
p	number of model parameters
α	significance level
β	vector of real model parameters
ϑ	acidogenic fraction

generate bioenergy and recycle valuable nutrients [1]. The anaerobic digestion process is highly complex, and the performance of the digester can be affected by a myriad of factors including organic loading rates, presence of inhibitory or toxic substances, reactor configuration, hydraulic retention time and environmental factors such as temperature [2]. For this reason, careful design of methane bioreactors is central to the optimal operation of anaerobic treatment process, as it is required to provide an appropriate environment for the complex interaction of anaerobic microorganisms to grow and produce biogas [3].

Studies have shown that when the reaction mechanism of a process is complex, the best performance is often achieved in a reactor network or reactor structure [4]. However, current configurations of methane bioreactors are simpler, employing one or rarely two different digesters in the so-called "rational basis of design," i.e., determination of digester capacity based on volatile solids (VS) loading, temperature, the extent of mixing, and so on [2]. It is well known that each digester has different characteristics often making them more adequate to treat waste of specific characteristics [5], and thus utilizing one reactor in one configuration may limit the possible combination of pathways, which may limit performance [6]. This is because anaerobic digestion involves multiple reactions and when operated as a single stage, the process conditions are only suitable for all the reactions with no particular reaction being optimized [7].

Multistage anaerobic digestion, in which multiple digesters are operated in a network are designed to optimize each process reaction for the breakdown of organics and generation of methane-rich biogas [7]. The most common techniques used for staging digester networks include [8]: Mesophilic Digester Staging in which two heated well-mixed digesters are operated in series, Acid/Gas (AG) Phased Staging in which acid-forming and methane-forming stages are physically separated, Temperature Phased Staging, which incorporates both thermophilic and mesophilic conditions in a series operation and Thermophilic Staging in which one or smaller digesters follow a large digester to prevent pathogen short-circuiting. Several studies focusing on anaerobic digester networks have been published using either two, three or four individual digesters operating in a particular configuration. Zhang et al. [9] presented a novel compact three-stage anaerobic digester (TSAD) for methane production from food waste. The functionalized staging using the Acid/Gas (AG) Phased technique significantly resulted in a 24–54% increase in methane production. Akobi et al. [10] investigated the effect of staging on the anaerobic digestibility of hydrolysates obtained from pretreated poplar wood biomass. The authors reported that the two-stage process resulted in a 16% increase in COD removal efficiency compared to the single-stage process. Furthermore, Nasr et al. [11] achieved an increase of 18.5% in the total energy yield by using a two-stage digester as opposed to a single-stage digester for digestion of thin stillage. While a handful of studies have demonstrated the ability of digester networks to enhance process performance, there still exists a high degree of empiricism in the design of digester networks. The aforementioned and all existing studies often predefine the network configuration, mostly assuming series digester connection, with no systematic approach to answer the following three questions: (1) How many individual digesters should be included in an optimal network (2) what type of digesters should be considered (PFR, CSTR, UASB, etc.) (3) Do we include recycle and bypass streams? If so, where are they placed within the structure? Very few writers have drawn on any systematic procedure for the design of anaerobic digester networks, and systematic procedures based on optimization techniques can further increase the ability of multistage digesters to improve process stability and operation or improve process economics. Also, using empirical methods to optimize the design of anaerobic digesters often requires construction of expensive prototype systems and time-consuming studies, which has been a key motivation for reliance on model-based techniques [12].

A previous article, which attempted to address such digester network synthesis problem involved creating a very large, generalized, digester superstructure [13]. However, a major challenge with this approach is that of multiple solutions or the existence of local optima, which illustrated the following two questions [4]: (1) Are there similar superstructure configurations that achieve the same result? And (2)

Does a better superstructure exist? Hence a more reliable and robust technique for the synthesis and operation of methane bioreactor networks will be a major breakthrough in extending the use of digester networks to solve more operational challenges in anaerobic digestion. This study is about multistage anaerobic digesters, but contrary to traditional multistage digestion articles that describe the experimental evaluation of a predefined network configuration, this study presents a systematic methodological framework developed for the design of multistage digester networks. The framework is based on the concept of attainable regions, which represents a collection of all possible outputs for all possible reactor designs by interpreting chemical processes as geometric objects that define a region of achievability without having to explicitly enumerate all possible design combinations [4]. The main advantage of this approach over the use of superstructure optimization is that it enables knowledge of all possible states for all possible digester configurations (even those that have not yet been devised) to be first obtained, before looking for configurations to achieve the maximum attainable states. The application of this concept to synthesize anaerobic digesters has not been recorded so far, which is why the current paper aims to develop a theoretical framework to support the application of attainable regions to model anaerobic digester networks. As required by the AR technique, the major contribution of this work is the development of a simplified model of the anaerobic treatment process, which has been used to account for the mathematical and geometric characteristics of fundamental anaerobic reactor types. This is followed by model identification with test experimental data sets, model dimensionality reduction, and construction of attainable regions. Further to a proof-of-concept for the geometric optimization technique, two optimization problems are formulated and solved geometrically using attainable region, to provide methane bioreactor structures that maximize volumetric methane production rate and volatile solids reduction.

2. Theoretical developments

2.1. Anaerobic digestion and reactor network synthesis problem

The anaerobic digestion process occurring in methane bioreactors is a multi-step process involving series and parallel reactions, which are either biochemical or physicochemical in nature (Fig. 1).

As shown in Fig. 1, compounds can traverse along many different paths, which makes it difficult to predict the flow of material in the anaerobic digestion chain, or to know what conditions favour a particular pathway for the production of a desired intermediate product or a final product in the chain. For this reason, a network of methane bioreactor becomes interesting because of complexities in the metabolic pathways, since a reactor network often gives the best performance when the reaction mechanism is complex [4].

2.2. Modeling the anaerobic treatment process

Dynamic models that describe the transient behavior of anaerobic digestion process occurring in methane bioreactors are based on systems of ordinary differential equations, which represent material balances for the various components in the metabolic pathway. For using the model in attainable region synthesis of the reactor, we set four main requirements an ideal model should attain:

- Present a compromise between being highly accurate but very complex input requirement and highly simplified but very limited predictive ability.
- Represent the effect of temperature on the anaerobic treatment process since the system is highly sensitive to operating temperature.
- Consider the effect of waste characteristics as different types of organic waste are normally used in anaerobic systems.
- Predict process optima and instability due to reactor overload or presence of toxic components in waste stream.

In this study, the objective is to maximize methane productivity, which is the final product in the chain and the scheme presented in Fig. 1 is thus simplified to focus on methane production. In our next paper, we will focus on maximizing an intermediate product, hydrogen, and the model would be extended to include the effect of hydrogen, constituting a multidimensional case of attainable region compared to the two-dimensional case presented in this study.

In order to meet the model requirements for maximizing methane production through the use of attainable regions, the anaerobic digestion process is simplified into two main biological processes (Fig. 2); acid formation stage for waste conversion and methane production for waste stabilization [2]. This considers four main state variables, for both groups of bacteria as well as their substrates, which include, acid-forming bacteria, methane-forming bacteria, biodegradable organic substrate, and organic acids. Bastone, [14] also confirmed that for designing of anaerobic processes, simplified models of at least two stage are more appropriate since the focus is on hydrodynamics and behaviour of solids.

(a) Model state equations

From the scheme shown in Fig. 2, we derive the rate expressions of the four states, biodegradable volatile solids (BVS); volatile fatty acids (VFA) as acetate; acidogenic bacteria; methanogenic archae, as expressed by Eqs. (1)–(4) respectively.

$$\frac{dS_{BVS}}{dt} = r_{BVS} = -k_1\mu_{ac}X_{ac} \tag{1}$$

$$\frac{dS_{VFA}}{dt} = r_{VFA} = k_2\mu_{ac}X_{ac} - k_3\mu_{me}X_{me} \tag{2}$$

$$\frac{dX_{ac}}{dt} = r_{ac} = \mu_{ac}X_{ac} \tag{3}$$

$$\frac{dX_{me}}{dt} = r_{me} = \mu_{me}X_{me} \tag{4}$$

The model assumes the specific death rate of both microbial populations is negligible compared to the specific growth rate. The specific growth rate of acidogenic bacteria is modeled using the Monod

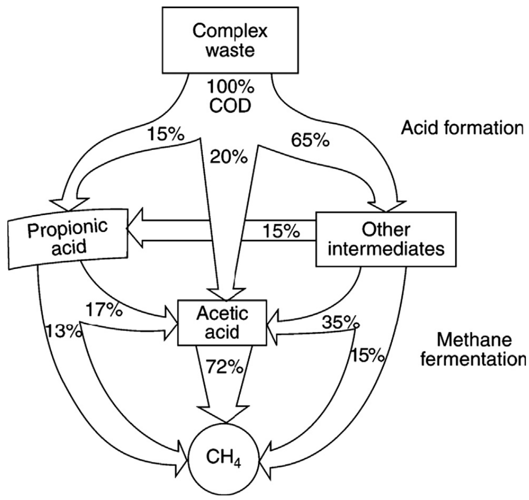

Fig. 1. Biochemical pathways for volatile solids reduction and methane generation [2].

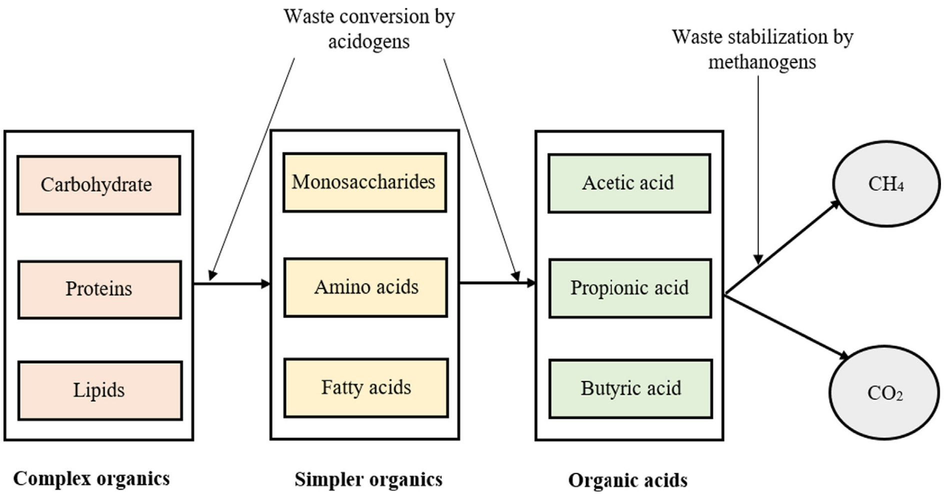

Fig. 2. Simplified two-stage scheme for anaerobic treatment process.

equation, Eq. (5) while an uncompetitive inhibition term is added to that of methanogenic archae, Eq. (6) to account for volatile acid inhibition during reactor upset or failure.

$$\mu_{ac} = \mu_{m_{ac}} \frac{S_{BVS}}{K_{s_{ac}} + S_{BVS}} \tag{5}$$

$$\mu_{me} = \mu_{m_{me}} \frac{S_{VFA}}{K_{s_{me}} + S_{VFA}\left(1 + \frac{S_{VFA}}{K_{i_{me}}}\right)} \tag{6}$$

(b) Model inputs

The model is made to have four inputs; temperature, volatile solids loading, digestion time and type of organic waste to be digested. The waste type is characterized by two parameters, the biodegradability constant (B_0) and the acidity constant (A_f), which are unique to each type of waste [12]. The two constants are modelled by Eqs. (7) and (8) respectively.

$$S_{BVS_0} = B_0 (VS)_0 \tag{7}$$

$$S_{VFA_0} = A_f S_{BVS_0} \tag{8}$$

The initial concentrations of acidogenic and methanogenic archae contained in the inoculum are expressed as a function of acidogenic fraction (ϑ) of the inoculum as shown by Eqs (9) and (10). Knowing the initial concentration of biomass, X_{in} the acidogenic fraction is estimated using test data.

$$X_{ac_0} = \vartheta X_0 \tag{9}$$

$$X_{me_0} = (1 - \vartheta) X_0 \tag{10}$$

The maximum growth rates of acid-forming ($\mu_{m_{ac}}$) and methane forming bacteria ($\mu_{m_{me}}$) are functions of the digestion temperature and this dependence was modeled using the Ratkowsky expanded square root model, Eq. (11) [15], which describes the effect of temperature over the entire temperature range of the anaerobic digestion process.

$$\mu_{m_{ac}}(T) = \mu_{m_{me}}(T) = [B(T - T_{\min})]^2\{1 - exp[C(T - T_{\max})]\}^2 \tag{11}$$

$$T_{min} < T < T_{max}$$

T_{min} and T_{max} are respectively the maximum and minimum temperatures at which the growth rate is zero while the constants B(°C^{-1}h$^{-0.5}$) and C(°C^{-1}) are known as Ratkowsky parameters, which are normally estimated from test data to reflect the process being modelled.

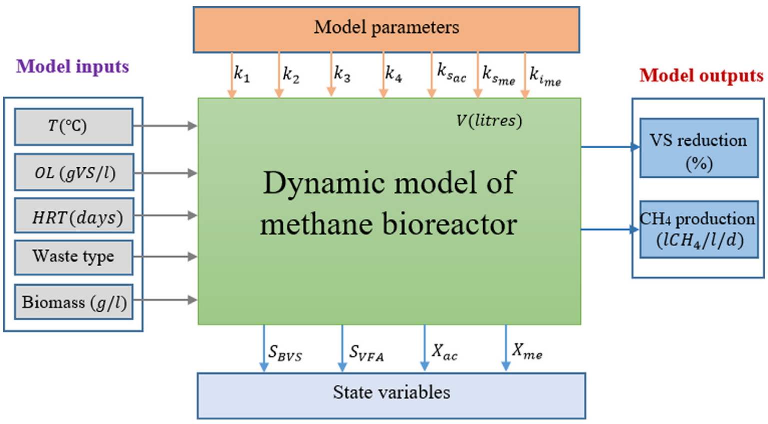

Fig. 3. Model of methane bioreactor showing inputs, outputs, parameters and state variables.

(c) Model outputs

The model inputs are propagated to model outputs (methane productivity and volatile solids reduction) through the state variables. The volumetric methane productivity and percentage volatile solids reduction are modelled using Eqs. (12) and (13) respectively [16].

$$\gamma_{CH_4} = \mu_{me} X_{me} (k_3 - 1) \times 1000 \gamma_s \tag{12}$$

$$VSR = HRT \frac{k_3 \mu_{me} X_{me}}{S_{VS_{in}}} \times 100 \tag{13}$$

Fig. 3 presents a summary of the model scheme, clearly outlining the model inputs, model outputs, kinetic constants as well as state variables. The result is a much-simplified model of anaerobic process, which meets all the requirements set above. (See section 2).

The effect of factors such as alkalinity, concentration of cation, dissolved CO_2 and ammonia gas is not considered because their effect is already lumped into B_0 and A_f. The model parameters now only depend on the bacterial consortium present in the methane bioreactor.

2.3. Hydrodynamic configurations of methane bioreactors

Over the past years, a variety of different methane bioreactors have been designed and are currently in use at industrial and domestic levels. By using the geometric approach of attainable regions to optimize the process operation, we provide a general classification of the existing reactor configurations. Methane bioreactors can be designed using a number of different hydrodynamic configurations, mainly derived from a combination of three fundamental regimes: flow regime, mixing regime and reactor regime, as shown in Fig. 4. Under flow regime, methane bioreactors can be operated in a batch, fed-batch or continuous mode; under mixing regime, they can be operated as completely mixed or with no axial mixing and under reactor regime, they are classified as conventional or modified. A continuous flow regime operated with no axial mixing gives a plug flow operation and when operated as completely mixed gives a continuous stirred tank operation. When including the reactor regime the flow and mixing regimes for a conventional reactor ends at plug flow reactor and continuously stirred tank reactor. Finally, for modified plug flow reactor regime, we have a variety of methane bioreactors, which include anaerobic filter (AF), upflow anaerobic sludge blanket (UASB), anaerobic baffled reactor (ABR) and Expanded granular sludge blanket (EGSB). A modified continuous stirred tank reactor gives an anaerobic contact reactor (ACR).

The attainable region for anaerobic treatment process defines all possible states that can be achieved by a given organic load and reaction kinetics, using a combination of two fundamental processes only; reaction and mixing [17]. The plug flow reactor represents an extreme case of reaction while the continuous stirred tank reactor represents an extreme case of mixing. As such, methane bioreactors with plug flow operation can be considered provide reaction while those continuous stirred operations are considered to provide mixing as illustrated in Fig. 5.

Since there are several reactors that can be considered to provide reaction, and/or mixing, the choice of which reactor to use depends on other operational constraints of anaerobic treatment process such as the strength of the waste, organic load, type of substrate, etc. [18]. Table 1 presents an overview of other parameters considered for the selection of an appropriate methane bioreactor.

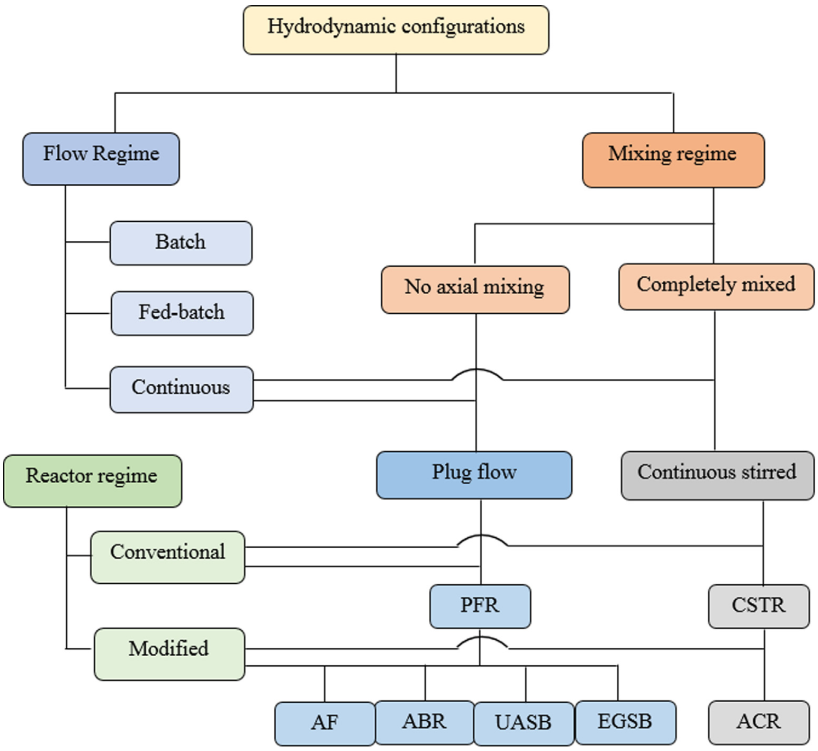

Fig. 4. Classification of hydrodynamic configurations of methane bioreactors.

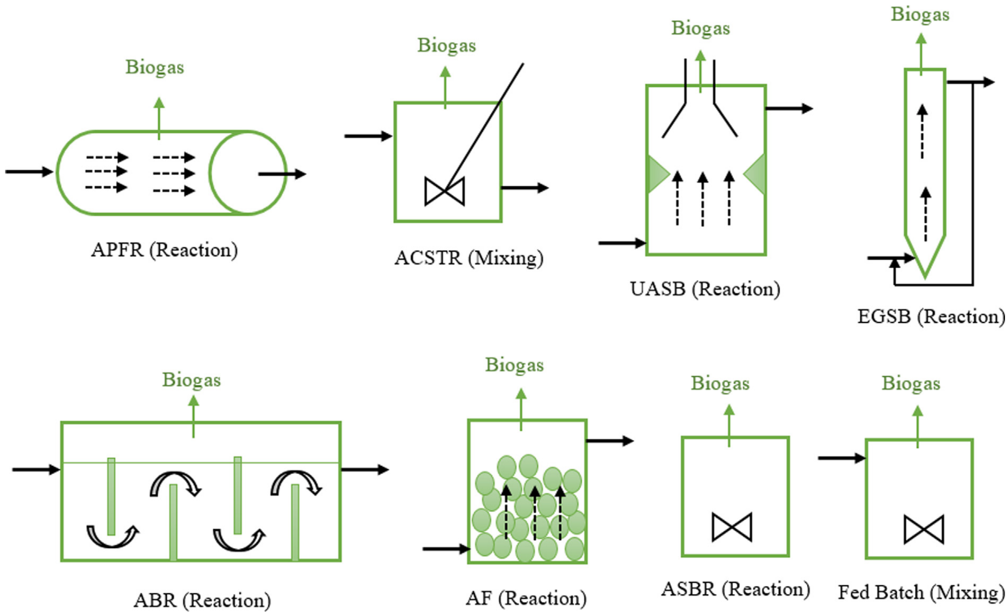

Fig. 5. Grouping of methane bioreactors types into fundamental processes of mixing and reaction.

2.4. Geometric interpretation of fundamental methane bioreactors

AR approach applied in this study seeks to incorporate geometry, calculus and mathematical optimization to understand how methane bioreactor networks can be designed systematically. This requires knowledge of the physical, mathematical and geometric properties of the different methane bioreactors that will be used to construct the attainable region. In illustrating these properties, we define two important vectors, the concentration vector (C) and the rate vector, $r(C)$ of the anaerobic treatment process, which are used to study the characteristics of the fundamental reactor types in section (a) and (b) below.

$$C = [S_{BVS} \;\; S_{VFA} \;\; X_{ac} \;\; X_{me}]^T \tag{14}$$

$$r(C) = [r_{S_{BVS}} \;\; r_{S_{VFA}} \;\; r_{X_{ac}} \;\; r_{X_{me}}]^T \tag{15}$$

(a) Continuous stirred tank anaerobic reactors (CSTR)

The anaerobic CSTR is presented mathematically as a system of nonlinear equations Eq. (16), where solving the system to obtain the roots at a given organic load (C_f) and for different digestion times (τ_i *for* $i = 1$ *to* n) results in a set of points referred to as a CSTR locus [17].

$$C = C_f + \tau r(C) \tag{16}$$

In a geometric interpretation, if we define the mixing vector as ($C - C_f$), then for a given rate expression, $r(C)$ and organic load (C_f), the roots (C) of the system of nonlinear equations results in a mixing vector which is collinear to the rate vector $r(C)$, evaluated at the roots [4]. This implies states generated by an anaerobic CSTR cannot be part of a true AR boundary since the rate vectors evaluated at CSTR points may point out of the boundary otherwise; it becomes possible to extend the region.

(b) Anaerobic plug flow reactors (APFR)

The governing equations of anaerobic plug-flow reactor is a system of first order ordinary differential equations Eq. (17), where a phase plane presentation of the solution of the system for a given organic load and digestion time is called PFR trajectory [17].

$$\frac{dC}{d\tau} = r(C) \tag{17}$$

Geometrically, the rate vector evaluated at points on the PFR trajectory is tangent to all points on the trajectory [4]. This implies that if rate vectors on the AR boundary evaluated at points on CSTR locus point out of the region, then it is also possible to extend the AR by

Table 1
Summary of the operational guidelines for selecting methane bioreactors.

Methane bioreactor	Effluent characteristics	Loading capacity ($kgCOD/m^3d$)	Ref.
Expanded Granular Sludge Blanket (EGSB)	Cold and dilute wastewater, foaming, long chain fatty acids	40–45	[1]
Upflow Anaerobic Sludge Blanket (UASB)	More concentrated wastewaters	15–32	[19]
Anaerobic Filter (AF)	Soluble types of wastewater	5–15	[19]
Anaerobic Baffled Reactor (ABR)	Mostly used for blackwater	1–12	[19]
Anaerobic Contact Reactor (ACR)	High strength COD and lipid concentrations higher than 150 mg/l	2–5	[20]
		< 10	[1]
Anaerobic Sequential Batch Reactor (ASBR)	Low-flow applications wider variations in wastewater strength	Not applicable	[21]
Anaerobic Continuous Stirred Tank Reactor (ACSTR)	Slurries with a %TS between 2 and 10	Retention time of 14 to 28 days	[20]
Anaerobic Plug Flow Reactor (APFR)	Slurries with TSS between 11 and 14% TS	Retention time of 15 to 20 days	[5]

running a PFR after a CSTR. In industrial practice, for an anaerobic treatment process operated using a CSTR the gas production and solids reduction can be improved if a PFR is combined with the CSTR and operated in series.

(c) Batch and fed-batch methane bioreactors

For methane bioreactors operated with batch and fed-batch regimes, [17] introduced the use of appropriate transformations whereby reactor structures in continuous flow systems can use to form related batch and fed-batch structures. This implies methane bioreactors operated in batch and fed-batch modes can also be designed and improved using techniques in AR theory.

It is not the objective of this article to go into a full description of techniques in AR theory. Readers interested in the technique can find relevant resources presented in Ming et al. [4].

3. Framework for designing methane bioreactor network

3.1. Process characteristics for model simulation

Anaerobic digestion of five different organic substrates has been considered to run the models and estimate the kinetic parameters required to completely define the rate vectors necessary for construction of AR. Experimental data for digestion of the different substrates, cow manure, diary manure, horse manure, chicken manure, and swine manure were obtained from [22]. Amongst the two model outputs, $\%VSR$ was reported directly from the experiments while Y_{CH_4} was computed using Eq. (18).

$$\gamma_{CH_4} = \frac{EMY_{90}}{HRT} \times VSL \tag{18}$$

Table 2 presents the computed/obtained values of Y_{CH_4} and $\%VSR$ to be used for model validation as well as the operational parameters for anaerobic digestion of the different substrates. The duration for 90% methane production was used as the hydraulic retention time (HRT) and the corresponding 90% of the methane yield value was computed.

3.2. Parameter estimation and model validation

(a) Identification of temperature dependence model

The determination of the Ratkowsky parameters (B and C) as well as T_{min} and T_{max} was made by fitting the Chen and Hashimoto curve (Fig. 6) for temperature dependence on growth rate, cited by [16] to the Ratkowsky expanded square root model. This was done using the Matlab routine '*nlinfit*', for nonlinear regression (Mathworks Natick, NA).

The 95% marginal confidence intervals and joint confidence regions of the estimated Ratkowsky parameters were computed using Eqs. (19) and (20) respectively.

$$\hat{\beta} \pm t_{\upsilon,\alpha/2} s_{\hat{\beta}_i} \tag{19}$$

$$(\beta - \hat{\beta})^T (J^T J)(\beta - \hat{\beta}) \leq p\sigma^2 F_{(1-\alpha),p,(n-p)} \tag{20}$$

where $s_{\hat{\beta}_i}$ is the approximate standard errors of the parameter estimates, computed by Eq. (21).

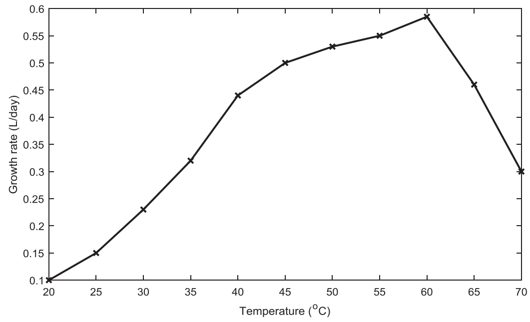

Fig. 6. Chen and Hashimoto curve for temperature dependence of growth rate.

$$s_{\hat{\beta}_i} = \sqrt{diag(cov(\beta))} \tag{21}$$

(b) Identification of the anaerobic digestion model

The following parameters $k = [k_1, k_2, k_3, \vartheta, \gamma_s]$ are to be estimated while the values of all other parameters particularly $K_{s_{ac}}, K_{s_{me}}$ *and* $K_{i_{me}}$ are maintained as in the original Hill model. The parameter estimation consisted of iteratively searching for parameter values that minimizes the squared error between the outputs predicted by the parameterized model and observed experimentally, Eq. (22).

$$\min_k \varepsilon(x, k) = \sum (\gamma_{CH_4}(k) - \gamma^e_{CH_4})^2 + (VSR(k) - VSR^e)^2 \tag{22}$$

For this purpose, the Matlab optimization routine, *fmincon* was used, where the dynamic methane bioreactor model integrated numerically using the Runge-Kutta 4–5th order method implemented by the Matlab *ode45* routine

3.3. Defining attainable region for anaerobic treatment process

After estimating the model parameters, the complete model for the anaerobic process becomes defined and can now be used for AR analysis, which is the object of the following section. A stoichiometric scheme of the bioreaction occurring in the methane bioreactor consists of two main reactions catalyzed by acid-forming bacteria, Eq. (23) and methane-forming bacteria Eq. (24).

$$k_1 S_{BVS} \xrightarrow{rX_{ac}} X_{ac} + k_2 S_{VFA} \tag{23}$$

$$k_3 S_{VFA} \xrightarrow{rX_{me}} X_{me} + k_4 CH_4 \tag{24}$$

Letting rows 1–5 correspond to S_{BVS}, X_{ac}, S_{VFA}, X_{me} and CH_4 respectively, the stoichiometric coefficient matrix A is therefore a 5×2 matrix, given by Eq. (25).

$$A = \begin{bmatrix} -k_1 & 0 \\ 1 & 0 \\ k_2 & -k_3 \\ 0 & 1 \\ 0 & k_4 \end{bmatrix} \tag{25}$$

Table 2
Process characteristics and experimental data for model validation.

Type of waste to be treated	HRT (T_{90}) (*days*)	EMY (mL/gVS)	90%EMY (mL/gVS)	VSL (gVS/l)	VSR (%)	γ_{CH_4} ($l/m^3/d$)
Diary manure (DM)	28	204	183.6	3.5	58.6	22.95
Horse manure (HM)	37	155	139.5	3.5	52.9	13.20
Goat manure (GM)	44	159	143.1	3.5	46.4	11.38
Chicken manure (CM)	18	259	233.1	3.5	81.4	45.32
Swine manure (SM)	17	323	290.7	3.5	81.4	59.85

Since there are two independent reactions participating in the system ($Rank(A) = 2$), we expect the set of points generated by the anaerobic treatment process to reside in a two-dimensional subspace in $\mathbb{R}^5$. As all model outputs are functions of volatile fatty acids and concentration of methanogenic archae, it is sensible to generate the AR in ($S_{VFA} - X_{me}$) space, which provides information required to maximize gas production and volatile solids reduction.

The number of dimensions in which the AR must be constructed was reduced using the concept of yield coefficients, which has been used previously to reduce the number of dimensions during AR analysis [23]. This is possible because using yield coefficients, we can calculate the reaction rates of S_{BVS} and X_{ac} as functions of production rates of S_{VFA} and X_{ac} as shown in Eqs. (26) and (27).

$$r_{X_{ac}} = \frac{1}{k_2}(r_{S_{VFA}} + k_3 r_{X_{me}}) \tag{26}$$

$$r_{S_{BVS}} = -\frac{k_1}{k_2}(r_{S_{VFA}} + k_3 r_{X_{me}}) \tag{27}$$

This implies that the concentrations of BVS and acidogenic bacteria can be expressed as a function of VFA and methanogenic archae concentrations as in Eqs. (28) and (29).

$$X_{ac} = X_{ac_0} + \frac{1}{k_2}[S_{VFA} - S_{VFA_0} + k_3(X_{me} - X_{me_0})] \tag{28}$$

$$S_{BVS} = S_{BVS_0} - \frac{k_1}{k_2}[S_{VFA} - S_{VFA_0} + k_3(X_{me} - X_{me_0})] \tag{29}$$

The ability to calculate X_{ac} and S_{BVS} as a function of X_{me} and S_{VFA} allow us to also express the rate and concentration vectors of X_{me} and S_{VFA} exclusively. In other words, for each X_{me} and S_{VFA} in the ($S_{VFA} - X_{me}$) space we can calculate a rate vector that uniquely determines the CSTR locus and PFR trajectory from a specified organic load.

Four main steps used to construct the AR include

- A determination of the PFR trajectory from the organic load.
- A determination of the CSTR locus from the organic load.
- An extension of the AR boundary by running a series of PFR from each CSTR point.
- Convexifying the entire set of points and test the AR against necessary conditions.

The CSTR equations were solved using Newton method, implemented by the Matlab routine ‘fsolve’ while the PFR equations were solved using the Matlab *ode45* routine for solving non-stiff differential equations. The convex hall of the entire set of geometric points is obtained by using the Matlab ‘*convhull*’ routine, which implements the Qhull algorithm (Mathworks, Natick NA).

3.4. Design optimization with attainable regions

AR theory offers advantages compared to other optimization techniques in that by computing the AR, we have all answers to all possible optimization problems, and all that is left is to introduce an objective function that answers our specific design objective. This is done by formulating the objective function in the ($S_{VFA} - X_{me}$) space and determining the point where the objective function intersects the AR boundary.

Our two design objectives, volumetric production rate, Eq. (12) and percentage of volatile solids reduction, Eq. (13) are reformulated in a way that can be plotted on the AR as shown in Eqs. (30) and (31) respectively.

$$X_{me} = \frac{\gamma_{CH_4}}{0.5\mu_{me}(k_3 - 1) \times 1000} \tag{30}$$

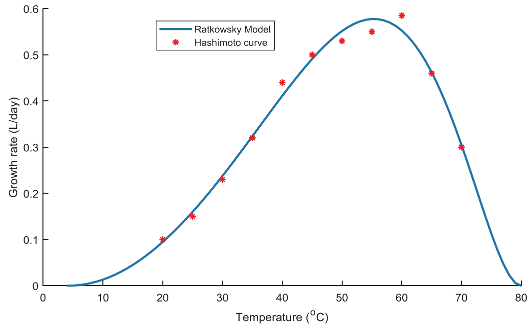

Fig. 7. Fitting of temperature dependence model to test data.

$$X_{me} = \frac{VSR \times S_{VS_0}}{k_3\mu_{me} \times 100} \tag{31}$$

Eq. (30) and (31) can respectively be used to graphically determine the volumetric methane productivity and volatile solids reduction in the ($S_{VFA} - X_{me}$) space.

4. Results and discussions

4.1. Parameter estimates and model validation

(a) Fitting of temperature dependence model

The first set of analyses estimated the parameters of the Ratkowsky model and examined its ability to predict the temperature dependence of the specific growth rate using the Hashimoto curve. As shown by Fig. 7, the Ratkowsky model gives a good prediction of the experimental data and can be used to model the temperature dependence of the methane bioreactor. In accordance with the present results, previous studies have demonstrated that the model can predict temperature dependence in bioreactors producing hydrogen under anaerobic conditions [24].

Fig. 8 shows the confidence contours of model parameter estimates, which show varying degrees of correlation, some being positively correlated and others being negatively correlated. The correlation amongst the model parameters does not have an “intuitive” explanation because it is a consequence of the estimation procedure itself, and does not reflect some aspect of the temperature dependence. The two-dimensional regions only show where it is reliable to select a parameter value taking into consideration correlation from the other parameters. The actual parameter estimates are 0.02, 0.05, 4.22 and 79.96 respectively for B, C, $T_{min} and T_{max}$. The results imply that at a minimum temperature of 4.22 °C and a maximum temperature of 79.96 °C, growth rate in the methane bioreactor becomes zero. The model offers advantage over the conventional Arrhenius model in that it represents realistic aspects of the anaerobic digestion process where the growth rate initially increases with increasing temperature up to a maximum after which its starts decreasing with increasing temperature.

(b) Validation of the dynamic state model

The parameter estimates of the methane bioreactor model for each of the organic substrates have been made using a nonlinear optimization solver, with the gradients computed using numerical perturbations at every iteration. The convergence history of the sum of squared error for all the organic substrates is presented in Fig. 9, which reveals two important findings. First, the different substrates show different convergence developments, and a possible explanation could be that the differences in substrate parameters offer different degrees of stiffness to

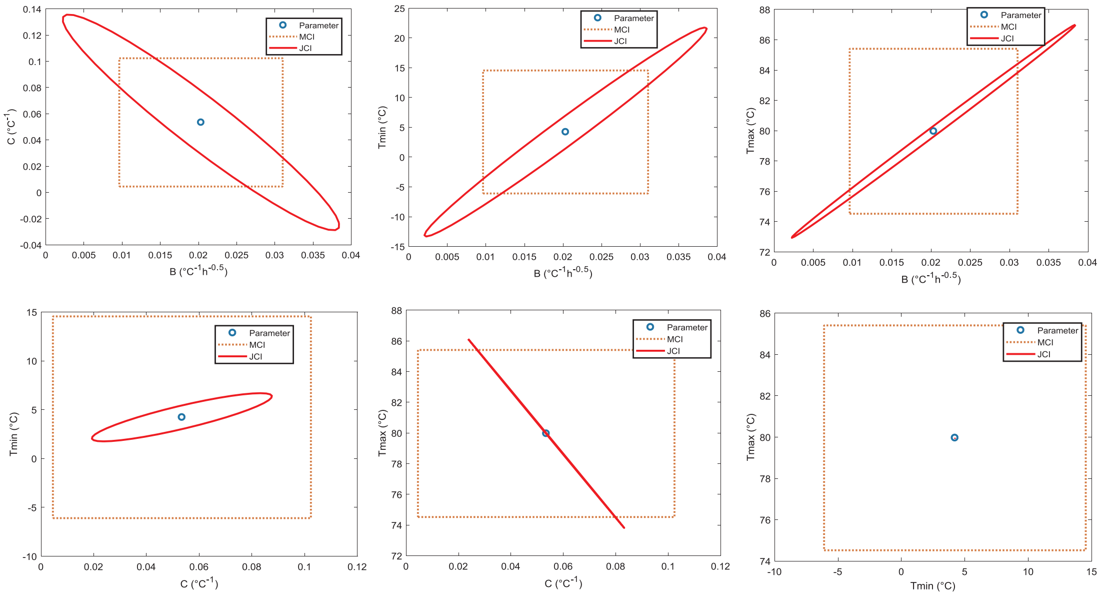

Fig. 8. Confidence contours of parameter estimates for temperature dependence model.

the optimization problem. Secondly, the errors between model and data approach zero for all the data sets as the number of iterations increases, implying the problem converges to a feasible solution as local minimizers normally select model parameters at every iteration such that the objective function is monotonically decreasing [25]. Table 3 presents the parameter estimates and compares the simulated and experimental values. It is apparent from the table that the model gives a good prediction of the experimental data.

4.2. Geometric representations and methane bioreactor structures

The objective was to propose optimal methane bioreactor structures

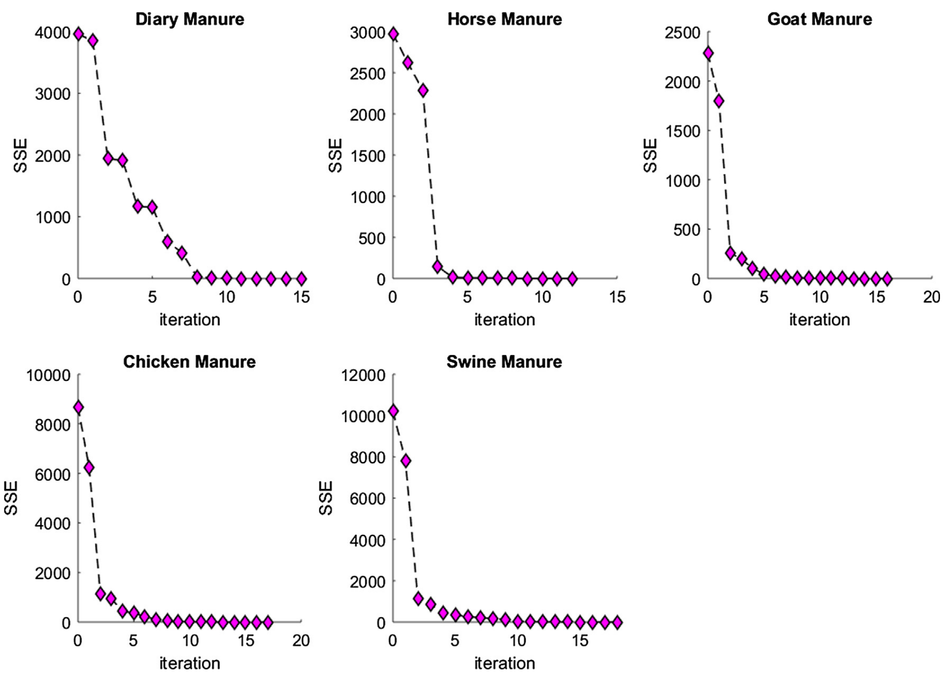

Fig. 9. Convergence history of parameter estimation process for different organic substrates.

Table 3
Parameter estimates and comparison of simulated and experimental data.

Substrate	Parameter estimates					Experimental digester values		Predicted digester values		Model error
	k_1	k_2	k_3	ϑ	γ_s	VSR	γ_{CH_4}	VSR	γ_{CH_4}	ε
DM	1.096	0.096	5.351	0.519	0.503	58.62	22.95	58.60	22.91	0.0435
HM	1.140	0.140	2.344	0.671	0.460	52.91	13.20	52.97	13.25	0.0603
GM	4.074	3.074	6.341	0.854	0.366	46.41	11.38	46.42	11.36	0.0153
CM	1.251	0.251	2.772	0.334	0.433	81.43	45.32	81.41	45.34	0.0300
SM	1.408	0.408	9.02	0.535	0.346	81.44	59.85	81.43	59.82	0.0154

for anaerobic treatment of different substrates by constructing candidate ARs in two-dimensional $S_{VFA} - X_{me}$ space. Figs. 10–14 present attainable regions for anaerobic treatment of the different substrates in a two-dimensional space of volatile fatty acids (x-axis) and methanogenic concentration (y-axis). The reaction rate vectors generated by the system of rate expressions $r(C) = [r_{S_{VFA}}\, r_{X_{me}}]^T$ evaluated at $C = [S_{VFA} X_{me}]^T$ is plotted over the regions for the different substrates. Two very important observations can be made from the figures. (1) The nature of the attainable region changes with different substrates. This is because the attainable region is unique for a given kinetics and organic load, and a change in kinetics generally affect the region and its associated reactor structures [4]. (2) All the rate vectors either point into the region (along the mixing line) or are tangent to the AR boundary (along the PFR trajectory), which is an interesting property indicating that there are no combinations of reactors that extend the region further.

As mentioned in Section 2.4, each methane bioreactor type exhibits unique geometric properties, which can be used together with the AR boundary to obtain reactor structures that define the limits of achievability for every substrate. The boundary of the attainable regions is the convex hull for the set of all points achievable by reaction and mixing. In AR theory, the convex hull is the smallest subset of a set of points that can be used to generate all other points by reaction and mixing [4]. Geometrically, a convex hull is a finite convex polytope enclosed by a finite number of hyperplanes, which is interpreted in a two-dimensional space as the smallest polygon enclosed by planar facets such that all of the elements lie on or in the interior of the polygon [26]. The interpretation of the boundary into reactor structures will be illustrated using Fig. 10. The point A is the feed, while the region defined by ABC is the AR. The convex segment AB is the PFR trajectory while segment A to D is the CSTR locus. The curves represented by E (which ends at the point C) are trajectories obtained by running PFR from points on CSTR locus. The point C is therefore obtained by running a CSTR from point A followed by a PFR from CSTR. Straight lines on the AR boundary represent mixing (lines AC and BC) while curved surfaces represent reaction (section AB). Concentrations along the line AC (C_{AC}) can be obtained by mixing point A and C, Eq. (32) (the lever-arm rule) and the reactor structure is therefore given by a CSTR + PFR (point C) with a bypass from point A. Concentrations on the line BC (C_{BC}) can be obtained by mixing point B and C, Eq. (33) and the required reactor structure is given by a PFR + CSTR (point C) run in parallel with a PFR (point B) with both contents mixed at the end. Similar reactor interpretations were made for the other substrates as presented in Figs. 11–14.

$$C_{AC} = \alpha C_A + (1 - \alpha)C_C,\ 0 \le \alpha \le 1 \tag{32}$$

$$C_{BC} = \alpha C_B + (1 - \alpha)C_C,\ 0 \le \alpha \le 1 \tag{33}$$

where α is known as the mixing ratio.

4.2.1. Reactor structures for optimal methane productivity and volatile solids reduction

Once the AR has been determined, the limits of achievability by the system for the different substrate degradation kinetics and organic load are known. The boundary of the AR can then be used to answer different design or optimization questions related to the system. This is done by defining an appropriate objective function in terms of the AR space variables and overlaying onto the AR to see where intersects the boundary [4]. The reactor structures corresponding to sections of the

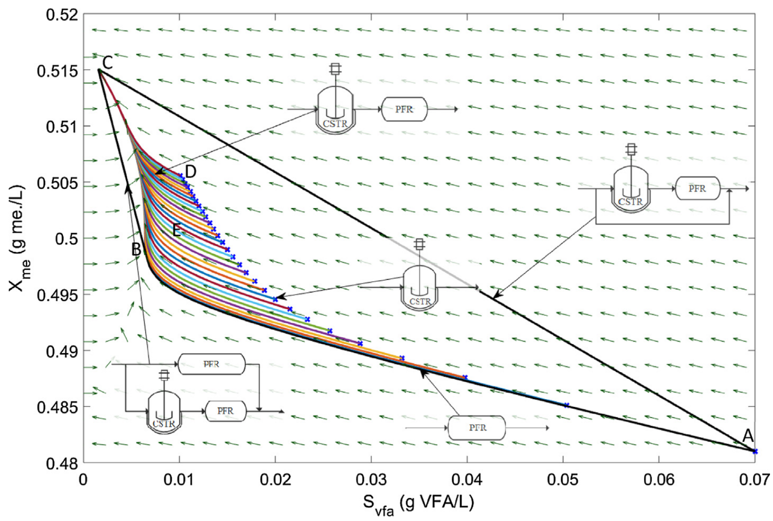

Fig. 10. Attainable region for anaerobic treatment of diary manure in 2D space.

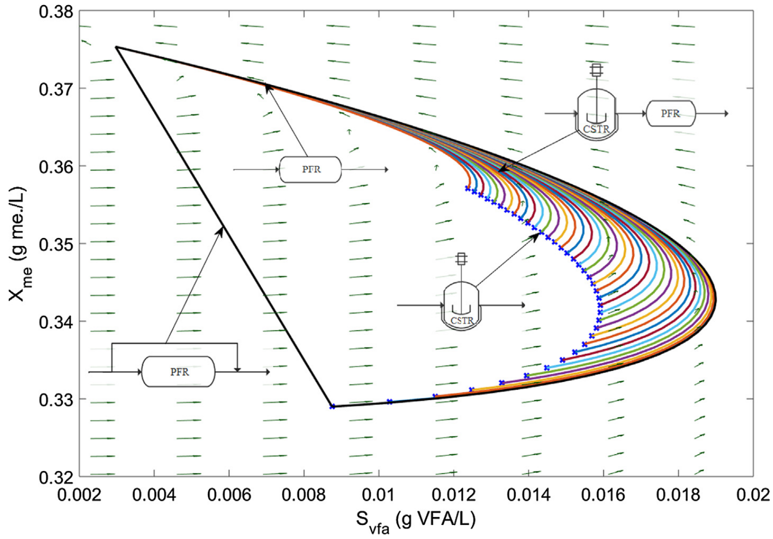

Fig. 11. Attainable region for anaerobic treatment of horse manure in 2D space.

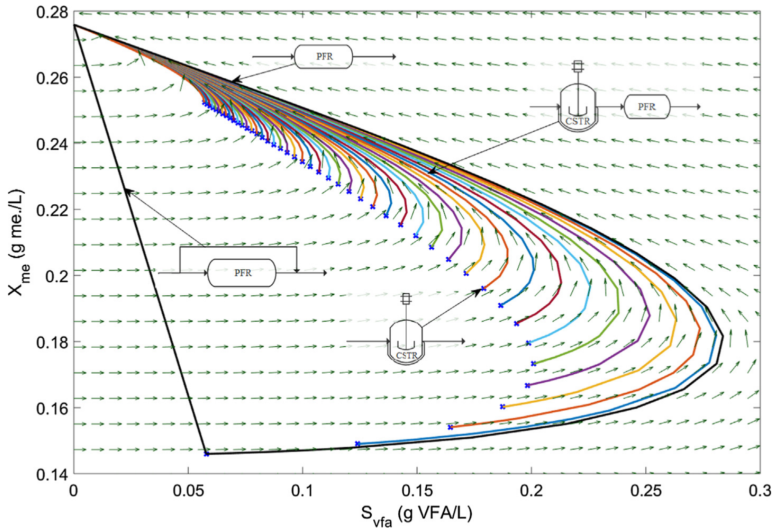

Fig. 12. Attainable region for anaerobic treatment of goat manure in 2D space.

AR that intersect the objective function are optimal structures relative to the specified objective function. Figs. 15–24 presents a number of contour lines for methane productivity (Y_{CH_4}), Eq. (30) and percentage volatile solids reduction (*VSR*), Eq. (31) overlaid onto the AR for the different organic substrates (plots to the right of each figure is a closer zoom of that to the left). Analyzing the figures reveals four important remarks. (1) For each value of Y_{CH_4} and *VSR*, there exist many points of intersection with the AR, with every intersection point being an optimal operating point. This implies that there are multiple optima for the objective functions and more strikingly an infinite number of optima, if we include all concentrations on the mixing line joining the two points of intersection on the AR boundary. The results corroborate the findings of some of the previous studies using AR to optimize for a reactor structure, where multiple optima is sometimes observed [4]. However, if we limit our choice to the points on the AR boundary, we have two possible operating points and their associated reactor structure, which can be used to achieve a specified objective for the different substrates (see Figs. 25–27). (2) As the value of Y_{CH_4} and *VSR* increases, the objective function shifts diagonally towards the positive quadrant and reaches a point where it no longer intersect the AR. This observation is quite interesting as it illustrates the limits of achievability of the system. The values of Y_{CH_4} and *VSR* where the objective function no longer intersects the AR are values that cannot be attained by the system for the specified organic load and reaction kinetics. The diagonal shift of the curve implies higher concentrations of methanogens and volatile acids are required to achieve higher methane productivity and volatile solids reduction, which is true for the anaerobic treatment process [2]. (3) The values of Y_{CH_4} and *VSR* for which the objective functions no longer

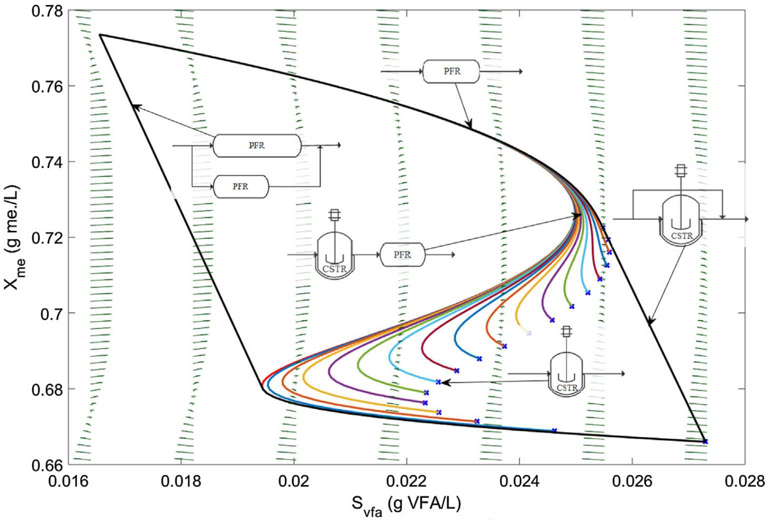

Fig. 13. Attainable region for anaerobic treatment of chicken manure in 2D space.

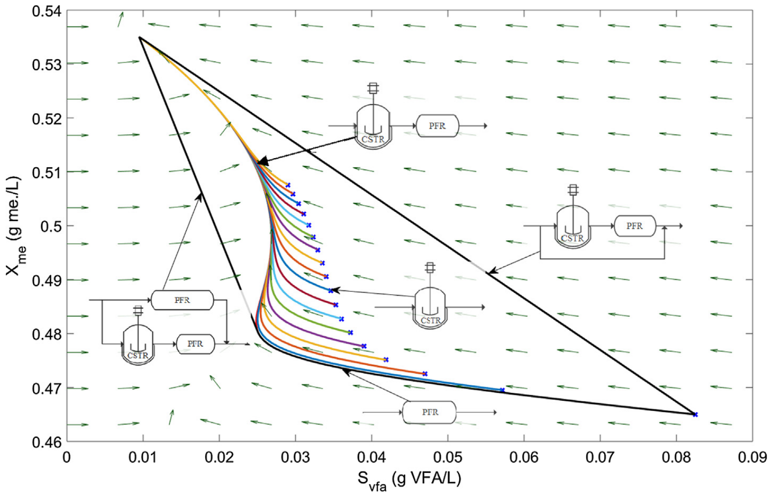

Fig. 14. Attainable region for anaerobic treatment of swine manure in 2D space.

intersect the AR differ for each organic substrate. This is explained by the fact that different substrates have different degradation kinetics and as the kinetics of the system changes, the limits of achievability changes [4]. (4) Both objective functions show similar patterns but with different magnitudes. This is a realistic observation because during anaerobic digestion, volatile solids are not consumed as all input VS minus the one incorporated in bacterial mass ends up in the methane produced [1]. This implies waste stabilization (VS reduction) only occurs in the methane formation step and the profile for methane recovery should therefore be similar to the profile for volatile solids reduction [2]. The difference in magnitude comes from the fact that some of the input VS is incorporated in new cell biomass.

Figs. 25–29 presents an illustration of the methane bioreactor structures required to attain specific methane productivities for the five organic substrates considered. It should be noted that if the specified methane productivity is changed the optimal reactor structure would also change. Observe that the optimal reactor structure has not changed in this instance even though the kinetics and associated AR have changed. However, this result is unique to the kinetics. Generally, a change in the kinetics may affect the AR and hence the optimal reactor structure associated with it.

As earlier mentioned in Section 2.3 there exist different methane bioreactors with a plug flow model of operation and the actual choice is to be made by the designer based on the criteria presented in Table 1. If the process is to be operated in batch mode, the transformations mentioned in Section 2.4 can be applied to the continuous reactor system to get corresponding batch reactors.

The results show that the attainable regions and their optimized parameters differ for each digested substrate and the optimal networks are made of different combinations of digesters operated in a continuous (axial mixing) and/or plug flow (no axial mixing) mode. This substrate effect on attainable regions shows great promises as it paves

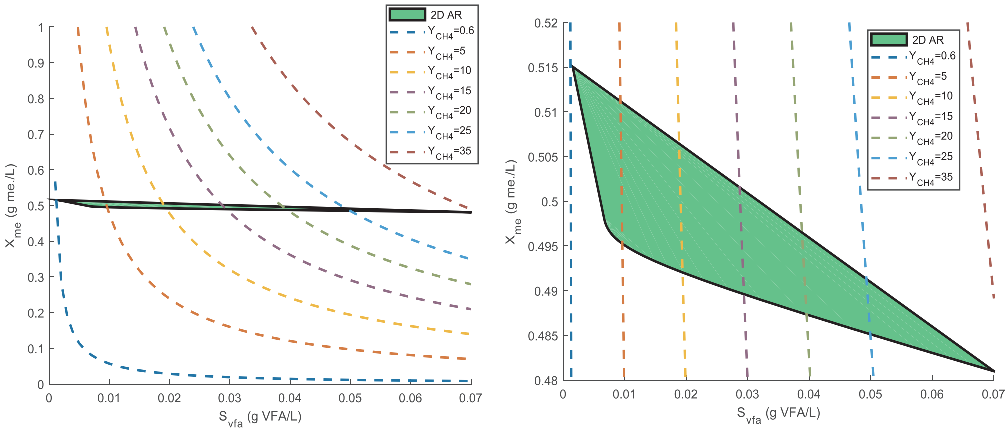

Fig. 15. Contours of volumetric methane productivity overlaid onto AR for diary manure.

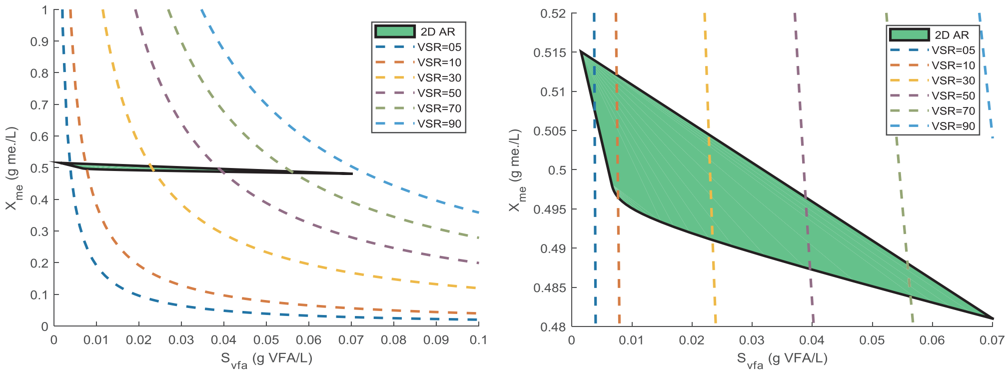

Fig. 16. Contours of volatile solids reduction overlaid onto AR for diary manure.

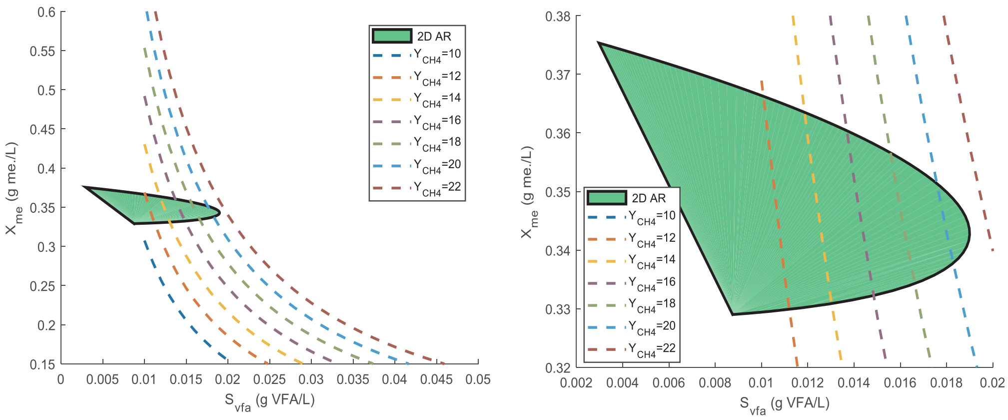

Fig. 17. Contours of volumetric methane productivity overlaid onto AR for horse manure.

the way for other substrates such as food waste, lignocellulosic waste, co-digested feeds, etc. This study though preliminary presents a major breakthrough in extending the use of digester networks to solve more operational challenges as well as support retrofitting multi-stage systems into facilities where single-stage digesters already exist. Multi-stage digesters systems have gained increasing importance due to their ability to optimize every step in the anaerobic treatment process. For an already existing digester system, the attainable region concept presented in this study will show the proximity of the existing system in relation to the absolute best performance, which is important in

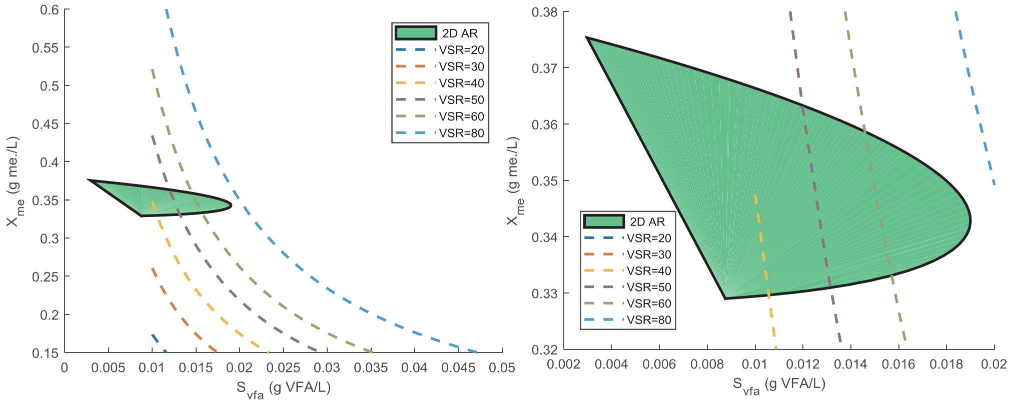

Fig. 18. Contours of volatile solids reduction overlaid onto AR for horse manure.

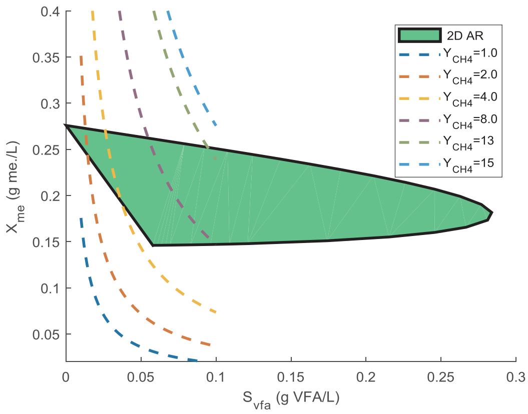

Fig. 19. Contours of volumetric methane productivity overlaid onto AR for goat manure.

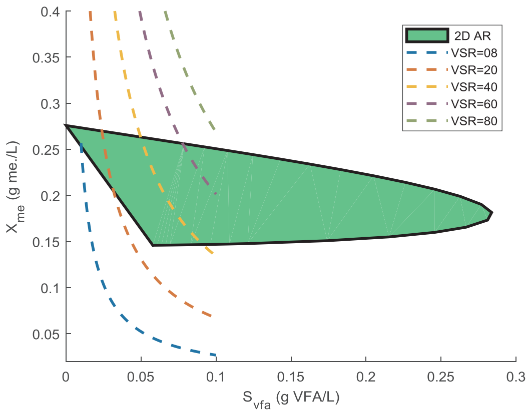

Fig. 20. Contours of volatile solids reduction overlaid onto AR for goat manure.

deciding whether or not to invest additional effort and resources to further revise the system. This is because by interpreting the anaerobic treatment process as geometric objects, we obtain (by constructing the AR) a collection of all possible output for all possible reactor designs that define a region of achievability without having to explicitly enumerate all possible design combinations. In the case where a decision is made to revise the network or to synthesize a new multistage digester network, the following steps are required:

- Identity the parameters of the simplified model presented in this study using data from the existing plant or anaerobic treatability studies. In this study, we used experimental data of diary, horse, goat, chicken and swine manure, and obtained errors between 0.01 and 0.06.
- Use the identified model to construct the attainable region and optimize a defined parameter of the plant in order to obtain an optimal network structure.
- By comparing the optimal and the existing network, points of modifications in practical operation will be evident, which includes answers to three main questions: (1) How many individual reactors do we consider in each structure? (2) What type of anaerobic reactors (CSTRs or PFRs) do we consider in each structure? (3) Whether and/or where to include recycle or bypass streams within the structure?

It should, however, be noted that unlike the superstructure optimization method [13] for reactor network synthesis, which requires defining an initial reactor structure, the AR approach does not require an existing network to synthesize an optimal network. The attainable region technique does not only define the limit of achievability of the system, but it provides reactor structures that can answer key design question relative to methane productivity and waste stabilization. This study, therefore, bridges the gap between research, development, and implementation of digester networks.

It is also interesting for readers to note that the network synthesis approach utilized in this study can also be applied for synthesis and optimization of other energy conversion processes (e.g., alcohol fermentation, gasification, pyrolysis, etc.) as well as for planning and scheduling of energy production processes. The approach provides information for both performance targeting and reactor network problems. Therefore the study offers great promises for widespread application to enhance energy generation.

5. Conclusion

The development of a systematic methodological framework for optimal synthesis of multistage digester networks has been presented. This is the first study indicating the usefulness of attainable regions, a global optimization technique for modeling configurations of multistage anaerobic digesters. A simplified model for anaerobic digestion is

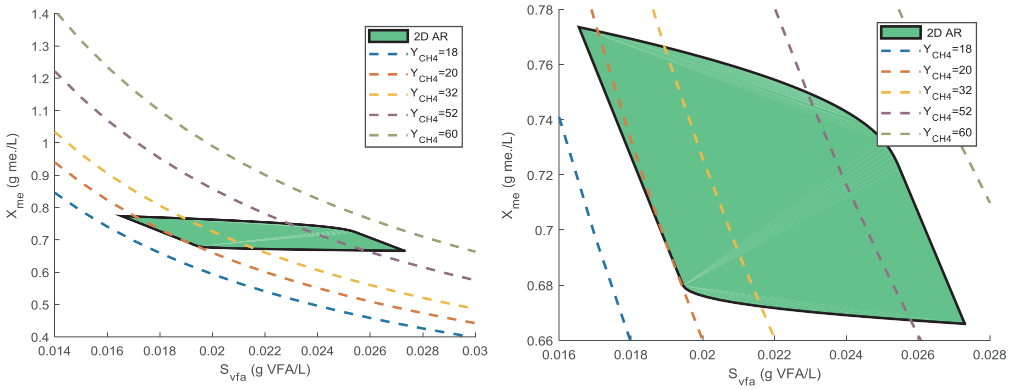

Fig. 21. Contours of volumetric methane productivity overlaid onto AR for chicken manure.

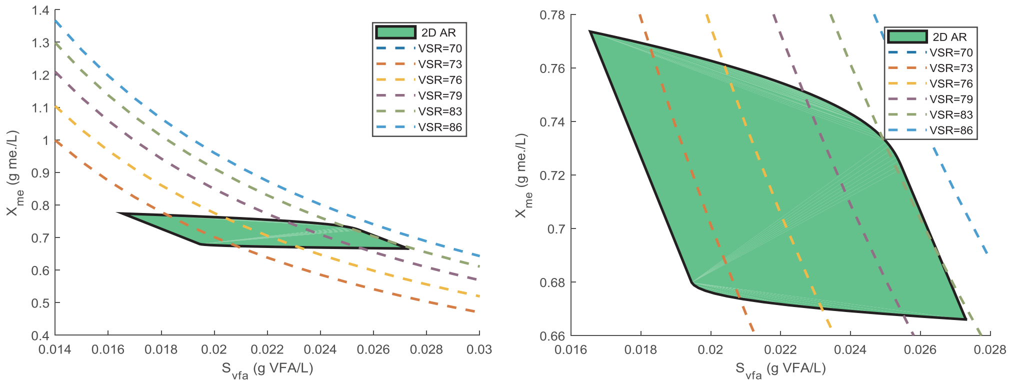

Fig. 22. Contours of volatile solids reduction overlaid onto AR for chicken manure.

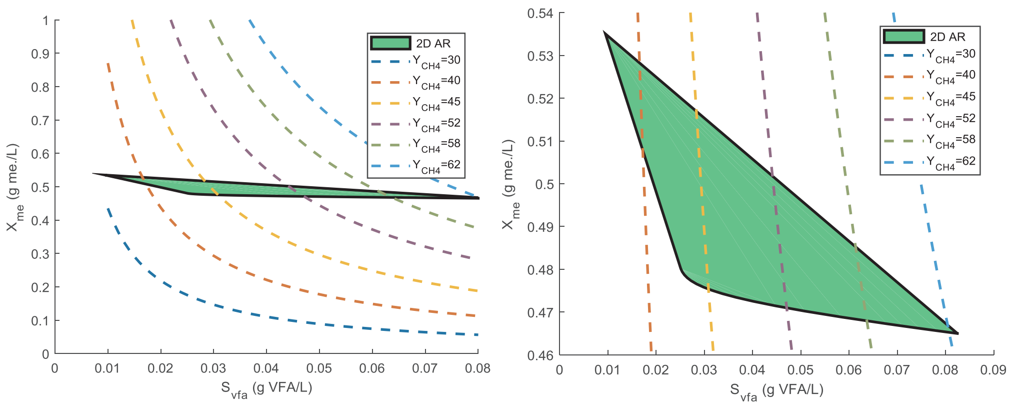

Fig. 23. Contours of volumetric methane productivity overlaid onto AR for swine manure.

formulated, and the Ratkowsky expanded square root model is presented as a reliable alternative to Arrhenius for modeling temperature dependence in methane bioreactors. Parameter estimation shows that the model predictions agree well with experimental data of diary, horse, goat, chicken and swine manure (model errors between 0.01 and 0.06). The model has been used to account for the mathematical and geometric characteristics of fundamental anaerobic digesters (Plug Flow and Continuous Stirred Tank digesters), and the results have been generalized to advanced anaerobic digesters. Two-dimensional attainable regions reveal that the optimal reactor structure differs for each digested substrate and all structures are made of digesters operated in a continuous (axial mixing) and/or plug flow (no axial mixing) mode.

This knowledge is very useful as it enables the definition of appropriate performance targets for different organic substrates, which is

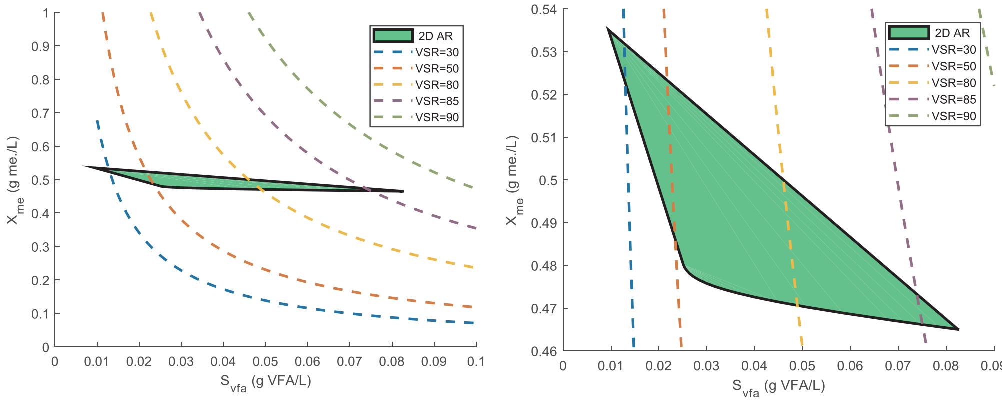

Fig. 24. Contours of volatile solids reduction overlaid onto AR for swine manure.

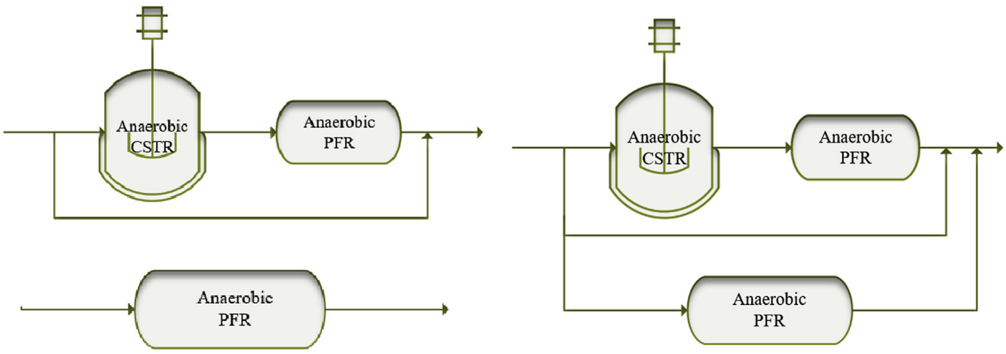

Fig. 25. Digester structures to attain methane productivity of 25 and 52 $l/m^3/d$ respectively diary manure and swine manure.

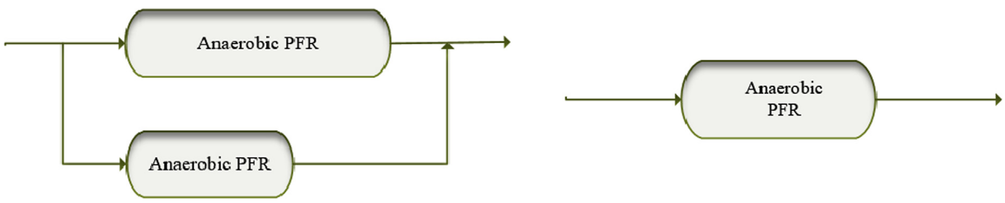

Fig. 26. Digester structures that can attain methane productivity of 18 and 20 $l/m^3/d$ respectively for horse manure, chicken manure.

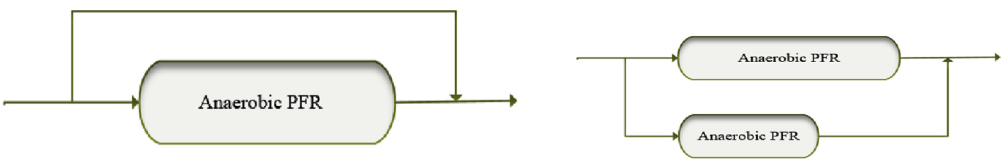

Fig. 27. Digester structures to attain methane productivity of 4.0 $l/m^3/d$ for goat manure.

useful to make design and feasibility decisions as well as support retrofitting multi-stage systems into facilities where single-stage digesters already exist. In addition, the substrate effect observed on the limits of achievability of the system shows great promises as it paves the way for other substrates such as food waste, lignocellulosic waste, co-digested feeds, etc. Further to a proof-of-concept for the geometric optimization technique, two optimization problems are formulated and solved geometrically to obtain optimal structures for anaerobic digesters that maximize volumetric methane production rate and volatile solids reduction for five different organic substrates.

As a natural progression of this study, it will be important to subject the optimized parameter and reactor structures obtained to actual experimental verification. For this reason, our next study considers residence time in a three-dimensional attainable region framework where residence time adds the third dimension. This has permitted us to design and dimension a novel compact digester consisting continuously stirred tank digester, plug flow digester as well as by-pass and recycle streams (currently under fabrication for experimental testing).

In this study, the anaerobic digester networks have been staged based on the Acid/Gas Phased Digestion technique (two-stage biochemical kinetics) in which acid-forming stage is physically separated from the methane gas-forming stage. Other studies could consider applying different staging techniques such as Staged Mesophilic Digestion; Temperature Phased Anaerobic Digestion or Staged Thermophilic Digestion.

Finally, readers should note that the attainable region technique is suitable for use not because of multiple reactors but because of multiple reactions, such as the biological reactions in anaerobic digestion involving complex metabolic pathways. However, for practicality, we have applied 2-stage lumped reaction models focusing on acid producing bacteria and methanogenic archaea to make the problem more tractable. Further studies can also consider more complex reaction schemes are comprising hydrolysis, acidogenesis, acetogenesis and parallel reactions for acetoclastic and hydrogenotrophic methanogenesis. In such cases, instead of using the graphical approach for attainable region construction presented in this study, automated approaches such as the recursive constant control policy algorithm should be adopted. This will lead to a generalization of the attainable region concept for synthesis and optimization of anaerobic digester networks.

Acknowledgments

Our team expresses gratitude to the following institutions; The Brew-Hammond Energy Centre, KNUST Ghana, The Water and Environmental Engineering Group, NTNU Ålesund and the Abunde Sustainable Engineering Group (AbundeSEG) for its immense technical support.

Funding

This work was supported by EnPe-NORAD under the project Upgrading Education and Research Capacity in Renewable Energy Technologies (UPERC-RET).

References

[1] Henze M, van Loosdrecht MCM, Ekama GA, Brdjanovic D. Biological wastewater treatment – principles, modelling and design. IWA-publishing; 2008.
[2] Wang LK, Shammas NK, Hung Y-T. Biosolids treatment processes. Handbook of Environmental Engineering 6. New Jersey: Humana Press Inc; 2007.
[3] Alford JS. Bioprocess control: advances and challenges. Comput Chem Eng 2006;30(10):1464–75.
[4] Ming D, Glasser D, Hildebrant D, Glasser B, Metzger M. Attainable region theory: an introduction to choosing an optimal reactor. Hoboken, New Jersey: John Wiley & Sons Inc; 2016.
[5] Mao C, Feng Y, Wang X, Ren G. Review on research achievements of biogas from anaerobic digestion. Renew Sustain Energy Rev 2015;45:540–55.
[6] Chong S, Sen TK, Kayaalp A, Ang HM. The performance enhancements of upflow anaerobic sludge blanket (UASB) reactors for domestic sludge treatment – a state-of-the-art review. Water Res 2012;46(11):3434–70.
[7] United States Environmental Protection Agency (EPA). Biosolids technology fact sheet, multi-stage anaerobic digestion. National Service Center or Environmental Publications (NSCEP); 2006.
[8] Metcalf, Eddy. Wastewater engineering: treatment and reuse. 4th Edition New York: McGraw-Hill; 2003.
[9] Zhang J, Loh K-C, Li W, Lim JW, Dai Y, Tong YW. Three-stage anaerobic digester for food waste. Appl Energy 2016.
[10] Akobi C, Yeo H, Hafez H, Nakhla G. Single-stage and two-stage anaerobic digestion of extruded lignocellulosic biomass. Appl Energy 2016;184:548–59.
[11] Nasr N, Elbeshibishy E, Hafez H, Nakhla G, El Naggar MH. Comparative assessment of single-stage and two-stage anaerobic digestion for the treatment of thin stillage. Bioresour Technol 2012;111:122–6.
[12] Yu L, Wensel PC, Ma J, Chen S. Mathematical modeling in anaerobic digestion (AD). J Bioremed Biodeg 2013;S4:003.
[13] Pontes RFF, Pinto JM. Optimal synthesis of anaerobic digester networks. Chem Eng J 2009;149(1):389–405.
[14] Batstone DJ. Mathematical modelling of anaerobic reactors treating domestic wastewater: rational criteria for model use. Rev Environ Sci Bio/Technol 2006;5(1):57–71.
[15] Ratkowsky DA, et al. Model for bacterial culture growth rate throughout the entire biokinetic temperature range. J Bacteriol 1983;154(3):1222–6.
[16] Hill DT. Simplified monod kinetics of methane fermentation of animal wastes. Agric Wastes 1983;5(1):1–16.
[17] Ming D, Glasser D, Hildebrandt D. Application of attainable region theory to batch reactors. Chem Eng Sci 2013;99:203–14.
[18] Angelidaki I, Ellegaard L. Anaerobic digestion in Denmark. Past, present and future. Department of Chemical Enzymology, Chemistry Faculty, Moscow State University; 2002. p. 129–38.
[19] Mang HP, Li Z. Technology review of biogas sanitation (draft). Eschborn: GIZ; 2010.
[20] Mes TZDD, Stams AJM, Reith JH, Zeman G. Methane production by anaerobic digestion of wastewater and solid wastes, in Bio-methane and bio-hydrogen: status and perspectives of biological methane and hydrogen production. The Hague, The Netherlands: Dutch Biological Hydrogen Foundation - NOVEM; 2003.
[21] Mohini S, K. SR. Sequencing batch reactor technology for biological wastewater treatment: a review. Asia-Pacific J Chem Eng 2011;6(1):3–13.
[22] Kafle GK, Chen L. Comparison on batch anaerobic digestion of five different livestock manures and prediction of biochemical methane potential (BMP) using different statistical models. Waste Manage 2016;48:492–502.
[23] Scott F, Conejeros R, Aroca G. Attainable region analysis for continuous production of second generation bioethanol. Biotechnology Biofuels 2013;6(1):171.
[24] Wang J, Wan W. Effect of temperature on fermentative hydrogen production by mixed cultures. Int J Hydr Energy 2008;33(20):5392–7.
[25] Hand DJ. Dynamic data assimilation: a least squares approach by John M. Lewis, S. Lakshmivarahan, Sudarshan Dhall. Int Statistic Rev 2007;75(3):410.
[26] Asiedu N, Hildebrandt D, Glasser D. Experimental simulation of three-dimensional attainable region for the synthesis of exothermic reversible reaction: ethyl acetate synthesis case study. Ind Eng Chem Res 2015;54(10):2619–26.

Paper 2:

Simulation of two-dimensional attainable regions and its application to model digester structures for maximum stability of anaerobic treatment process

A paper published in ***Water Research***

Citation:

ABUNDE NEBA, F., ASIEDU, N. Y., ADDO, A., MORKEN, J., ØSTERHUS, S. W. & SEIDU, R. 2019c. Simulation of two-dimensional attainable regions and its application to model digester structures for maximum stability of anaerobic treatment process. ***Water Research***, 114891.

Highlights:

- A framework is presented for using attainable regions to improve stability in low-rate digesters
- Five different patterns of kinetic inhibition (linear, exponential, competitive, non-competitive and uncompetitive) of methanogenic microorganisms are considered
- Two stability indicators: inoculum to substrate ratio and instantaneous methanogenic yield are used as design objectives
- Three digestion cases: diary manure, diary manure + lagoon inoculum and diary manure + granular sludge are considered.
- A change in the source of inoculum results in different optimal digester structures
- Methanogenic archaea are more viable in digester structures than in single digesters

Water Research 163 (2019) 114891

Contents lists available at ScienceDirect

Water Research

journal homepage: www.elsevier.com/locate/watres

Simulation of two-dimensional attainable regions and its application to model digester structures for maximum stability of anaerobic treatment process

F. Abunde Neba [a, d, e, *], Nana Y. Asiedu [c], Ahmad Addo [b], John Morken [f], Stein W. Østerhus [d], Razak Seidu [e]

[a] Abunde Sustainable Engineering Group (AbundeSEG), Ghana
[b] Department of Agricultural and Biosystems Engineering, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana
[c] Department of Chemical Engineering, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana
[d] Department of Civil and Environmental Engineering, Norwegian University of Science and Technology, Trondheim, Norway
[e] Institute for Marine Operations and Civil Engineering, Norwegian University of Science and Technology, Ålesund, Norway
[f] Faculty of Science and Technology, Drobakveien 31, 1432 Aas, Norwegian University of Life Sciences, Ås, Norway

A R T I C L E I N F O

Article history:
Received 17 April 2019
Received in revised form
18 July 2019
Accepted 19 July 2019
Available online 23 July 2019

Keywords:
Attainable regions
Low-rate digesters
Performance targets
Digester structures
Process stability

A B S T R A C T

Unlike high-rate anaerobic digesters that employ some mechanism to retain microbial sludge mass, low-rate systems use sufficiently long hydraulic retention times to ensure process stability, which becomes economically unattractive for treating large quantities of waste. This study presents the use of attainable region to develop a new strategy to enhance the stability of low-rate digesters. By considering three digestion cases, diary manure only (batch 1) or diary manure with granular (batch 2) or lagoon (batch) sludge as innoculum, the following findings were obtained. (1) For a given concentration of volatile acids in an anaerobic digester, higher concentrations of methanogenic archae can be attained using a digester structure (combination of different digesters) as opposed to single digester. (2) For a given digested substrate, a change in the source of inoculum results in a change in the limits of achievability by the system (attainable limits for batches 1, 2 and 3 were 46.486$(g/L)^2$, 5.562$(g/L)^2$ and 0.551$(g/L)^2$, which resulted in performance improvements of 118.604%,175.627% and 200.436% respectively), and hence optimal digester structure. The evidence from this study suggests that the technique can be used to simultaneously improve process stability, define performance targets and propose digester structures required to achieve a given target.

1. Introduction

The anaerobic digestion process for waste treatment and biogas generation has received considerable attention from the scientific community due to rising demand for renewable energy and environmental sanitation. As with any other bioprocess, central to the operation of the anaerobic treatment process is the anaerobic digester in which microorganisms grow, breakdown organic pollutants and produce methane-rich biogas (Alford, 2006). Unlike aerobic treatment systems in which the loading rate is limited by the supply of a reagent (such as O_2), the loading rate of anaerobic reactors is limited by the processing capacity of the microorganisms (Mes et al., 2003). These microorganisms generally include two groups: Acid-forming and methane producing microorganisms (Demirel and Yenigun, 2002), with the latter having a growth rate five times relatively higher than the former (Henze et al., 2008). Therefore the stability of anaerobic digesters is highly dependent on the viability and mass of methanogenic archae retained in the digester with respect to a given substrate concentration. The specific growth rate of methanogenic archae increases with concentration of volatile fatty acids until a maximum specific growth rate is reached above which volatile acids turn to inhibit growth rate (Henze et al., 2008; Chen et al., 2008, 2014). Hence an optimal archae to acid ratio (generally referred to as inoculum to substrate (I/S) ratio) is necessary to ensure an optimal efficiency of biogas production from anaerobic digesters. This explains why biodigester designs that

* Corresponding author. Department of Civil and Environmental Engineering, Norwegian University of Science and Technology, Trondheim, Norway.
E-mail address: fabricen@stud.ntnu.no (F. Abunde Neba).

https://doi.org/10.1016/j.watres.2019.114891
0043-1354/

maximize retention of microbial biomass are crucial to the stability of the anaerobic treatment process and hence its industrial efficiency compared to other biological treatment processes. One of the major causes of failure in the anaerobic treatment process is inhibition, which depends on the components of the digester, byproducts of microbial metabolism as well as a combination of loading rate and retention time, which can result in microbial wash out or inhibition from chemical species. Of the two main type of anaerobic digester systems, 'high rate' systems (e.g. Contact Process, Anaerobic Filter, Fluidized Bed, UASB, EGSB) enhance process stability by employing some mechanism either to retain microbial sludge mass in the digester or to separate the sludge from the effluent and return it to the digester (Mes et al., 2003; Henze et al., 2008). On the other hand, 'low rate' systems (e.g. CSTR or PFR), use sufficiently long hydraulic retention times to ensure process stability, which becomes economically unattractive for treating large quantities of waste or requiring large digester volumes if a given quantity of waste must be treated (Mes et al., 2003). Hence alternative techniques that maximize process stability in low-rate anaerobic digesters will be a major breakthrough in the application of anaerobic treatment process. The use of digester networks, in which multiple digesters are designed to operate as a single unit is such technology (EPA, 2006). It is well known that each type of anaerobic digester has specific characteristics often making them more appropriate under specific substrate or digester conditions. In addition, the anaerobic digestion (AD) process involves multiple reactions (each catalyzed by different groups of microorganisms) and when operated in a single digester, the process conditions are only suitable for all the reactions but not optimal for any particular reaction. Hence a combination of digesters allows for the flexibility and possibility of improving overall process performance. Previous experimental studies confirming the efficacy of digester networks have only been limited to series combinations (Zhang et al., 2017; Akobi et al., 2016; Nasr et al., 2012) with a lot of empiricism in the design process. In particular, some of the plants that use the series digester combinations cannot prove whether there exist (or not) other network configurations that produce better performance. In other words, there exist the problem of local optimum or multiple solutions (existence of other digester combinations that achieve same or improved results). In our recent publication, Abunde et al. (Abunde Neba et al., 2019) we solved this challenge by developing a novel theoretical framework for optimal synthesis of digester networks based on the concept of attainable regions. The attainable region is a collection of all possible output for all possible digester designs by interpreting the anaerobic digestion process as a geometric object that define a region of achievability without having to explicitly enumerate all possible design combinations (Ming et al., 2016). In the previous study, we concluded that a change in the type of digested substrate results in a change in the limits of ahievability (as well as the optimal combination of digesters), while considering the volumetric methane productivity and waste stabilization as design objectives. In the current study we aim to illustrate how the attinainable region concept can be used to solve instability problems in low rate anaerobic digesters. Unlike the previous study that considered different organic substrates, this study considers same substrate for different sources of inoculum and uses I/S ratio and instantaneous methanogenic yield as design objectives. In other words, we lay down a theoretical framework to design an optimal digester combination that gives the desired stability parameters (I/S ratio or instantaneous methanogenic yield) based on the concept of attainable regions.

It is important for readers to note that the attainable region is unique for given reaction kinetics (model structure and/or parameter values), and anaerobic biodegradation kinetics depends on the inhibitory conditions or type of organic substrate in the digester. All inhibitory conditions in anaerobic digesters will often upset the balance between acid-forming and methane-producing microorganisms resulting in accumulation of volatile acids (Chen et al., 2014). Different inhibitory conditions and/or substrates will result in different kinetic behaviour of volatile acids on methanogenic archae, and some of the published inhibitory patterns include: competitive, non-competitive, uncompetitive, linear or exponential kinetic behaviors (Kythreotou et al., 2014). Hence by using attainable regions, we can understand how the performance of the digester (concentration of methanogens) can be enhanced (under higher concentration of volatile acids) using digester structures as opposed to single digesters.

The determination of performance targets for anaerobic digestion of different organic substrates has been investigated extensively in the past using either experimental methods (such as the biomethane potential test and spectroscopy) or theoretical methods (based on chemical composition, chemical oxygen demand or elemental composition) (Jingura and Kamusoko, 2017). However these approaches are limited to only methane yield and gives no information about the other states and hence cannot predict exact cause of process failure or inhibition. In addition, it provides no information with respect to the digester design required to achieve a defined target. This paper discusses how the attainable region concept can be used as a technique to define performance targets under different inhibitory conditions as well as model anaerobic digester configurations to optimize process stability.

2. Process modeling and model identification

2.1. State dynamic model of anaerobic treatment process

For synthesis of low rate anaerobic digesters using attainable regions, simplified models are considered most appropriate as the geometric and hydrodynamic analysis are relatively more complex. The attainable region (AR) technique is suitable for use because it can solve problems not because of multiple reactors but because of multiple reactions, such as the biological reactions in anaerobic digestion involving complex metabolic pathways. However, for practicality, the authors have applied 2-stage lumped reaction models focusing on acid producing bacteria and methanogenic archae to make the problem more tractable. Our subsequent studies will seek to consider more complex (parallel and series) reaction set to align more closely with the biochemical pathways, i.e. series of rate equations for hydrolysis, acidogenesis, acetogenesis and parallel reactions for acetoclastic and hydrogenotrophic methanogenesis. The modified Hill model (Finn et al., 2013), which was developed for anaerobic digestion of animal manure (diary, poultry, beef and swine wastes) has been selected for this study. The model presents a compromise between the overly simplistic models capable of predicting only gas production and sometimes substrate consumption and simplistic models (such as the AM2) (Bernard et al., 2001) that include a hydrolysis step, alkalinity, cation concentration, dissolved carbon dioxide and ammonia. These effects are 'lumped' into and become part of the biodegradability constant (Bo) and acidity factor (AF) present in the modified Hill model (Finn et al., 2013). The species conservation equations for the modified Hills model are presented as follows:

a) Total biodegradable volatile solids (S_1) in the liquid phase of the bioreactor

$$\frac{dS_1}{dt} = (S_{1_{in}} - S_1)D - k_1\mu_1 X_1 \quad (1)$$

b) Volatile fatty acids (S_2) in the liquid phase of the bioreactor

$$\frac{dS_2}{dt} = (S_{2_{in}} - S_2)D + k_2\mu_1 X_1 - k_3\mu_2 X_2 \tag{2}$$

c) Acidogens (X_1) in the liquid phase of the bioreactor

$$\frac{dX_1}{dt} = (\mu_1 - K_{d_1} - D)X_1 \tag{3}$$

d) Methanogens (X_2) in the liquid phase of the bioreactor

$$\frac{dX_2}{dt} = (\mu_2 - K_{d_2} - D)X_2 \tag{4}$$

e) Methane gas flow rate

$$Q_{CH_4} = V\mu_2 k_4 X_2 \tag{5}$$

The organic waste is characterized by using the two parameters, which are biodegradability (B_o), Eq. (6) and acidity (A_f), Eq. (7). B_o measures the ease with which the organic substrate can be broken down and stabilized by anaerobic bacteria while A_f of a substrate can be defined as the amount of volatile fatty acids contained in the substrate per unit mass of biodegradable volatile solids

$$S_{1_{in}} = B_0 S_{in} \tag{6}$$

$$S_{2_{in}} = A_f S_{1_{in}} \tag{7}$$

The anaerobic biodegradability can be computed via Eq. (8) while the acidity factor is computed using Eq. (19).

$$B_0 = \frac{g\ VS_{destroyed}}{g\ VS_{added}} \quad as\ HRT \rightarrow \infty \tag{8}$$

$$A_f = \frac{VFA_{in}}{B_0 \times VSL} \tag{9}$$

The modified Hill's model considers temperature dependence of the anaerobic treatment process through an empirical model, Eq. (10) and since the death rates are set to one tenth of the maximum reaction rates, Eq. (11) they are also show temperature dependent.

$$\mu_{1m}(T) = \mu_{2m}(T) = 0.012T - 0.086 \tag{10}$$

$$K_{d1} = K_{d2} = 0.1\mu_{1m} \tag{11}$$

$$10^\circ C < T < 60^\circ C$$

For the purpose of our study, the model is adapted as follows: The Monod function used to describe the growth rates of acidogenic and methanogenic microorganisms in the original model will be used only for acidogenic bacteria, Eq. (12). The growth model for methanogenic archae will vary depending on the cases presented in Table 1.

$$\mu_1 = \mu_{m1}\frac{S_1}{K_{s1} + S_1} \tag{12}$$

In addition, a new parameter, known as the acidogenic fraction in inoculum (ϑ) is however included to characterize the inoculum. The value of this parameter is lies in the range $0 \le \vartheta \le 1$ and is selected to give the best fit between model and experimental predictions.

2.2. Kinetic patterns of volatile acid inhibition

Anaerobic digestion involves the complex interaction of different groups of microorganisms but the methanogenic archae are known to be the most sensitive to inhibition (Chen et al., 2008). As the volatile acid concentration is increased, a maximum specific growth rate of methanogenic archae will be reached at a certain concentration. A further increase of the substrate concentration results in a decrease of the specific growth rate. The kinetic patterns for volatile acid inhibition have been based on modification of the Monod model, Eq. (13) for growth of methanogenic archae to include inhibition term.

$$\mu_2 = \mu_{m2}\frac{S_2}{K_{s2} + S_2} \tag{13}$$

The effect of volatile acid on microbial inhibition in anaerobic digestion has generally been modeled through two main approaches: The empirical approach, which include a linear or, exponential inhibition patterns and the enzyme kinetic approach, which include a competitive, non-competitive and uncompetitive inhibition patterns. Both approaches are lumped into Eq. (14) by multiplying the Monod model with a factor that describe the different inhibition patterns.

$$\mu_2 = \frac{\mu_{m2}S_2(1 - K_i S_2)^a}{K_{s2}\left(1 + \frac{S_2}{K_i}\right)^e + S_2\left(1 + \frac{S_2}{K_i}\right)^d}\left(1 + \frac{S_2}{K_i}\right)^c\left(e^{-K_i S_2}\right)^b \tag{14}$$

Eq. (14) presents a generalized modified Monod model to describe volatile acid inhibition on methanogenic archae from which the different inhibition cases can be derived as shown in Table 1.

It should be noted that even though there exist other product inhibition models that have been used to model growth of anaerobic microorganisms, we consider the most common ones to illustrate the effect of anaerobic digester conditions on the type of kinetic pattern used to describe the effect of volatile acids on methanogenic archae. Instead of predefining an inhibition pattern as practiced by modelers of anaerobic digestion, the authors of this study present a framework for determining the inhibition patterns before using the model for digester synthesis. Since it is not feasible to measure the specific growth rate of both microbial populations during the anaerobic treatment process, the strategy consist of using the kinetic models in a full dynamic model so that the kinetic constants can be estimated from easily measurable parameters such as volumetric biogas and total volatile fatty acid concentration.

2.3. Model identification

In order to better illustrate the different kinetic patterns and how the patterns will change with characteristics of digestion substrate, AD experiment, using diary manure (1.7% TS) mixed inoculum from different sources was selected for model identification (Zaher et al., 2009). The experiments were conducted in continuously mixed batch reactors at 35 °C. Further details on the experimental study can be obtained from the cited literature.

In order to identify the model parameters for the different kinetic cases, the adjoint-based gradient algorithm defined in Fig. 1 is implemented. First, the gradient algorithm fits the whole set of model parameters and assesses the variability of the fit using marginal and joint confidence regions of the model parameters. Second, for parameters that show a high correlation, one of them is kept constant and a readjustment of the uncorrelated set of

Table 1
Structural patterns of volatile acid inhibition.

Empirical constant					Kinetic Pattern	Model
a	*b*	*c*	*d*	*e*		
1	0	0	0	0	Linear inhibition	Dagley and Hinshelwood
0	1	0	0	0	Exponential inhibition	Aiba et al. model
0	0	0	0	1	Competitive inhibition	Anonymous
0	0	−1	0	0	Non-competitive inhibition	Haldane model
0	0	0	1	0	Uncompetitive inhibition	Andrews model

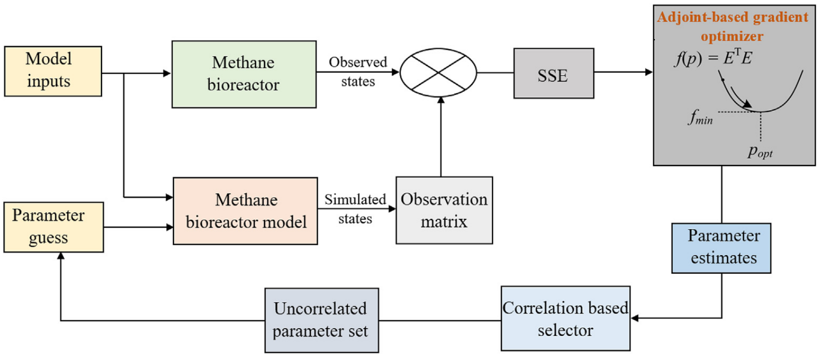

Fig. 1. Model identification framework using the adjoint-based gradient optimizer.

parameters is performed using the algorithm. This allows a more accurate adjustment of the whole set of model parameters to experimental data, as illustrated in subsequent sections.

The advantage of this procedure is that parameter estimation and variability assessment is performed simultaneously, which allows the user to better understand the model's sensitivity to different influences and obtain reliable estimates. It is not the intention of this article to go into the mathematical formulations leading to a full description of the adjoint-based gradient method for parameter estimation. Interested readers can find a detailed description of the procedure in the following literature (Benítez et al., 2017).

3. Attainable region analysis

3.1. Brief theoretical overview

The AR theory is a technique for process synthesis and optimization, which incorporates elements of geometry to understand how networks of chemical reactors can be designed and improved (Hildebrandt and Glasser, 1990; Hildebrandt et al., 1990). The attainable region is defined as the set of all possible output for all possible reactor designs that can be achieved by using the fundamental processes occurring within the system and that satisfies all the constraints placed by the system. Geometrically, the attainable region represents the region bounded by the convex hull for the set of points achievable by the fundamental processes occurring in the system. Once the AR has been determined, the limits of achievability by the system for the given kinetics and feed point is known and the boundary of the AR can then be used to answer different design or optimization questions related to the system (Ming et al., 2016). The theory provides guidelines for construction of attainable regions as well as some necessary conditions to test the results.

The following requirements are necessary before an AR analysis can be performed (Glasser et al., 1987, 1993).

Choose the fundamental processes occurring in the system.

- ➢ Choose the state variables
- ➢ Define the reaction scheme and process kinetics
- ➢ Determine the geometry of the process units.
- ➢ Define the process conditions
- ➢ Determine the objective of the optimization

Given a set of reactions and associated kinetics, the following five key steps need to be performed in order to complete an attainable region analysis.

- ➢ Define the reaction dimension and feed set
- ➢ Generate the AR using combinations of the fundamental processes
- ➢ Interpret the AR boundary in terms of reactor equipment
- ➢ Define the objective function and overlay this onto the AR to determine point of intersection with the AR boundary
- ➢ Determine the specific reactor configuration required to achieve the intersection point

Some necessary conditions for AR can be summarized as follows:

- ➢ The AR includes all feed points to the system.
- ➢ The AR is convex.
- ➢ No rate vectors point out of the AR boundary.
- ➢ Backward extension of rate vectors in the complement region do not intersect the AR

The following section outlines the methodological flow for AR construction and application for process synthesis and optimization. The framework involves five main steps (Ming et al., 2016):

3.1.1. Step 1: Preparation

This involves definition of the reaction kinetics, AR dimension, state variables (those used to represent the AR) as well as the feed point used to generate the AR. The feed point defines the initial value or the concentration of states fed into the reactor.

3.1.2. Step 2: AR construction

This step generates the AR using a combination of PFR, CSTR and mixing for two-dimensional ARs or a combination of PFR, CSTR, DSR (Differential side-stream reactor) and mixing for higher dimensional constructions. This is the most difficult and time-consuming step but also provides the most valuable information about the operating limits of the system. AR construction typically begins by determining the PFR trajectory and CSTR locus from the feed. The PFR trajectory is the set of points generated by solving the steady state model of a PFR reactor (a set of ordinary differential equations) while the CSTR locus is the set of points generated by solving the CSTR model (a set of nonlinear equations).

3.1.3. Step 3: Boundary interpretation

This step involves interpretation of the AR boundary into reactor structures, based on the fundamental characteristics of the AR boundary. The boundary of the AR is composed of reaction and mixing surfaces only. Reaction surfaces are always convex and the points that form convex sections of the AR boundary arise from effluent concentrations specifically from PFR trajectories. For a two-dimensional system, points on the AR boundary that initiate these convex PFR trajectories arise from specialized CSTRs while for a three dimensional system, they arise from DSRs. The convex hull of the set of points generated by all possible combinations of fundamental reactor types and mixing defines the attainable region.

3.1.4. Step 4: Overlay objective function

The objective function is modeled in terms of the variables used to represent the AR and then overlaid onto the AR. The points of intersection between the objective function and the AR boundary represent the optimal points of operation.

3.1.5. Step 5: Optimize

Since the entire boundary of the AR has been interpreted in terms of reactor structures (step 3), the particular reactor structure required to achieve the optimal operating points (point of intersection) is known.

Summarily, starting from the feed point, the procedure entails finding all possible achievable outputs for the system under consideration, from the trajectory of the states of interest describing the system operation. These trajectories are convexified to obtain candidate attainable regions, which are tested against the necessary conditions and recursively updated so that any violated necessary conditions is eliminated. The process continues until no other necessary conditions are violated otherwise, a candidate AR (subset of the true AR) is obtained, which can still provide better understanding of the achievable limits of the system. It is not the intention of this article to present a detailed explanation of the AR theory. Interested readers can consult the above sited literature for a more in-depth understanding.

3.2. Application of AR approach to maximize methanogenic activity

3.2.1. Reaction scheme and process kinetics

Using the estimated kinetic constants, a stoichiometric scheme of the bioreaction occurring in the anaerobic digester consist of two main reactions catalyzed by acid-forming bacteria, Eq. (15) and methane-forming bacteria Eq. (16)

$$k_1S_1 \xrightarrow{r_{X_1}} X_1 + k_2S_2 \tag{15}$$

$$k_3S_2 \xrightarrow{r_{X_2}} X_2 + k_4CH_4 \tag{16}$$

If we assume the specific death rate to be negligible compared to the specific growth rate of both microbial populations, the rate expressions for the different reaction species is defined by Eqs. (17) – (20)

$$r_{X_1} = \mu_1X_1 \tag{17}$$

$$r_{X_2} = \mu_2X_2 \tag{18}$$

$$r_{S_1} = -k_1\mu_1X_1 \tag{19}$$

$$r_{S_2} = k_2\mu_1X_1 - k_3\mu_2X_2 \tag{20}$$

3.3. Fundamental processes

Various fundamental processes can occur within a system, which for bioreactors may include: mass transfer, mixing, bioreaction (biodegradation, bioconversion), adsorption, heat transfer, etc. The AR approach requires the fundamental processes taking place in the system be identified. The following two main fundamental processes are identified to be associated with the anaerobic treatment process: Biodegradation and mixing. The attainable region (AR) for the anaerobic treatment process therefore represents the set of all possible states that can be achieved by a combination the two fundamental processes, biodegradation and mixing. In AR theory, mixing is performed by a continuous stirred tank reactor (CSTR) while reaction (biodegradation) is achieved in a plug flow reactor (PFR), since the operation of both reactors respectively mimic the two fundamental processes. At steady state operation, the general mathematical representation of a CSTR and PFR are given by Eqs. (21) and (22) respectively.

$$C = C_f + \tau r(C) \tag{21}$$

$$\frac{dC}{d\tau} = r(C) \tag{22}$$

C is the state vector while $r(C)$ is the reaction rate vector as shown by Eqs. (23) and (24) respectively.

$$\mathbf{C} = [X_1 \quad X_2 \quad S_1 \quad S_2\,]^T \tag{23}$$

$$\mathbf{r}(\mathbf{C}) = [r_{X_1} \quad r_{X_2} \quad r_{S_1} \quad r_{S_2}\,]^T \tag{24}$$

Solving the CSTR system to obtain the roots at a given feed point (C_f) and for different residence times (τ_i *for* $i = 1$ *to* n) results in a set of points referred to as a CSTR locus. In the same way, integrating the PFR system for a given feed point and residence time results in a set of points referred to as PFR trajectory.

3.3.1. Dimensionality analysis and model reduction

The reaction stoichiometry of the system can be used to determine the dimension of the system. The dimension of the AR is determined from the number of independent reactions occurring in the reactor system, which defines the dimension of the stoichiometric subspace (the rank of the stoichiometric coefficient matrix A), in which the AR must reside. Since there are two independent reactions occurring in the system, the set of points generated by the anaerobic treatment process must reside in a two-dimensional subspace in $\mathbb{R}^5$ (Ming et al., 2016).

Before constructing the AR, the space wherein the AR must reside (by choosing unique species components in the reactions that will represent the AR) must first be determined. Methanogenesis has been known to be the most sensitive step of the anaerobic treatment process and since the volatile fatty acids and methanogenic microorganisms, are the key player in this stage, it is sensible to generate the candidate AR in $(S_2 - X_2)$ space, which provides information required to maximize gas production as well as process stability. However, even if only a subset of the states is used to construct the AR (candidate AR), it can still be transformed in terms of the other variables if required (Ming et al., 2016). The reduced state and reaction rate vectors are therefore presented by Eqs. (25) and (26).

$$\mathbf{C} = [S_2 \;\; X_2]^T \tag{25}$$

$$\mathbf{r}(\mathbf{C}) = \left[r_{S_2} \;\; r_{X_2} \right]^T \tag{26}$$

This reduction in the dimensions of the state and rate vectors is possible because the reaction rate of biodegradable volatile solids (r_{S_1}) can be expressed in terms the reaction rate of acidogenic bacteria (r_{X_1}), which can in turn be expressed as functions of reaction rates of volatile acids (r_{S_2}) and methanogenic archae (r_{X_2}) as shown by Eqs. (27) and (28):

$$r_{S_1} = -k_1 r_{X_1} \tag{27}$$

$$r_{X_1} = \frac{1}{k_2}\left(r_{S_2} + k_3 r_{X_2}\right) \tag{28}$$

This implies that S_1 can be expressed in terms of X_1, which can in turn be expressed as a function of S_2 and X_2, illustrated by Eqs. (29) and (30). Notice the presence of two new terms in Eqs. (29) and (30), $X_{1_{in}}$ and $X_{2_{in}}$, which represent the respective feed concentrations of acidogenic bacteria and methanogenic archea. These terms are absent in Eqs (1)–(5) because the material balance assumes that the concentration of anaerobic microbes in the feed is negligible compared to that inside the digester (Finn et al., 2013; Hill, 1983). So in Eqs (29) and (30), $X_1 - X_{1in} \cong X_1$ and $X_2 - X_{2in} \cong X_2$.

$$S_1 = S_{1_{in}} - k_1\left(X_1 - X_{1_{in}}\right) \tag{29}$$

$$X_1 = X_{1_{in}} + \frac{1}{k_2}\left[S_2 - S_{2_{in}} + k_3\left(X_2 - X_{2_{in}}\right)\right] \tag{30}$$

The model reduction assumes that the specific death rates of acidogens and methanogens is negligible compared to their respective specific growth rates.

3.3.2. AR construction

After stating the process kinetics, the AR construction process is initiated by defining feed point and process conditions that influence the system. In this study, three anaerobic digestion batches: diary manure, diary manure + granular sludge and diary manure + lagoon inoculum were considered each with respective feed concentrations, $C_f = [S_{2f}, X_{2f}]^T$ of $[1.89, 0.84]^T$, $[1.89, 0.84]^T$ and $[1.62, 1.53]^T$. The controlled process condition was mainly temperature, which was maintained at a constant value of $35°C$ throughout retention time. Using the specified feed, kinetics and temperature conditions, the set of points generated by solving the PFR equation are called the PFR trajectory and those generated by solving the CSTR equation are called the CSTR locus. The convex hull for the set of points generated by all possible combinations of CSTR, PFR and mixing defines the AR. The attainable region is unique for a given kinetics and feed point and process conditions. A change in any of these may result in a change in the AR and hence the operating limits of the system.

3.4. Objective function for optimizing microbial activity

Since the methanogens are most susceptible to process instabilities, we are interested in determining the optimal operating point that ensures stability of methanogenic microorganisms. For doing this, we define two objective functions, which translate the stability of methanogenic archae: The inoculum to substrate (I/S) ratio and the instantaneous yield of methanogens from volatile acids.

The inoculum to substrate ratio describes the concentration of volatile acids that should be maintained in the digester for optimal activity methanogenic archae. Studies have reported the optimal tolerance range of volatile acids, above which the methanogens experience inhibition or toxicity (Chen et al., 2008). The Instantaneous yield is defined as the rate of formation of the desired product (methanogens), divided by the rate of consumtption of the reactant (volatile acids). The inoculum to substrate ratio (I/S) was modeled using Eq. (31) while the instantaneous yield of methanogenic archae (Y_{X_2}) from volatile acids was modeled by Eq. (32).

$$IS = \frac{X_2}{S_2} \tag{31}$$

$$Y_{X_2} = \frac{r_{X_2}}{-r_{S_2}} = \frac{\mu_2 X_2}{-k_2\mu_1 X_1 + k_3\mu_2 X_2} \tag{32}$$

Eqs. (31) and (32) can be rearranged to express X_2 as a function of S_2, presented by Eqs. (33) and (34) respectively. It should be noted that the term $\mu_1 X_1$ in Eq. (34) contains X_2 and the numerical computations additionally made use of Eqs. (29) and (30).

$$X_2(S_2) = IS \times S_2 \tag{33}$$

$$X_2(S_2) = \frac{Y_{X_2} k_2 \mu_1 X_1}{\mu_2(S_2) \times \left(Y_{X_2} k_3 - 1\right)} \tag{34}$$

Eqs. (33) and (34) can seperately be plotted over the AR boundary as contours to determine the intersection with the boundary. Sections of the objective function that intersect the AR are optimal points, relative to the I/S ratio or Y_{X_2} specified. The points of intersection can be interpreted in terms of digester networks depending on the manner in which the AR is constructed (Ming et al., 2016), and the reactor structure corresponding to the I/S ratio or Y_{X_2} of interest is the optimal reactor structure.

4. Results and discussion

4.1. Model fits and estimate of kinetic constants

We have explored the capabilities of the different biokinetic models to describe the degradation of organic substrate in the anaerobic digestion process. Experimental results for three anaerobic-digestion batches of diary manure, each under different process conditions were considered (Zaher et al., 2009). In Batch 1, no external inoculum was added during start-up of the digester. In Batch 2, granular sludge is added into the digester as inoculum while in Batch 3, sludge from a lagoon was used as the inoculum. Fig. 2 presents the fitting results for all the 5 biokinetic models with experimental measurements of volatile fatty acids and methane flow rate obtained from batch 1. From the fitting results, it can be concluded that the models give a good prediction of the experimental data. However, the competitive model shows the smallest SSE (see Table 2) and can thus be considered to more closely represent the experimental data. Hence anaerobic digestion of diary manure with no external inoculum leads to methanogenic inhibition described as being competitive. Similar fittings were performed for Batches 2 and 3, which was observed that the linear model more closely represented the experimental data for both cases. Figs. 3 and 4 present the fitting for the linear model with experimental measurements of volatile acid and methane flow rate obtained from batches 2 and 3. Table 2 presents the parameter estimates for all the fitting cases. Even though batches 1 and two fit well with the linear model, the kinetic constants are different and we can thus conclude that inhibition characteristics exerted by volatile acids on methanogenic archae differs based on the conditions in the anaerobic digester. This kinetic behaviour of methanogenic archae might be explained in this way. The growth kinectis of microorganisms widely depends on nutritional availability as well as operational and environmental conditions, which in turn vary for different digester worts. The different sources of inoculum results in different wort characteristics, which can be measured in terms of nutritional differences, presence of different toxicants or other competitive microorganisms in the digester, thereby varying the kinetic behaviour of the methanogens.

The findings of the current study are consistent with those of Yang et al. (Ref) who studied the effect of temperature and substrate characteristics on kinetic behaviour of anaerobic digestion process. The authors considered four different substrates (swine wastewater, palm oil mill wastewater, protein production wastewater, synthetic wastewater and pharmaceutical wastewater), five temperature regimes ($10°C$, $15°C$, $20°C$, $25°C$, $30°C$) and four kinetic models (modified Stover-Kincannon, Chen and Hashimoto, Deng and modified Deng) were tested. It was observed that changes in substrate and temperature as well as a combination of thereof resulted in different fitting characteristics of the different kinetic models. This effect of substrate and operating conditions on the kinetic behaviour of the anaerobic treatment process has very important implications in the concept of attainable regions. This is because the attainable region is unique for a given kinetics (Ming et al., 2016) and a change in kinetics therefore results in a change in the limits of achievability by the system. What this implies practically is that the reactor structures required to achieve the optimal operating point will differ for each digested substrate, which paves the way to use the concept of attainable regions to solve operational challenges for different types of wastewaters. In addition, the study uses the adjoint method for computing gradient of the parameter estimation objective function before using the conjugate gradient method for model calibration. Gradient-based methods are widely used for calibration of anaerobic digestion models (Donoso-Bravo et al., 2011) with gradients mostly computed using the finite difference method. The adjoint method presents an alternative approach for computing gradients. Other model calibration methods that have been applied to anaerobic digestion models include the asymptotic state observers (López and Borzacconi, 2009), Simplex algorithm (Zaher et al., 2009), genetic algorithm combined with the gradient descent method (Martinez et al., 2012), etc. Even though we have presented the kinetic analysis of anaerobic digestion process using the adjoint-based gradient method, the emphasis of this paper is not necessarily on the fitting performance of the different biokinetic models, but on how we use the models to develop new policies for operation of anaerobic digesters to ensure stability of methanogenic archae.

4.2. *Effects of process kinetics on optimal reactor configuration*

4.2.1. *Attainable regions: limits of achievability by the system*

Fig. 5 presents the PFR trajectory and CSTR locus (dubed base trajectories) for the anaerobic digestion process in batch 1 while Fig. 6 presents the two-dimensional candidate AR obained from the based trajectories, using the specified feed and kinetics. Employing a CSTR gives a maximum attainable methanogenic concentration of 5.01gme/L (Fig. 5). A PFR however improves upon this concentration to an attainable value of 24.09gme/L. At this point, the reader can already notice that by using an anaerobic PFR as opposed to a CSTR, the concentration of methangoneic archae in the biodigester increases by approximately 5 times. It can be observed from Fig. 6 that constructing the attainable region further extends the concentration of methanogenic archae to 37.51 gme/L. This increase in the concentration of methanogenic archae for the same feed and kinetics is attributed to the fact that a systematic manipulation of the fundamental processs (mixing and reaction in this case) occurring in a system serves to expand the states that can be achieved by a system, which is one of the key strengths of the attainable region theory. If more fundamental processes are considered (e.g seperation), the limits of achievabtily by the states of the system can be further improved. Fig. 7 presents the base trajectories for the anaerobic digestion process in batch 2 while Fig. 8 presents the candidate AR. Recall that the AR is specific for a given kinetics, which explains why the nature of AR for batch 2 is different from that in batch 1. We oberve from the base trajectories that using a CSTR will result in higher concentrations of volatile fatty acids in the digester (an indication of process instability), while a PFR presents a maximum limit of volatile acid concentration that can be attained. Unlike the case of batch 1, constructing the AR doesn't serve to increase the maximum concentration of methanogenic archae attained (compared to the that attained with the base trajectories). However, in this case, the AR analysis shows that higher concentrations of methanogenic arhae can be attained at higher concentrations of volatile fatty acids by running a PFR from a CSTR and a bypass valve from feed (see mixing line AB on Fig. 8). Practically, this implies that using a digester network as opposed to a single digester results in an increased stability of the methananogenic archae. Fig. 9 presents the base trajectories for the anaerobic digestion process in batch 3 while Fig. 10 presents the candidate AR. Similar to the case of batch 2, constructing the AR doesn't serve to increase the maximum concentration of methanogenic archae attained but concentrations of methanogenic archae origninally not attainable at higher concentrations of volatile acids now become attainable by using a digester structure indicated by line BC of Fig. 10.

Even though we have observed a change in the nature of attainable region for the different kinetics, the boundary of ARs however have a simple fundamental structure irrespective of the kinetics used. This boundary is composed entirely of mixing surfaces (straight lines) and manifolds convex reaction surfaces (Ming et al., 2016). The points that form the convex reaction surfaces arise from effluent concentrations of the PFR trajectories, which are initiated by points from specialized CSTRs. We will now illustrate how interpret the AR boundary into anaerobic digester strucures by using the fundamental characteristics of the AR boundary. The illustration will be done by using the AR for batch 1 presented in Fig. 6. In Fig. 6, the point B is the feed point, while the region defined by ABC is the AR. The convex segment BA are trajectories obtained by running PFR from points on CSTR locus. The point A is therefore obtained by running a CSTR from point B followed by a PFR from CSTR while the point C is obtained by running a CSTR from feed (point B). The lines AC and BC are the mixing surfaces while AB is the reaction surface. Concentrations along the line AC (C_{AC}) can

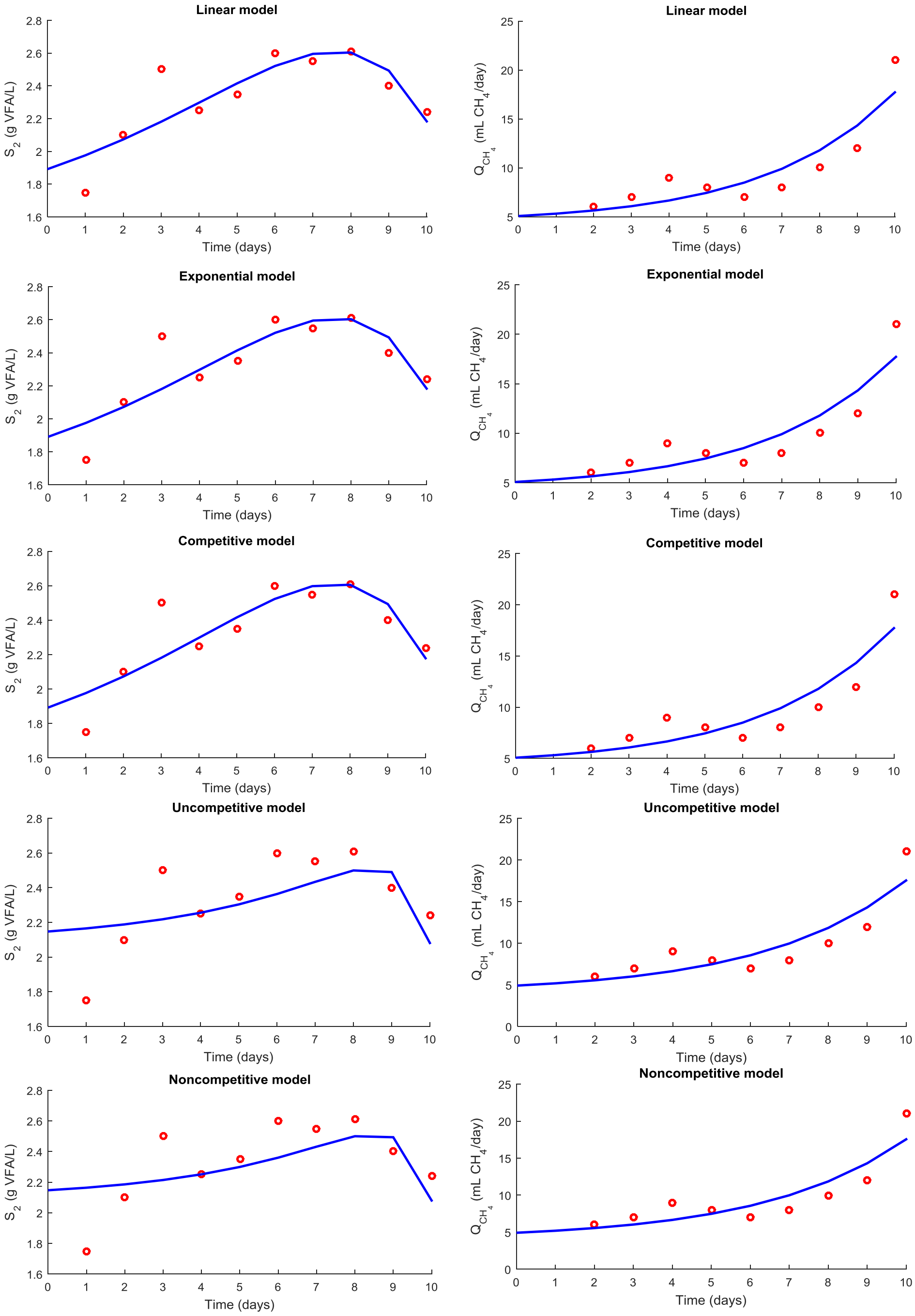

Fig. 2. Fitting of biokinetic models to experimental measurements from Batch 1. Columns one and two show the fitting of the volatile fatty acids, (S_2) and methane flow rate (Q_{CH_4}) with experimental measurements respectively while the rows show the performance of each biokinetic model (linear, exponential, competitive, non-competitve and uncompetitive).

Table 2
A generalized table for model parameters and fitting characteristics.

Model	Model parameters							SSE
	k_1	k_2	k_3	k_4	K_{s1}	K_{s2}	K_i	
Batch 1: Diary manure								
Linear	1.996e-04	3.515	4.915	1.117	0.779	0.278e-4	0.136e-04	5.67130
Exponential	1.999e-04	3.515	4.915	1.116	0.778	0.278e-4	0.135e-04	5.67129
Competitive	0.0011	9.376	13.712	1.118	0.278	0.056e-4	32.717	5.67124
Noncompetitive	0.421	7.754	11.581	1.3514	2.486	0.0215	45.338	5.86042
Uncompetitive	0.406	7.760	11.581	1.3499	2.487	0.0215	39.338	5.85868
Batch 2: Diary manure + granular sludge								
Linear	14.026	1.152	4.71e-7	112.995	0.061	28.432	61.065	14.1601
Exponential	7.294	0.742	14.501	97.558	1.498	31.401	45.438	43.3498
Competitive	9.1887	1.66e-6	56.367	53.318	26.965	1.1545	76.762	39.4748
Noncompetitive	1.1125	2.8272	60.143	2.73e-6	1.6375	6.8891	2.279	39.4221
Uncompetitive	4.1808	8.9413	23.552	7.66e-6	7.0412	0.8130	0.2758	39.4221
Batch 3: Diary manure + Lagoon innoculum								
Linear	9.3970	0.3433	0.5855	384.557	0.5103	2.3350	0.3960	10.7005
Exponential	12.0561	0.0004	0.8396	9.5819	4.9221	2.4868	0.0266	30.4303
Competitive	14.9995	1.0976	2.5811	15.3191	0.0024	0.0026	5.5585	30.2694
Noncompetitive	12.8286	2.9529	31.873	0.2078	271.81	32.576	0.0011	30.5584
Uncompetitive	12.869	4.465	13.474	56.4751	422.73	53.154	7.148e-8	30.5584

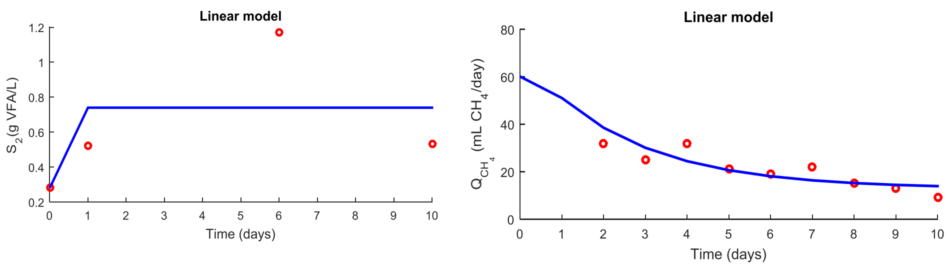

Fig. 3. Fitting of linear model to experimental measurements from Batch 2.

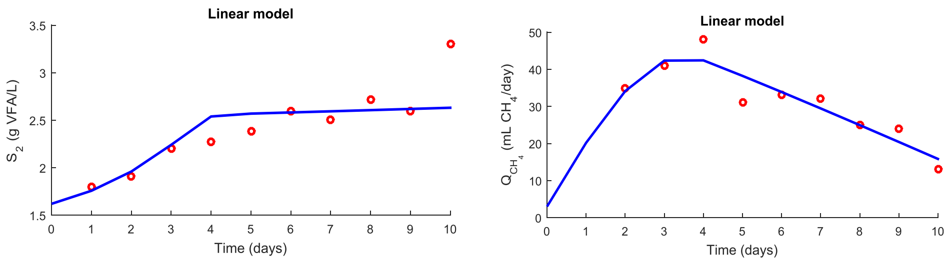

Fig. 4. Fitting of linear model to experimental measurements from Batch 3.

be obtained by mixing points A and C, Eq. (35) (the lever-arm rule) and the digester structure is therefore given by a CSTR + PFR (point A) run in parallel with a CSTR (point C) and both contents mixed at the end. Concentrations on the line BC (C_{BC}) can be obtained by mixing points B and C, Eq. (36) and the required digester structure is given by a CSTR (point C) with a bypass from feed (point B). Similar digester interpretations were made for batches 2 and 3 as displayed on Figs. 8 and 10.

$$C_{AC} = \alpha C_A + (1 - \alpha) C_C\ , \qquad 0 \le \alpha \le 1 \tag{35}$$

$$C_{BC} = \alpha C_B + (1 - \alpha) C_C, \qquad 0 \le \alpha \le 1 \tag{36}$$

Where α is known as the mixing ratio.

The results obtained imply practically that a systematic scheduling of the fundamental processes of mixing and reaction occurring in the anaerobic digester can result in an increased stability of methanogenic archae. It is interesting to note that for a two-dimensional attainable region, when mixing and reaction are the only fundamental processes occurring in a system, the AR may be constructed by a combination of reactors involving PFRs, CSTRs and mixing only (Ming et al., 2016). What this means is that there is no need to devise new or perhaps novel types of digesters with the aim of extending the limits of achievability by the system. Instead, it is required to focus attention on optiimally arranging combinations of these two fundamental digester types or researching more fundamental processes to the system.

Table 3 presents a summary of the performance characteristics

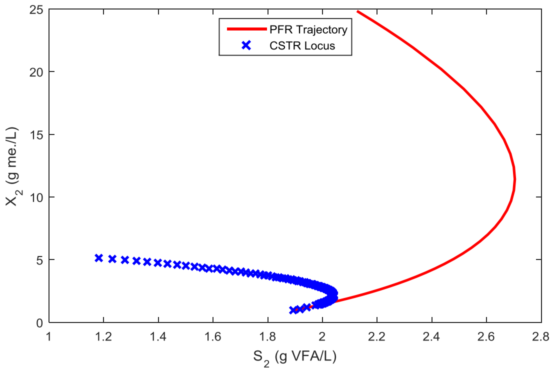

Fig. 5. Anaerobic base trajectories for digestion process in batch 1.

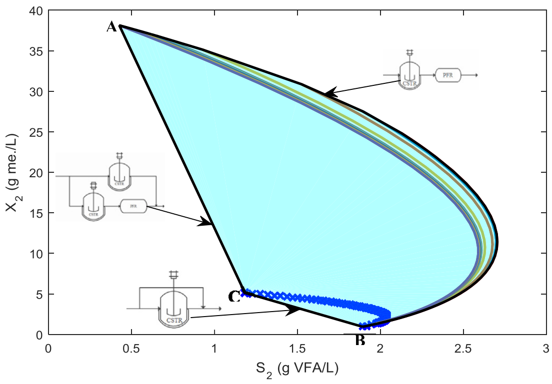

Fig. 6. Two-dimensional attainable region for anaerobic digestion process in batch 1.

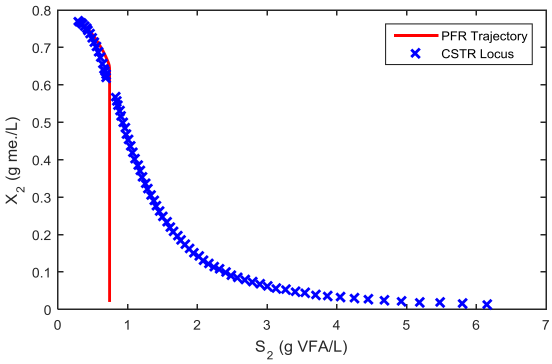

Fig. 7. Anaerobic base trajectories for digestion process in batch 2.

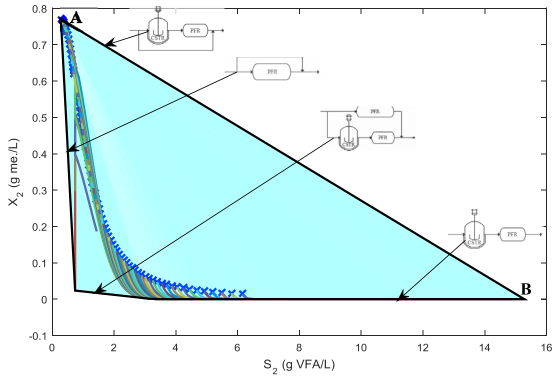

Fig. 8. Two-dimensional attainable region for anaerobic digestion process in batch 2.

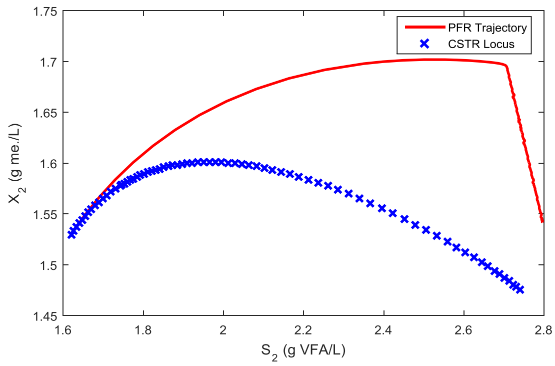

Fig. 9. Anaerobic base trajectories for digestion process in batch 3.

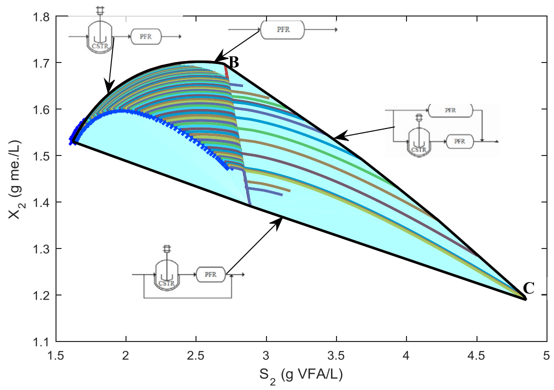

Fig. 10. Two-dimensional attainable region for anaerobic digestion process in batch 3.

of limits of achievability of the three batches of anaerobic digestion. The limits of achievability by the systems have been measured quantitatively in terms of the area of the convex hull. Note that the AR is defined by the convex hull of the set of points (states) generated by the fundamental processes occurring within the system. The convex hull represents the smallest subset of a set of points that can be used to generate all other points by reaction and mixing. Geometrically, a convex hull is a finite convex polytope enclosed by a finite number of hyperplanes, which is interpreted in a two-dimensional space as the smallest polygon enclosed by planar facets such that all of the elements lie on or in the interior of the polygon (Asiedu et al., 2015).

Table 3
Performance characteristics of the limits of achievability by the batches.

Batch	Digested condition	Area of convex hall (g/L)^2		Performance improvement
		Base trajectory	Attainable region	
1	Diary manure only	21.265	46.486	118.604%
2	Diary manure + granular sludge	2.018	5.562	175.627%
3	Diary manure + lagoon innoculum	0.183	0.551	200.436%

From the results in Table 3, The following two conclusions can be made. (1) The AR analysis serves to improve the peformance of the system (measured in thems of states attained by the concenertation of methanogenic archae) for all the three batches. (2) We observe that the % increase in peformance differs for each batch of anaerobic digestion. This is because a change in digester characterisitics (source of innoculum) results in a change in kinetics and the ability of the AR to improve the peformance of the system depends on the process kinetics. We can conclude that even if all the necessary conditions of the AR are not met, the candidate (otherwise true) AR still serves to improve the limits of achievability by the system. In the next section, we will present how the AR has been used to answer few design questions on the anaerobic digesters.

4.2.2. Digester structures for optimal methanogenic activity

The optimal digester structures for the different batches of anaerobic digestion have been obtained from the point of intersection between the objective function and boundary of the AR. Fig. 11 shows a number of contour lines for I/S ratio and the instantaneous methanogenic yield for batch 1. Recall that in the case of batch one, the competitive inhibition model was selected for use in modeling the digester configuration using attainable regions. So for optimizing the instantaneous yield, we substitute $\mu_2(S_2)$ corresponding to competitive inhibition model (see Table 1) into Eq. (34) to obtain objective function for competitive model, Eq. (37). Since the term $\mu_1 X_1$ in Eq. (37) contains X_2, the numerical computations additionally made use of Eqs. (29) and (30) in order to overlay Eq. (37) onto the AR boundary constructed in the $(S_2 - X_2)$ space.

$$X_2 = \frac{Y_{X_2} k_2 \mu_1 X_1 \left[K_{s2}\left(1+\frac{S_2}{K_i}\right)+S_2\right]}{\mu_{m2} S_2 (Y_{X_2} k_3 - 1)} \tag{37}$$

Observe that the two objective functions intersect the AR boundary at several points. The I/S ratio becomes smaller while the instantaneous methanogenic yield becomes larger as we move toward the horizontal line $X_2 = 0$. This suggests that for a given concentration of methanogens in the digester, higher I/S ratio corresponds to lower concentration of volatile acids while the instantaneous methanogenic yield corresponds to higher concentration of volatile acids.

Fig. 12 shows a number of contour lines for I/S ratio and instantaneous yield for batch 2. In the case of batch 2, the linear inhibition model was selected to describe the anaerobic digestion kinetics. So for optimizing the instantaneous yield, we substitute $\mu_2(S_2)$ corresponding to linear inhibition model (see Table 1) into Eq. (36) to obtain Objective function for the linear inhibition model (Eq. (38)) as in the case for batch 1.

$$X_2 = \frac{Y_{X_2} k_2 \mu_1 X_1 (K_{s2} + S_2)}{\mu_{m2} S_2 (1 - k_i S_2)(Y_{X_2} k_3 - 1)} \tag{38}$$

Similarly to the case of batch 1, the two objective functions intersect the AR boundary at several points and the I/S ratio becomes smaller w as we move toward the horizontal line $X_2 = 0$. However, contrarily to batch 1, the instantaneous methanogenic yield becomes smaller as we move toward the horizontal line $X_2 = 0$. This suggests that for a given concentration of methanogens in the digester, higher I/S ratio and higher instantaneous methanogenic yields corresponds to lower concentration of volatile acids. A possible explanation for the reversal of trend observed in instantaneous methanogenic yield can be attributed to the fact that the range of concentrations of volatile acids attained in batch 2 fall within the inhibitory range there by causing inhibition to the growth of methanogenic archae. Another possible explanation could be that the granular sludge used as inoculum for batch 2 is less adapted to higher concentrations of volatile acids and studies have confirmed that acclimation or adaptation of methanogens greatly influence their ability to withstand higher concentrations of inhibitory substances (Asiedu et al., 2015; Chen et al., 2008, 2014).

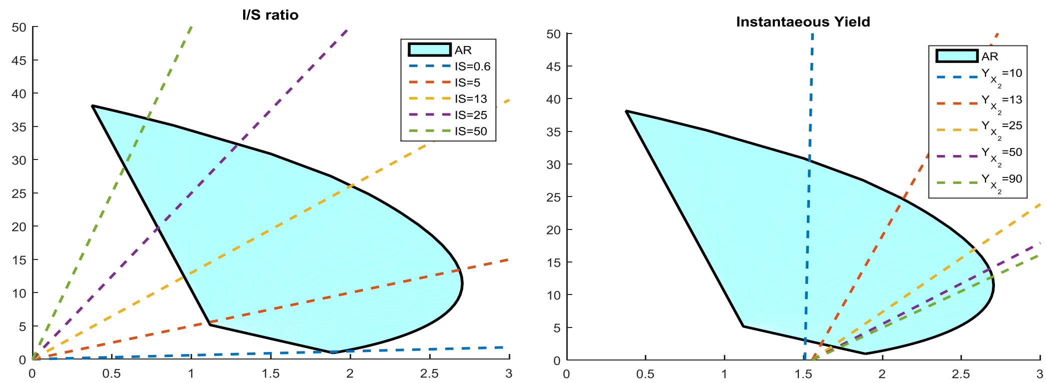

Fig. 11. Contour lines for I/S ratio and instantaneous methanogenic yield for batch 1.

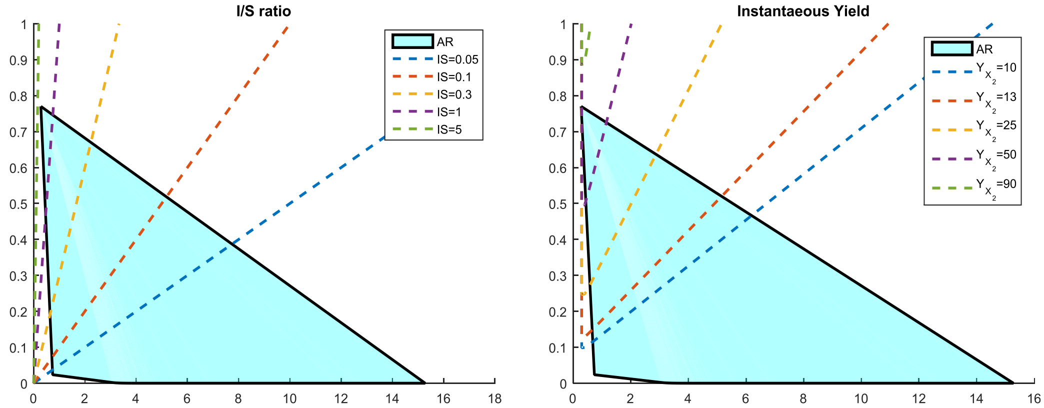

Fig. 12. Contour lines for I/S ratio and instantaneous methanogenic yield for batch 2.

Fig. 13 shows a number of contour lines for I/S ratio and instantaneous yield for batch 3. Contrarily to batches 1 and 2, for some concentrations of methanogenic archae in the digester, certain values of instantaneous methanogenic yield correspond to two different concentrations of volatile acids within the limits of achievability by the system.

Multiple points of intersection between objective functions and boundary of the attainable region is an indication of multiple operating points (multiple optima) for the system. If the only criteria for design is the I/S ratio and the instantaneous methanogenic yield, then the optimal digester structure to achieve a given I/S ratio or instantaneous methanogenic yield can be seelected from any of the intersection points. However, points corresponding to lower concentrations of methanogenic yield (points associated with the lower part of the AR) are preferable since the growth rate of these microbial population is about 5 times slower that of acidogens (Henze et al., 2008) hence making it difficult to maitain higher concentrations in the digester unless a seperation system is included.

It is interesting to compare the results of this study with that of Abunde et al. (Abunde Neba et al., 2019), who used attainable regions to compare the limits of achievability of five different digested substrates using volumetric methane productivity and waste stabilization as objective functions. The authors concluded that a change in the type of digested substrate results in a change in the limits of ahievability as well as the optimized AR parameter of an anaerobic digestion system. In this study, the results have demonstrated that for the same digested substrate (diary manure), different sourcess of inoculum will result in different limits of achievability by system and hence the optimal digester structure (using I/S ratio and instantaneous methanogenic yield as objective functions. The results have shown that using digester structues as opposed to single digesters can improve the viability of methanogenic archae at higher concentrations of volatile fatty and for an anaerobic digestion system, a change in digested subtrate and/or source of innoculum results in a change in the limits of achievabilty by the system. This study therefore lays down the theoretical framework for using attainable regions to define the anaerobic digetion performance targets for a given innoculum and/or substrate characteristics. Therefore, unlike the BMP assay and the Buxuells technique for defining performance targets (limits of achievability), the AR approach does not only provide information about the limits of achievabily, but it provides the optimal digester structures required to achieve a given target.

In our previous study (Abunde Neba et al., 2019), we considered measurable outputs from the digester (volumetric methane

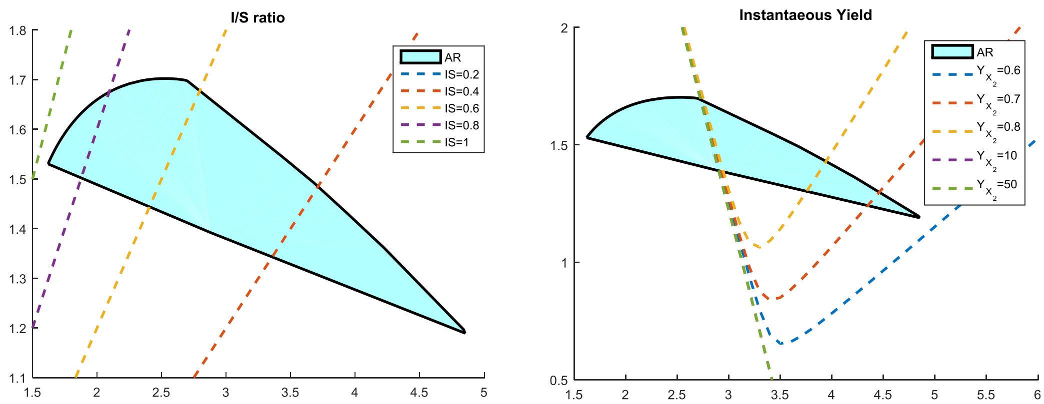

Fig. 13. Contour lines for I/S ratio and instantaneous methanogenic yield for batch 3.

productivity and waste stabilization) as objectives, while the current study we considered parameters directly linked to microbial stability (I/S ratio and instantaneous methanogenic yield) as objectives. The combination of results suggest that the AR can be used to answer any design and optimization questions. This is possible because for a defined kinetics (model structure and/or kinetic coefficients), the AR is fixed for a given feed point and multiple objective functions (and hence multiple optimizations) may be formed using a single AR. In other words, the attainable region represents the solution to several different optimization problems implying several optimization scenarios can be performed without any requirement to perform further optimizations (or reconstruct the attainable region) when the objective function is changed. Therefore the design approach presented in this study can be used to optimize any process and design parameter of the anaerobic treatment processes.

5. Conclusion

Returning to the problems posed at the beginning of this study, it is now possible to state that the use of digester structues as opposed to single digesters can improve process stability and performance. For a given concentration of volatile acids in an anaerobic digester, higher concentrations of methanogenic archae can be attained using a digester structure (network) as opposed to single digester. This study has shown that a for a given digested substrate, a change in the source of innoculum results in a change in the limits of achievabilty by the system and hence the optimal digester structures required to achieve a given objective. Another major finding was that the attainable region technique can be used as reliable alternatives to the BMP assay and the Buxuells technique for defining performance targets (limits of achievability), because the AR approach does not only provide information about the limits of achievabily, but it provides the optimal digester structures required to achieve a given target.

The design technique presented in this study can be used to answer any design and optimization questions regarding the anaerobic treatment process. This concept has been proven by formulating and solving two optimization problems to obtain optimal structures for anaerobic digesters to achieve a given inoculum to substrate ratio as well as instantaneous methanogenic yield. The evidence from this study suggests that the technique of using digester structures presents a break through in the application of low-rate anaerobic digesters as it can be used to improve upon the process dynamics. The current findings add to a growing body of literature on the application of attainable regions for solving operational challenges in processs engineering.

These findings enhance our understanding of that a systematic manipulation of the fundamental processes (mixing and reaction in this case) occurring in a system serves to expand the states that can be achieved by a system, which is one of the key strengths of the attainable region theory. It is highly interesting for readers to note the geometric optimization technique presented in this study can also be to optimize operation of other wastewater treatment processes (e.g., activated sludge treatment, coagulation, etc.). The technique is suitable not because of multiple reactors, but because of multiple reactions in a process.

In order to subject the operational technique to actual experimental verification, pilot scale studies are currently under design. In the mean time, interested researchers could consider using economic indicators such as net present value, internal rate of returns, benefit cost ratio or payback period as objective function for attainable region optimization, which would present a key motivation for investors.

Further theoretical study is needed to account for the effect of temperature regimes (pychrophillic, mesophillic and thermophillic) on the limits of avhievabilit by the system. In this study, the anaerobic digester networks have been staged two-stage biochemical kinetics in which acid-forming stage is physically separated from the methane gas-forming stage. Other studies could consider applying thermodynamic staging techniques where digester networks are designed based on different temperature regimes in order to take advantage of the higher stability of the mesophilic digestion as well as the higher digestion rate of thermophilic digestion.

Acknowledgments

Our team expresses gratitude to the following institutions; The Brew-Hammond Energy Centre, KNUST Ghana, The Water and Environmental Engineering Group, NTNU Ålesund and the Abunde Sustainable Engineering Group (AbundeSEG) for its immense technical support.

Nomenclature

Y_{X_2}	Instantaneous methanogenic yield
A_f	Acidity factor $(g\ VFA/L)/(g\ BVS/L)$
B_0	Biodegradability constant $(g\ BVS/L)/(g\ VS/L)$
K_{s_1}	Monod half-saturation constant for acidogenic bacteria $(g\ BVS/L)$
K_{s_2}	Monod half-saturation constant for acidogenic bacteria $(g\ VFA/L)$
K_{d1}	Specific death rate of acidogenic bacteria (d^{-1})
K_{d2}	Specific death rate of methanogenic archae (d^{-1})
K_i	VFA inhibition constant for methanogenic archae $(g\ VFA/L)$
Q_{CH_4}	Volumetric methane flowrate $(mL\ CH_4/d)$
$S_{1_{in}}$	Input concentration of biodegradable volatile solids $(g\ BVS/L)$
$S_{2_{in}}$	Initial concentration of volatile fatty acids $(g\ VFA/L)$
S_2	Concentration of biodegradable volatile solids $(g\ VFA/L)$
S_2	Concentration of volatile fatty acids in bioreactor $(g\ VFA/L)$
S_{in}	Input concentration of volatile solids $(g\ VS/L)$
VFA_{in}	Inlet concentration of volatile fatty acids $(g\ VFA/L)$
$X_{1_{in}}$	Initial concentration of acidogenic bacteria $(g\ ac./L)$
$X_{2_{in}}$	Initial concentration of methanogenic archae $(g\ me./L)$
X_1	Concentration of acidogenic bacteria in bioreactor $(g\ ac./L)$
X_2	Concentration of methanogenic archae in bioreactor $(g\ me./L)$
k_1	Yield constant $(g\ BVS/g\ ac./L)$
k_2	Yield constant $(g\ VFA/g\ ac./L)$
k_3	Yield constant $(g\ VFA/g\ me./L)$
r_{S_1}	Reaction rate for biodegradable volatile solids $(g\ BVS/L/d)$
r_{S_2}	Reaction rate for volatile fatty acids $(g\ VFA/L/d)$
r_{X_1}	Reaction rate for acidogenic bacteria $(g\ ac./L/d)$
r_{X_2}	Reaction rate for methanogenic archae $(g\ me./L/d)$
μ_{m_1}	Maximum specific growth rate of acidogenic bacteria (d^{-1})
μ_{m_2}	Maximum specific growth rate of methanogenic archae (d^{-1})
μ_1	Specific growth rate of acidogenic bacteria (d^{-1})
μ_2	Specific growth rate of methanogenic archae (d^{-1})
AD	Anaerobic digestion
AR	Attainable Regions
CSTR	Continuous Stirred Tank Reactor
DSR	Differential Side-stream reactor

I/S	Inoculum to substrate ratio
PFR	Plug Flow Reactor
T	Reactor temperature ($^{\circ}C$)
V	Volume of digester (L)
VSL	Volatile Solids Load ($g\ BVS\ /L$)
α	Mixing ratio
τ	Residence time (days)
ϑ	Acidogenic fraction

Funding

This work was supported by EnPe-NORAD under the project Upgrading Education and Research Capacity in Renewable Energy Technologies (UPERC-RET).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

Abunde Neba, F., Asiedu, N.Y., Addo, A., Morken, J., Østerhus, S.W., Seidu, R., 2019. Use of attainable regions for synthesis and optimization of multistage anaerobic digesters. Appl. Energy 242, 334–350.
Akobi, C., Yeo, H., Hafez, H., Nakhla, G., 2016. Single-stage and two-stage anaerobic digestion of extruded lignocellulosic biomass. Appl. Energy 184, 548–559.
Alford, J.S., 2006. Bioprocess control: advances and challenges. Comput. Chem. Eng. 30, 1464–1475.
Asiedu, N., Hildebrandt, D., Glasser, D., 2015. Experimental simulation of three-dimensional attainable region for the synthesis of exothermic reversible reaction: ethyl acetate synthesis case study. Ind. Eng. Chem. Res. 54, 2619–2626.
Benítez, M., Bermúdez, A., Rodríguez-Calo, J.F., 2017. Adjoint method for parameter identification problems in models of stirred tank chemical reactors. Chem. Eng. Res. Des. 123, 214–229.
Bernard, O., Hadj-Sadok, Z., Dochain, D., Genovesi, A., Steyer, J.-P., 2001. Dynamical model development and parameter identification for an anaerobic wastewater treatment process. Biotechnol. Bioeng. 75, 424–438.
Chen, J.L., Ortiz, R., Steele, T.W.J., Stuckey, D.C., 2014. Toxicants inhibiting anaerobic digestion: a review. Biotechnol. Adv. 32, 1523–1534.
Chen, Y., Cheng, J.J., Creamer, K.S., 2008. Inhibition of anaerobic digestion process: a review. Bioresour. Technol. 99, 4044–4064.
Demirel, B., Yenigun, O., 2002. Two-phase anaerobic digestion processes: areview. J. Chem. Technol. Biotechnol. 77, 743–755.
Donoso-Bravo, A., Mailier, J., Martin, C., Rodríguez, J., ACEVES-Lara, C.A., Wouwer, A.V., 2011. Model selection, identification and validation in anaerobic digestion: a review. Water Res. 45, 5347–5364.
Epa, U.S.E.P.A., 2006. Biosolids Technology Fact Sheet, Multi-Stage Anaerobic Digestion. National Service Center or Environmental Publications (NSCEP).
Finn, H., Rune, B., Bernt, L., 2013. Adapting dynamic mathematical models to a pilot anaerobic digestion reactor. Model. Identif. Control 34, 35–54.
Glasser, D., Crowe, C., Hildebrandt, D., 1987. A geometric approach to steady flow reactors: the attainable region and optimization in concentration space. Ind. Eng. Chem. Res. 26, 1803–1810.
Glasser, D., Hildebrandt, D., Godorr, S., Jobson, M., 1993. A geometric approach to variational optimization: finding the attainable region. Ifac Proc. Vol. 26, 451–457.
Henze, M., Van Loosdrecht, M.C.M., Ekama, G.A., Brdjanovic, D., 2008. Biological Wastewater Treatment. Iwa Publishing.
Hildebrandt, D., Glasser, D., 1990. The attainable region and optimal reactor structures. Chem. Eng. Sci. 45, 2161–2168.
Hildebrandt, D., Glasser, D., Crowe, C.M., 1990. Geometry of the attainable region generated by reaction and mixing: with and without constraints. Ind. Eng. Chem. Res. 29, 49–58.
Hill, D.T., 1983. Simplified monod kinetics of methane fermentation of animal wastes. Agric. Wastes 5, 1–16.
Jingura, R.M., Kamusoko, R., 2017. Methods for determination of biomethane potential of feedstocks: a review. Biofuel Res. J. 4, 573–586.
Kythreotou, N., Florides, G., Tassou, S.A., 2014. A review of simple to scientific models for anaerobic digestion. Renew. Energy 71, 701–714.
López, I., Borzacconi, L., 2009. Modelling a full scale Uasb reactor using a Cod global balance approach and state observers. Chem. Eng. J. 146, 1–5.
Martinez, E., Marcos, A., AL-Kassir, A., Jaramillo, M.A., Mohamad, A.A., 2012. Mathematical model of a laboratory-scale plant for slaughterhouse effluents biodigestion for biogas production. Appl. Energy 95, 210–219.
Mes, T.Z.D.D., Stams, A.J.M., Reith, J.H., Zeeman, G., 2003. Methane Production by Anaerobic Digestion of Wastewater and Solid Wastes. Bio-Methane and Bio-Hydrogen : Status and Perspectives of Biological Methane and Hydrogen Production. Dutch Biological Hydrogen Foundation - NOVEM, The Hague, The Netherlands.
Ming, D., Glasser, D., Hildebrandt, D., Glasser, B., Metzer, M., 2016. Attainable Region Theory: an Introduction to Choosing an Optimal Reactor. John Wiley & Sons, Inc, Hoboken, New Jersey.
Nasr, N., Elbeshbishy, E., Hafez, H., Nakhla, G., Hesham El Naggar, M., 2012. Comparative assessment of single-stage and two-stage anaerobic digestion for the treatment of thin stillage. Bioresour. Technol. 111, 122–126.
Zaher, U., Pandey, P., Chen, S., 2009. A simple elemental continuity based model application to study the anaerobic microbial activity for the treatment of dairy manure. Appl. Math. Model. 33, 3553–3564.
Zhang, J., Loh, K.-C., Li, W., Lim, J.W., Dai, Y., Tong, Y.W., 2017. Three-stage anaerobic digester for food waste. Appl. Energy 194, 287–295.

Paper 3:

Biodigester rapid analysis and design system (B-RADeS): A candidate attainable region-based simulator for the synthesis of biogas reactor structures

A paper published in ***Computers & Chemical Engineering***

Citation:

ABUNDE NEBA, F., ASIEDU, N. Y., ADDO, A., MORKEN, J., ØSTERHUS, S. W. & SEIDU, R. 2020. Biodigester rapid analysis and design system (B-RADeS): A candidate attainable region-based simulator for the synthesis of biogas reactor structures. ***Computers & Chemical Engineering*,** 132, 106607.

Highlights:

- The concept of attainable regions is used to couple biokinetics and macroeconomic parameters and digester network synthesis
- Ten microbial growth models are adapted in to one-stage kinetic models for synthesis of anaerobic digesters
- An economic feasibility model involving payback period is developed as the design objectives
- A software is developed to implement the framework and tested on three substrates: swine wastewater, pharmaceutical wastewater and palm oil mill effluent
- The choice of macroeconomic parameter and type of feedstock influences the optimal digester structure
- The framework is useful in cases of data limitations as only experimental measurements of biogas yield are required for synthesis

Computers and Chemical Engineering 132 (2020) 106607

Contents lists available at ScienceDirect

Computers and Chemical Engineering

journal homepage: www.elsevier.com/locate/compchemeng

Biodigester rapid analysis and design system (B-RADeS): A candidate attainable region-based simulator for the synthesis of biogas reactor structures

F. Abunde Neba[a,d,e,f,*], Nana Y. Asiedu[c], Ahmad Addo[b], John Morken[g], Stein W. Østerhus[d], Razak Seidu[e]

[a] Abunde Sustainable Engineering Group (AbundeSEG), Kumasi, Ghana
[b] Department of Agricultural and Biosystems Engineering, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana
[c] Department of Chemical Engineering, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana
[d] Department of Civil and Environmental Engineering, Norwegian University of Science and Technology, Trondheim, Norway
[e] Institute for Marine Operations and Civil Engineering, Norwegian University of Science and Technology, Ålesund, Norway
[f] Department of Mathematics, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana
[g] Faculty of Science and Technology, Norwegian University of Life Sciences, Drobakveien 31, 1432 Aas, Ås, Norway

ARTICLE INFO

Article history:
Received 17 April 2019
Revised 10 September 2019
Accepted 16 October 2019
Available online 19 October 2019

Keywords:
Attainable regions
Kinetic models
Macroeconomic parameters
Process economics
Biodigester design

ABSTRACT

Anaerobic digesters are seldom designed based on process kinetics, but rather on a combination of hydraulic and organic loading, which may limit operational performance. This study focuses on the incorporation of process kinetics in the design of anaerobic digesters, within the attainable region conceptual framework. Candidate attainable regions for anaerobic digesters are identified using the software environment Biodigester Rapid Analysis and Design System (B-RADeS), which couples, biodegradation kinetics as well as economic parameters for the synthesis of biodigester structures. By considering swine, palm oil and pharmaceutical wastewaters, payback periods of 0.5, 1 & 2 years, and substrate, kinetic model and/or economic parameters, a promising digester structure (and associated hydraulic retention times) is synthesized, consisting of a CSTR followed by PFR (15 days), CSTR (4.8 hours) and a PFR with bypass of feed (3 days). The framework offers great promise for widespread practical application.

1. Introduction

Anaerobic digestion of solid waste and/or wastewater sludges has long been used for stabilization of these wastes prior to disposal. Among the benefits involved in anaerobic waste treatment compared to the aerobic counterpart are: improved dewaterability of the treated waste and generation of renewable bioenergy (Mes et al., 2003). The construction and operation of anaerobic treatment plants requires optimizing the techno-economic feasibility by defining optimal process configuration of anaerobic digesters. There exist several types of anaerobic digesters each of which have specific characteristics making them more adequate to treat specific types of organic wastes (Mao et al., 2015). For treatment of solid waste and sludges, low-rate anaerobic systems are more appropriate due to their use of long but coupled hydraulic and sludge retention times to ensure a stable operation of the process (Mes et al., 2003). On the other hand, a major breakthrough in the anaerobic treatment technology has been the development of high-rate systems, which use biomass retention to employ shorter hydraulic retention times, but this technology is mainly adapted to the treatment of wastewaters (Henze et al., 2008). Design of high-rate systems for wastewater treatment has received considerable attention over the past years and a variety of novel or improved digester designs and hydrodynamic configurations have been proposed in the literature (Zhang et al., 2016; Mao et al., 2015). The motivation for designing novel digester configurations has been to increase process stability, simplify construction and operation as well as improve process economics. However, few researchers have been able to draw on any systematic research into the improved design and operation strategies of low rate anaerobic reactors and the use of long process times (which is linked to economic feasibility of a system) remains a challenge to such systems. Both efficient and economical performances of low-rate digesters are extremely important to promote their widespread adoption for treatment of

* Corresponding author.
E-mail address: fabricen@stud.ntnu.no (F. Abunde Neba).

https://doi.org/10.1016/j.compchemeng.2019.106607
0098-1354/

Nomenclature

B_t	Annual savings from electricity consumption ($)
C_{Gen}	Cost of biogas electricity generator ($)
C_{Inv}	Investment cost ($)
C_{con}	Cost of digester construction ($)
C_m	Annual cost of digester maintenance ($)
C_{misc}	Cost associated to miscellaneous activities ($)
C_{pf}	Annual cost of biogas purification ($)
C_t	Annual operating cost ($)
K_*	Kinetic constant of Chen and Hashimoto based on specific methane production rate $(-)$
K_{ha}	Maximum rate of substrate degradation by the acidogenic bacteria (day^{-1})
K_{Lr}	Monod-like half saturation constant for continuous mode operation $(gVS/L/day)$
K_{am}	Maximum rate of substrate utilization by the acetogenic/methanogenic microorganisms
K_i	Inhibition constant for biogas yield (g/L)
K_s	Monod-like half saturation constant for continuous mode operation (gVS/L)
L_r	Organic loading rate $(gVS/L/day)$
P_{el}	Percentage of methane utilized for electricity generation
Pr_{pf}	Price for biogas purification per unit volume $(\$/m^3)$
P_t	Net annual benefit ($)
R^2	coefficient of determination
R^2Adj	Adjusted coefficient of determination
R_f	Recalcitrant fraction of initial volatile substrate that is non-biodegradable.
R_p	Specific rate of biogas production by acetoclastic methanogens $(mL_{biogas}/gVS/day)$
R_{pm}	Maximum specific rate of biogas production by acetoclastic methanogens $(mL_b/gVS/day)$
S_i	Initial concentration of substrate taken up by acetogenic/methnogenic micororgansims (g/L)
S_o	Initial substrate concentration (gVS/L)
S_u	Concentration of acidified substrate produced by acidogenic bacteria (g/L)
T_{el}	Annual time period for use of electricity (d)
T_{pr}	Annual time period for biogas purification (d)
V_D	Volume of digester (mL)
X^TX	Characteristic matrix
X_{am}	concentration of acetogenic/methanogenic microorganisms
Y_{PS}	Biogas yield coefficient $(mL_{biogas}/gVS)/(gVS_{utilized}/L)$
Y_{XS}	Cell yield coefficient
b_e	Unit conversion coefficient (kWh/m^3CH4)
dy_t/dt	Rate of change in biogas yield
$-dS/dt$	Rate of decrease in the concentration of hydrolyzed substrate $(gVS/L/day)$
k_i	Inhibition constant for cell growth(g/L)
$k_{n(s)}$	Rate of substrate degradation by acidogenic bacteria $(g/g/day)$
k_s	Monods' half saturaion contant for substrate uptake
m_{VS}	Mass of volatile solids added into the digester (gVS)
n_t	Project lifespan (years)
r_p	Modified rate of biogas production by acetogenic/methanogenic microorganisms $(mL_b/gVS/day)$
$s_{\hat{\beta}_i}$	Approximate standard error of parameter estimates
$t_{\upsilon,\alpha/2}$	Student t-distribution parameter$(-)$
$t_{\upsilon,\alpha/2}$	student's t-distribution parameter
y_t	Biogas yield (mL_{biogas}/gVS)
y_{tm}	Maximum attainable biogas yield (mL_{biogas}/gVS)
$(\beta-\hat{\beta})$	Deviation between the real and the estimated model paramters
$\hat{\beta}$	Vector of estimated model parameters $(-)$
$\hat{\beta}$	Vector of estimated model parameters
μ_{max}	Maximum specific growth rate of acetogenic/methanogenic bacteria (day^{-1})
σ^2	True variance
χ^2	Reduced chi-square
AR	Attainable Regions
B-RADeS	Biodigester Rapid Analysis and Design System
CSTR	Continuous Stirred Tank Reactor
DSR	Differential Sidestream reactor
GUI	Graphical User Interface
GUIDE	Graphical User Interface Development Environment
IDEAS	Infinite DimEnsionAl State-space
NRT	Network Residence Time
NRT-C-AR	Network Residence Time Constrained Attainable Region
PFR	Plug Flow Reactor
ν	Number of degrees of freedom
C	Two-dimensional state vector
F	F-distribution varince comparism parameter
Pr_{el}	Feed in tariff rate for biogas based electricity $(\$/kWh)$
Q	Volumetric flow rate (mL/day)
$RMSE$	Root mean square error
S	Substrate concentration (g/L)
X	Acidogenic bacteria concentration (g/L)
b	Fraction of initial volatile solids remaining in effluent.
$cov(\beta)$	Covariance of estimated model parameters $(-)$
$cov(\beta)$	Covariance matrix of estimated model parameters
gVS	Gram volatile solids
gVS	Gram volatile solids
k	Kinetic constant of Chen and Hashimoto $(-)$
m	Measure of microbial adoption to stationary processes by mutation $(-)$
n	Could provide a useful measure of microbial cooperativity $(-)$
p	Number of model parameters
r	Discount rate (%)
$r(C)$	Two-dimensional reaction rate vector
α	Significance level
β	Vector of model parameters $(-)$
β	Vector of model parameters
μ	Specific growth rate of acetogenic/methanogenic bacteria (day^{-1})
τ	Retention time $(days)$

sludges and solid wastes. Using empirical methods to optimize design of anaerobic digesters often requires construction of expensive prototype systems and time consuming studies, which has been a key motivation for reliance on model-based techniques (Yu et al., 2013). Use of the models is again highly dependent on the availability of kinetic coefficients and hence modelling requirements for design of anaerobic digesters are often simplified to a minimal number of inputs and experimental states (most commonly biogas yield) required for model identification (Batstone, 2006). The simplified models employed for digester design can be single stage, based on the rate limiting step approach, or two-stage, based on lumping the process in to acid-forming and methane-producing microorganism. The single stage models previously employed in digester design include first order models (Linke, 2006; Momoh and Nwaogazie, 2011) and the models based on maximum bacteria growth rate (Fdez.-Güelfo et al., 2011; Fdez-Güelfo et al., 2012) while the two stage models include the biogas yield models presented by Momoh et al. (2013). Although simplified models have been used extensively in digester design, published articles are limited to mainly to determination of digester capacity based on parameters such a VS loading, temperature, etc. with first order models being mostly used (Momoh et al., 2013; Wang et al., 2007). For design, critical issues are hydrodynamics, as well as the behaviour of solids, which requires at least two-stage, with hydrolysis and biological steps (Batstone, 2006). This study focuses on hydrodynamics and an approach to optimize hydrodynamic configuration of anaerobic digesters based on simplified models will be a highly practicable as it will require less experimental measurements to estimate kinetic constants. Although the study develops two-stage models which can be identified based using only biogas yield measurements, the emphasis of the paper is not necessarily on the models but on how the authors use the models to develop new hydrodynamic configurations for operating low rate anaerobic digesters. The design objective is to minimize the process time as well as payback period by considering biodegradation and mixing as the only permitted fundamental processes occurring in the digester. The approach is based on the concept of attainable regions (AR), which is a technique for process synthesis and optimization that incorporates elements of geometry to understand how networks of chemical reactors can be designed and improved. "Following this initial work, many other researchers advanced AR research. Glasser et al. (1987) proposed a geometric approach that identified candidate AR's satisfying a number of necessary conditions that the AR must possess. Burri et al. (2002) demonstrated that, within the Infinite DimEnsionAl State-space (IDEAS) conceptual framework, construction of the true AR, and increasingly accurate AR approximants, can be carried out through Infinite Linear Programming (ILP), and a sequence of approximating finite Linear Programs (LP) respectively. Subsequently, Manousiouthakis et al. (2004) developed, within IDEAS, necessary and sufficient conditions that the true AR must satisfy, proposed the Shrink–Wrap algorithm for AR construction, and established, this algorithm's equivalence to the aforementioned LP based AR construction methods. They also demonstrated that the true AR can be potentially larger than the candidate AR's identified by geometric methods. Zhou and Manousiouthakis (2006) demonstrated that the true AR for reactor networks involving only reaction and mixing may be smaller than the true AR for reactor networks also incorporating diffusion effects (e.g. by considering non-ideal dispersion reactor models). Zhou and Manousiouthakis carried out pollution prevention studies using the AR approach (Zhou and Manousiouthakis, 2007a), extended the AR approach to reactor networks involving variable density fluids (Zhou and Manousiouthakis, 2007b), discussed the dimensionality of the space in which AR construction can be pursued (Zhou and Manousiouthakis, 2008), and extended the AR approach to non-isothermal reactor networks (Zhou and Manousiouthakis, 2009). Around the same time, Posada and Manousiouthakis (2008), proposed AR construction methods for reactor networks with multiple feeds, while Davis et al. extended the AR approach to batch reactor networks (Davis et al., 2008). More recently, Conner and Manousiouthakis extended the AR approach to general process networks (Conner and Manousiouthakis, 2014), while Ming et al. (2016) summarized many of the AR literature results." Geometrically, the attainable region represents the region bounded by the convex hull for the set of points achievable by the fundamental processes occurring in the system (Asiedu et al., 2015; Hildebrandt and Glasser, 1990; Hildebrandt et al., 1990). Once the AR has been determined, the limits of achievability by the system for the given kinetics and feed point is known and the boundary of the AR can then be used to answer different design and/or optimization questions related to the system. Our recent publication, Abunde Neba et al. (2019) has been first of its kind laying down theoretical framework for use of attainable regions to model optimal configurations of multistage anaerobic digesters. The study employed a four-state dynamic model of anaerobic treatment process and the attainable region analysis has been based on concentration (state) space but not residence time. The lack of residence time makes it impossible to size the digester structure or perform economic feasibility studies on the optimal digester structure. In addition, the four-state model (compared to the simplified model in this study) poses requirement for more experimental measurements hence limiting its application to situations where process measurements are limited. The current study is designed to illustrate how simplified models (requiring only biogas yield measurements) of the anaerobic treatment process can be used for attainable region analysis involving residence time space. "AlHusseini and Manousiouthakis (2013) were the first to incorporate residence-time considerations in the AR conceptual framework, by first introducing a production normalized, capital cost measure for a reactor network, that they termed "Network Residence Time" (NRT), and defined as "the ratio of the sum of the volumes of all reactors participating in the reactor network over the total volumetric flowrate entering the network." They subsequently introduced the Network Residence Time Constrained Attainable Region (NRT-C-AR), which they then proceeded to quantify using a Linear Programming formulation within the IDEAS conceptual framework. The advantage of constructing an AR of the anaerobic treatment process that incorporates residence time considerations, over the AR presented in our previous study, is that it enables the coupling of biodegradation kinetics, economic feasibility objectives and country specific macroeconomic parameters for the synthesis of biogas digester structures. By its use of attainable regions, knowledge of all possible states, for all possible digester configurations can be obtained considering biodegradation and mixing as the only fundamental processes occurring in the digester. Unlike previous studies where economic analysis is performed to determine the feasibility parameters of a predefined digester configuration, this study rather determines the biodigester network configuration required to achieve a given economic objective based on the macroeconomic situation of a given country. Finally, the study seeks to deploy the theoretical framework into a software in order to save time and effort for designers who are planning and designing biogas plants for different process or economic scenarios.

2. Attainable region theory for process synthesis and optimization

The Attainable Region (AR) theory is a technique that incorporates elements of geometry and mathematical optimization, to design and improve operation of chemical reactors (Ming et al., 2016). The power of the AR approach to process optimization is that the answer to all possible optimization problems, even the ones not considered are first determine, and then we look for ways of achieving that answer. In reactor operation knowledge of all possible reactor states for all possible reactor configurations, even those that have not yet been devised, is obtained. For a two-dimensional system, the convex hull for the set of all points achievable by all possible combinations of CSTR + PFR and mixing defines the attainable region. For higher dimensional systems, the attainable region is the convex hull for the set of points generated by all possible combinations of CSTR, PFR, DSR and mixing lines. The convex hull is understood as the smallest subset of a set of points that can be used to generate all other points by reaction and mixing (Ming et al., 2016). Geometrically, a convex hull is a finite convex polytope enclosed by a finite number of hyperplanes, which is interpreted in a two- dimensional space as the smallest polygon enclosed by planar facets such that all of the elements lie on or in the interior of the polygon (Asiedu et al., 2015). Once the AR has been determined, the limits of achievability by the system for the given kinetics and feed point is known, which can then be used to answer different design or optimization questions related to the system.

Given a set of reactions and associated kinetics, the following five key steps needs to be performed in order to complete an attainable region analysis (Ming et al., 2016):

- ➢ Define the reaction, dimension and feed set.
- ➢ Generate the AR using combinations of the fundamental processes.
- ➢ Interpret the AR boundary in terms of reactor equipment.
- ➢ Define the objective function and overlay this onto the AR to determine point of intersection with the AR boundary.
- ➢ Determine the specific reactor configuration required to achieve the intersection point.

Some necessary conditions for AR derived from the work of Glasser et al. (1987) can be summarized as follows:

- ➢ The AR includes all feeds to the system.
- ➢ The AR is convex.
- ➢ No process vector point out of the AR boundary.
- ➢ No rate vectors in the complement of the AR when extended backward intersects the AR.

The objective of this section is to analyze the aforementioned necessary requirements with respect to its application to the anaerobic treatment process. However, AR analysis requires that the process kinetics is known and we therefore begin by modeling the kinetics of the anaerobic treatment process.

2.1. Reaction kinetics of the anaerobic treatment process

In the present paper, the mathematical models describing the kinetics of substrate utilization and methane production in anaerobic treatment process are developed based on the approach presented by Momoh et al. (2013). The approach assumes that the AD process takes place in three stages. (i) hydrolysis/acidogenesis of the organic substrates in wastewater by acidogenic bacteria to produce acidified substrate; (ii) uptake of acidified substrate by acetogenic/methanogenic bacteria and (iii) acidified substrate assimilation, growth and biogas production by the acetogenic/methanogenic bacteria.

2.1.1. Enunciation of the process model

Fig. 2 presents the algorithm used to develop and validate the simplified two-stage based modes to predict biogas yield. The model development involves five main aspects, which include:

- ➢ Develop substrate degradation model.
- ➢ Formulate substrate uptake model.
- ➢ Choose microbial kinetic model.
- ➢ Derive models for substrate assimilation and biogas production.
- ➢ Identification of the developed model.

Considering these aspects led to a series of ordinary differential equations to predict biogas yield based on microbial growth kinetics

Stage 1: Hydrolysis and acidogenesis

Many constituents of organic wastes behave as complex substrates (polysaccharides, proteins, fats etc.). The Grau model presented in Eq. (1), which has widely been used to model multiple substrate removal kinetics (Kim et al., 2006; Liu, 2006) was therefore adopted for this study.

$$-\frac{dS}{dt} = k_{n(s)} X \left(\frac{S}{S_o}\right)^n \tag{1}$$

where $-dS/dt$ represents the rate of decrease in concentration of substrate being hydrolyzed, S is the concentration of initial substrate left at every instant following onset of hydrolysis, S_o is the concentration of initial substrate, $k_{n(s)}$ is the rate of substrate degradation by acidogenic bacteria, X is the concentration of acidogenic bacteria and n defines the degree of adaptation by acidogenic bacteria for substrate degradation.

The multicomponent substrate degradation model is based on the assumption that the different components are simultaneously removed and transported into the cells (Grau et al., 1975). Assuming hydrolysis and acidogenesis are catalyzed by acidogenic bacteria, whose concentration is constant, then Eq. (1) can be re-written as Eq. (2).

$$-\frac{dS}{dt} = K_{ha} \left(\frac{S}{S_o}\right)^n \tag{2}$$

Where, K_{ha} is the maximum rate of substrate degradation by acidogenic bacteria. Since anaerobic digestion is a biological process and the AR approach considers biodegradation and mixing as the only fundamental processes occurring in the digester, it becomes important to consider the non-biodegradable part of the substrate. The model is then modified as shown in Eq. (3)

$$-\frac{dS}{dt} = k_{n(s)} X \left(\frac{S}{S_o} - R_f\right)^n \tag{3}$$

Eq. (3) represents the kinetics of substrate degradation, where R_f is the recalcitrant fraction of initial volatile substrate that is non-biodegradable.

Stage 2: Substrate uptake by acetogenic/methanogenic microorganism

The hydrolytic model of Momoh et al. (2013), Eq. (4), which represents a modified version of the hydrolytic model presented by previous studies (Barthakur et al., 1991; Faisal and Unno, 2001; Zinatizadeh et al., 2006) was adopted.

$$S_u = S_o A_f \left(b - R_f\right)^n \tag{4}$$

This model takes into consideration the acidified substrate produced after substrate degradation by acidogenic bacteria as well as the uptake of acidified substrate by acetogenic/methanogenic microorganism. S_u represents the actual amount of the substrate that was acidified and utilized by the acetogenic/methanogenic bacteria while b is the fraction of initial volatile solids remaining in effluent. The coefficient $A_f = K_{ha}/(K_{am}(1-\alpha) + K_{ha})$ represents the rate limiting coefficient for very slow (case of $0<\alpha<1$) or very fast (case of $\alpha = 1$) metabolism of acidified substrate by the acetogenic/methanogenic bacteria (Momoh et al., 2013). The constant K_{am} is the maximum rate of substrate utilization by the acetogenic/methanogenic microorganisms.

Stage 3: Kinetics of bacteria growth and biogas production

The attainable region is unique for a given kinetics and a change in organic substrate can cause a change the kinetic model used to describe the growth of microorganisms. Table 1 presents a list of microbial growth models considered to model substrate assimilation. The table has been assembled from Kythreotou et al. (2014), who presented a comprehensive review of simple to scientific models for anaerobic digestion. As expected, the different models have different characteristics often making

Table 1
Microbial growth models selected to model substrate assimilation.

Author	Model equation	Eq. No.	Remark
Monod, 1949	$\mu = \mu_{max} \frac{S_u}{k_s+S_u}$	(5)	Describe growth processes for low substrate concentration
Moser, 1958	$\mu = \mu_{max} \frac{{S_u}^m}{k_s+{S_u}^m}$	(6)	Integrates effect of microbial adoption to stationary processes by mutation
Tessier model	$\mu = \mu_{max}(1 - e^{-\frac{S_u}{k_s}})$	(7)	An exponential function used to describe cell growth processes
Chen & Hashimoto, 1978	$\mu = \frac{\mu_{max} S_u}{kS_i+(1-k)S_u}$	(8)	Considers cell concentration depending on the level of substrate degradation
Haldane, 1930	$\mu = \frac{\mu_{max} S_u}{(k_s+S_u)(1+{S_u}/{k_i})}$	(9)	For growth process affected by the allosteric effectors present in the acidified substrate
Andrews, 1968	$\mu = \frac{\mu_{max} S_u}{k_s+S_u(1+{S_u}/{k_i})}$	(10)	Based on Haldane for enzyme inhibition at high substrate concentrations
Aiba et al., 1968	$\mu = \frac{\mu_{max} S_u}{k_s+S_u} exp(-{S_u}/{k_i})$	(11)	An empirical correlation to describe substrate inhibition
Dagley & Hinshelwood, 1983	$\mu = \mu_{max} \frac{S_u}{K_s+S_u}(1 - k_i S_u)$	(12)	An empirical correlation with critical inhibitor concentration of growth stop
Ierusalimsky, 1967	$\mu = \mu_{max} \frac{S_u}{k_s+S_u} \frac{k_i}{k_i+S_u}$	(13)	Haldane model for product inhibition
Moser, 1981 und Bergter, 1983	$\mu = \mu_{max} \frac{{S_u}^m}{k_s+{S_u}^m} \frac{k_i}{k_i+S_u}$	(14)	Production inhibition model with effect of microbial adoption

them more adequate to describe microbial growth of specific effluent and/or digester conditions rather than others.

k_s is the Monods' half saturaion contant for substrate uptake, μ_{max} is the maximum specific growth rate for methatnogenic archae, m is the coefficient of acetogenic/methanogenic microbial adaptation for cooperativity, S_i is the initial concentration of substrate taken up by acetogenic/methnogenic micrororgansims, k is the kinetic constant of Chen and Hasshimoto, and k_i is the substrate concentration where bacteria growth is reduced to 50% of the maximum specific growth rate due to substrate inhibition

Taking the case of the Monod model for growth of acetogenic/methanogenic microorganisms, and using product and cell yield coefficients, the rate of biogas production can be expressed by Eq. (15)

$$\frac{dy_t}{dt} = \frac{Y_{PS}}{Y_{XS}} \frac{\mu_{max} S_u}{k_s + S_u} X_{am} \tag{15}$$

Where dy_t/dt is the rate of change in biogas yield, y_t is the biogas yield, Y_{PS} is the biogas yield coefficient, Y_{XS} is the cell yield coefficient and X_{am} is the concentration of acetogenic/methanogenic microorganisms. If we consider the growth rate of the acetogenic/methanogenic bacteria is very slow or relatively constant while dy_t/dt can be described as the specific biogas yield rate (R_p) at the end of biogas production (Momoh et al., 2013), then Eq. (15) can re-written as Eq. (16). The parameter R_{pm}= ($X_{am}\, Y_{PS}/Y_{XS}$) is the maximum specific rate of biogas production.

$$R_p = \frac{R_{pm} S_u}{k_s + S_u} \tag{16}$$

Hence, by substituting Eq. (4) into Eq. (16) and rearranging, we obtain Eq. (15′).

$$R_p = \frac{R_{pm} S_o}{\frac{k_s}{A_f(b-R_f)^n} + S_o} \tag{15′}$$

The term $k_s/A_f(b - R_f)^n$ represents the Monod half saturation constant in terms of the fraction of acidified substrate taken up by acetogenic/methanogenic bacteria (represented as K_S) and the final biogas yield model considering the Monod kinetics is presented by Eq. (16′).

$$R_p = \frac{R_{pm} S_o}{K_S + S_o} \tag{16′}$$

Eq. (16) describes the biogas yield in anaerobic digester considering a batch operation mode. In cases where the system in operated in continuous mode, the initial substrate concentration (S_o) is converted to loading rate by multiplying the factor (Q/V) as shown in Eq. (17). The factor (Q/V) is the ratio of volumetric flow rate (Q) to volume of the digester (V).

Table 2
Two-stage based models to predict biogas yield rate.

Author	Model equation	Eq. No.
Monod, 1949	$R_p = R_{pm} \frac{S_o}{K_S+S_o}$	(19)
Moser, 1958	$R_p = R_{pm} \frac{{S_o}^m}{K_S+{S_o}^m}$	(20)
Tessier model	$R_p = R_{pm}(1 - k_p e^{-\frac{S_o}{K_S}})$	(21)
Chen & Hashimoto, 1978	$R_p = \frac{R_{pm} S_o}{K_* S_i+(1-k)S_o}$	(22)
Haldane, 1930	$R_p = \frac{R_{pm} S_o}{(K_s+S_o)(1+{S_o}/{K_i})}$	(23)
Andrews, 1968	$R_p = R_{pm} \frac{S_o}{K_s+S_o(1+{S_o}/{K_i})}$	(24)
Aiba et al., 1968	$R_p = R_{pm} \frac{S_o}{K_s+S_o} exp(-{S_o}/{K_i})$	(25)
Dagley & Hinshelwood, 1983	$R_p = R_{pm} \frac{S_o}{K_s+S_o}(1 - K_i S_o)$	(26)
Ierusalimsky, 1967	$R_p = R_{pm} \frac{S_o}{K_s+S_o} \frac{K_i}{K_i+S_o}$	(27)
Moser, 1981 und Bergter, 1983	$R_p = R_{pm} \frac{{S_o}^n}{K_s+{S_o}^n} \frac{K_i}{K_i+S_o}$	(28)

$$R_p = \frac{R_{pm}(QS_o/V)}{(K_s Q/V) + (QS_o/V)} \tag{17}$$

The resulting continuous mode counterpart of the biogas yield model considering Monod kinetics is shown by Eq. (18).

$$R_p = \frac{R_{pm} L_r}{K_{Lr} + L_r} \tag{18}$$

Where, the variable L_r is the organic loading rate into the biodigester and K_{Lr} is the Monod's half saturation constant defined in terms of the organic loading rate. Similar process was applied to develop the other biogas yield rate models by assuming that the growth process of the acetogenic/methanogenic microorganism can be described using the other growth models presented in Table 1. However, a parameter of k_p was introduced to the Tessier based model as a coefficient to the exponential term, which serves as an index of the processing speed of R_p as it approaches R_{pm} due to the change in S_o or L_r (depending on the mode of operation).The derived biogas yield models considering a two-stage biodegradation kinetics is presented in Table 2.

K is the kinetic constant of Chen and Hasshimoto defined in terms of specific biogas yield, K_i is the substrate concentration where specific biogas yield rate is reduced to 50% of the maximum specific biogas yield rate due to substrate inhibition and n provides a useful measure of microbial cooperativity to biogas production.

2.1.2. Parameter estimation and statistical methods

The kinetic constants of the different models were estimated using the Matlab nonlinear regression solver '*nlinfit*' (Mathworks Natick NA). In assessing the variability of the model identification process, we used the kernel density estimates and the parameter confidence regions. It is also interesting to note that marginal confidence intervals often used by several researchers do not account for correlations between the parameter estimates. Therefore, their use in parameter estimation can sometimes be misleading if there is strong correlation between several parameter estimates. In this study, we rather illustrate the use of joint confidence regions in assessing reliability of parameter estimates in least square regression.

The $100(1-\alpha)\%$ joint confidence region and the marginal confidence intervals of the parameter estimates is computed using Eqs. (29) and (30) respectively

$$\left(\beta-\hat{\beta}\right)^{T}\left(X^{T}X\right)\left(\beta-\hat{\beta}\right)\leq p\sigma^{2}F_{(1-\alpha),p,(n-p)} \tag{29}$$

$$\hat{\beta}\pm t_{\upsilon,\alpha/2}s_{\hat{\beta}_i} \tag{30}$$

Where $s_{\hat{\beta}_i}$ is the approximate standard error of the parameter estimates given by Eq. (31)

$$s_{\hat{\beta}_i}=\sqrt{diag(cov(\beta))} \tag{31}$$

$X^TX={s_{\hat{\beta}_i}}^2/cov(\beta)$ is the characteristic matrix, $cov(\beta)$ is the covariance matrix of estimated model parameters, $(\beta-\hat{\beta})$ is the deviation between the real (β) and the estimated model paramters $(\hat{\beta})$, $t_{\upsilon,\alpha/2}$ is the student's t-distribution parameter, ν is the number of degrees of freedom $(n-1)$, where n is the number of data points used to compute the variance and average, $F_{(1-\alpha),p,(n-p)}$ is the F-distribution varince comparism parameter, p is the number of model parameters being estimated, $(n-p)$ is the model degrees of freedom, and σ^2 is the true variance, and $\alpha=0.05$ is the level of significance.

2.2. AR analysis of the anaerobic treatment process

2.2.1. Reaction dimension and process vectors

Before it is possible to construct the AR, the engineer must first determine the space wherein the AR must reside (by choosing unique species components in the system that will represent the AR). These are variables required to characterize the state of the system, in this case, an anaerobic treatment process and must be sufficient to describe the dynamics of the fundamental processes chosen to describe the system. These variables would include biogas yield (y_t) and retention time (τ). A key criteria for selecting variables in AR is that they must obey the linear mixing law. The concept of "Network Residence Time" (NRT), as introduced by Al-Husseini and Manousiouthakis (2013) defines the residence time of a reactor network as the ratio of the sum of the volumes of all reactor units constituting the network to the total volumetric flowrate entering the network. Using this definition, it can be shown that the residence time of a network comprising two reactor units obey the linear mixing law, Eq. (32). This implies the overall residence time of the system must lie in a straight line between the residence times of the individual reactors, τ_1 and τ_2 comprising the system.

$$\tau_{mix}=\alpha\tau_1+(1-\alpha)\tau_2 \tag{32}$$

Where τ_{mix} is the overall residence time of the system comprising two individual reactors. The developed simplified kinetic models predict biogas yield (y_t), which is given in terms of volume of biogas produced (mL) per gram of volatile solids added to the digester (gVS). $y_t=V_g/m_{VS}$. Assume we have two digesters of known biogas yield, we can obtain the actual volume of biogas produced for digesters 1 and 2 by $V_{g1}=y_{t1}m_{VS1}$ and $V_{g2}=y_{t2}m_{VS2}$ respectively. Conservation of mass may be used to calculate the total biogas yield for both digesters. Conservation of mass ensures that the total mass of volatile solids in the mixture is equal to the sum of the individual masses contained in beakers 1 and 2, which is given by $m_{VST}=m_{VS1}+m_{VS2}$. Computing the biogas yield of the entire system is equivalent to determining the biogas yield for a mixture of digesters 1 and 2 since the density of the liquid phase of the digester can be assumed constant. The biogas yield of the mixture (y_{tM}) is given by the ratio of the total volume of biogas produced to the total mass of volatile acids added as shown by Eq. (33).

$$y_{tM}=\frac{y_{t1}m_{VS1}+y_{t2}m_{VS2}}{m_{VST}} \tag{33}$$

If we set $\alpha=m_{VS1}/m_{VST}$ then Eq. (33) can be written as Eq. (34), which is similar to the linear mixing law. What this means practically is that if we mix the contents of the liquid phase of two digesters, each of which contains a given quantity of volatile solids added, then the total biogas yield of the mixture will lie in a straight line joining that of both digesters.

$$y_{tM}=\alpha y_{t1}+(1-\alpha)y_{t2} \tag{34}$$

The process of combining the contents of two parallel digesters (or digester networks) of different volatile solids contents results in a linear mixing law measured in term of biogas yield. This implies biogas yield may be used in the construction of candidate ARs in a similar manner to that for concentration.

The biogas yield and the retention time grouped together form a vector called the characteristic vector; $\vec{C}=[y_t\ \ \tau]$, whose dimension determines the dimension of the optimization problem. We therefore have a 2-D optimization problem with the objective of minimizing $[\tau]$, time parameter.

If we assume that as substrate is consumed rate of change of biogas yield is directly correlated with the quantity of biogas to the biogas yield y_t, such that the driving force for gas production is disappearing when the biogas yield gradually approaches its maximum (y_{tm}) then for the mass of volatile solids added to the digester. This is modelled by Eq. (35)

$$r_p=\frac{dy_t}{dt}=R_p\left(1-\frac{y_t}{y_{tm}}\right) \tag{35}$$

Where, r_p is the modified rate of biogas production by acetogenic/methanogenic microorganisms. The reaction rate vector is therefore given by $\overrightarrow{r(C)}=[r_p\ \ r_\tau]$

2.2.2. Generate the AR using combinations of the fundamental processes

The attainable region (AR) represents the set of all possible states that can be achieved by a combination two fundamental processes, biodegradation and mixing in the case of the anaerobic treatment process. In AR theory, mixing is performed by a continuous stirred tank reactor (CSTR) while biodegradation (reaction) is performed by the plug flow reactor (PFR), since the operation of both reactors respectively mimics the two fundamental processes. At steady state operation, the general mathematical model of a CSTR and PFR are given respectively by Eqs. (36) and (37) respectively.

$$C=C_f+\tau r(C) \tag{36}$$

$$\frac{dC}{dx}=r(C) \tag{37}$$

C is the two-dimensional state vector made of biogas yield and the residence time, Eq. (38) while $r(C)$ is the reaction rate vector, which can be illustrated to be given by Eq. (39).

$$C = [y_t \quad \tau]^T \tag{38}$$

$$r(C) = [r_p \quad 1]^T \tag{39}$$

During construction of AR, the PFR trajectory is the set points generated by numerically solving the PFR equation while the CSTR locus is the set of points generated by solving the CSTR equation. The convex hull for the set of all points achievable by all possible combinations of CSTR + PFR defines the attainable region. The convex hull is understood as the smallest subset of a set of points that can be used to generate all other points by reaction and mixing (Ming et al., 2016). Geometrically, a convex hull is a finite convex polytope enclosed by a finite number of hyperplanes, which is interpreted in a two- dimensional space as the smallest polygon enclosed by planar facets such that all of the elements lie on or in the interior of the polygon (Asiedu et al., 2015)

The candidate attainable region was constructed with Matlab using the following five-steps

Step 1: Determine PFR trajectory from feed
Step 2: Determine the CSTR locus from feed
Step 3: Determine PFR trajectory from each CSTR point
Step 4: Construct the convex hull of the set of achievable points
Step 5: Verify the obtained AR against the necessary conditions of AR and if any condition is not met return extend the AR by running a PFR from the point of disagreement

The PFR equations are solved using the Matlab *ode45* routine for solving non-stiff differential equations while the system of nonlinear CSTR equations were solved using 'fsolve' routine The convex hall of the entire set of geometric points is obtained by using the Matlab '*convhull*' routine, which implements the Quickhull algorithm (Mathworks, Natick NA).

It is important to mention that even though this construction approach has been applied in a couple of studies (Ming et al., 2013; Ming et al., 2016), the NRT-C-AR obtained is a candidate and not the true NRT-C-AR. For a true AR, the Infinite DimEnsionAl State-space (IDEAS) conceptual framework is applied to obtain a general linear programming formulation for the construction of the true NRT-C-AR, as shown in (AlHusseini and Manousiouthakis, 2013). However, the interest of the study is not necessarily on the method used for AR construction, but on how the concept of attainable regions can be applied to optimized operation of the anaerobic treatment process. Also, even if just a candidate AR is obtained, it can still be used for process synthesis and optimization only that the totality of outputs is not obtained.

2.2.3. Interpret the AR boundary in terms of reactor equipment

The AR boundary is composed of two types of geometries: mixing lines referred to as lineations and manifolds of PFR trajectories referred to as protrusions. The role of PFRs on the AR boundary is to generate the outer extremities whereas CSTRs and DSRs (in the case of higher dimensional constructions) are used as connectors to these PFRs (Ming et al., 2016). This implies the AR boundary is defined in terms of reactor structures, and for two-dimensional constructions, the boundary is composed of combinations of the two fundamental reactor types and mixing lines. The PFR and CSTR each exhibit unique geometric interpretation and hence determining the reactor configurations that form the AR relates to interpreting the surfaces that form the AR boundary using its geometric properties.

2.2.4. Define the objective function and the optimal reactor structure

The AR, which defines the limits of achievability by the system for the given kinetics and feed point can be used to answer one or more design or optimization questions related to the system. Two-dimensional ARs involving residence time are particularly important for understanding the minimum reactor volume required for a given output. Since the construction and operation of anaerobic digesters generally requires capital investment, it would be interesting to use the AR concept in determining the profitability of the plant. However, we need to develop a suitable objective function that incorporates biogas yield and residence time (or digester volume).

The economic evaluation considers that biogas generated from the anaerobic digester is utilized for electricity generation. The total annual income, benefit (B_t) from installing the biomethane plant is determined by Eq. (40), which is benefit due to savings from electricity consumption.

$$B_t = 0.9P_{el} \times T_{el} \times b_e \times Pr_{el} \times gVS \times y_t \tag{40}$$

Where P_{el} is the percentage of methane utilized for electricity generation (which is 100% in the case of the current study), T_{el} is the annual time period for use of electricity, b_e is the unit conversion coefficient, Pr_{el} is the feed-in tariff rate for biogas based electricity, gVS is the gram mass of volatile solids fed in the digester.

The total annual expenses or operating cost (C_t) is computed by Eq. (41). The operating costs are assumed to be a function of two factors: the repair and maintenance costs, Eq. (41a), which is taken to by 1% of the cost of construction ($0.01C_{con}$) and the cost of biogas upgrading, which is a function of the biogas volume, Eq. (41b).

$$C_t = C_m + C_{pf} \tag{41}$$

$$C_m = 0.01C_{con} \tag{41a}$$

$$C_{pf} = gVS \times y_t \times T_{pr} \times Pr_{pf} \tag{41b}$$

Where, C_m is the annual cost of digester maintenance, C_{pf} is the annual cost of biogas purification, C_{con} is the cost of digester construction, T_{pr} is the annual time period for biogas purification and Pr_{pf} is the price for biogas purification per unit volume.

The cost of investment/construction is computed using the rates of a commercial biogas company in Ghana, stating the cost of digester construction to be $300 per cubic meter (Mohammed et al., 2017). This includes administrative, transport costs, consultancy fees and other logistic aspects. The final expression of the total annual cost of digester operation is given by Eq. (42).

$$C_t = 3V_D + gVS \times y_t \times T_{pr} \times Pr_{pf} \tag{42}$$

Where V_D is the volume of digester. The annual profit (P_t), is defined as the difference between annual benefit, B_t, due to savings from electricity consumptioin and the annual operating costs, C_t. This is expressed by Eq. (43).

$$P_t = B_t - C_t \tag{43}$$

Substituting the expressions for B_t and C_t into Eq. (43) the expression for the annual profit can be written as in Eq. (44)

$$P_t = gVS \times y_t \left(0.9P_{el} \times T_{el} \times b_e \times Pr_{el} - T_{pr} \times Pr_{pf}\right) - 3V_D \tag{44}$$

Since the AR is constructed in residence time space, it is necessary to express the volume of digester (V_D) in terms of residence time τ and volumetric flow rate Q. The expression for P_t as a function of residence time becomes;

$$P_t = gVS \times y_t \left(0.9P_{el} \times T_{el} \times b_e \times Pr_{el} - T_{pr} \times Pr_{pf}\right) - 3\tau Q \tag{45}$$

The economic evaluation of the digester investment is based on the payback period (PBP) (Gittinger, 1986) and the decision rule is that one generally accepts projects that require shorter number of years to recover the investment. The payback period is given by the annual profits, generated over n years, needed to recover the total

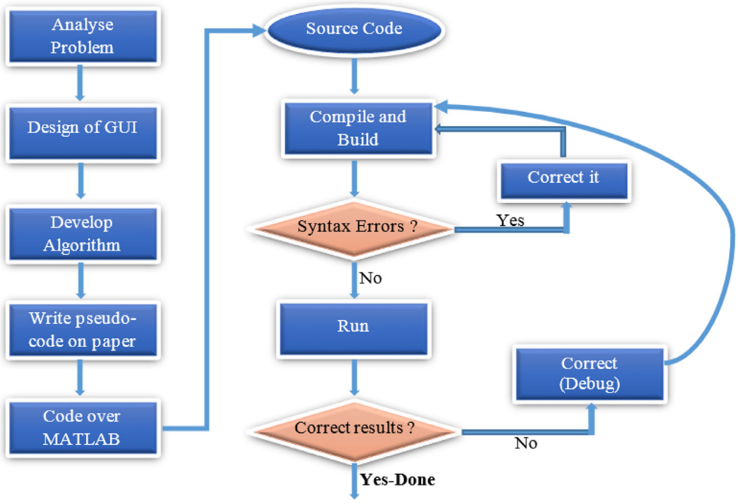

Fig. 1. B-RADeS software development procedure.

Table 3
Summary of input parameters used for the economic evaluation.

S.N	Parameter	Unit	Value
1	Discount rate	%	10
2	Average cost of digester and infrastructure	$\$/m^3(Base)$	300
3	Biogas-based electricity generator (500 kW)	$/4 *PCS*	600
4	Biodigester lifespan	*years*	20
5	Upper calorific value of methane gas	MJ/m^3	39.8
6	Density of methane	kg/m^3	0.75
7	Electricity equivalent of methane	kWh/m^3	11.06
8	Feed-in tariff rate for biogas based electricity	$\$/kWh$	17.5

future cost of the biogas plant, determined using the compounded interest formula. The payback period is evaluated using Eq. (46).

$$n_t P_t = C_{Inv}(1+r)^{n_t} \tag{46}$$

$$C_{Inv} = C_{con} + C_{Gen} + C_{misc} \tag{46a}$$

Where C_{Inv} is the cost of investment, r is the discount rate, C_{Gen} is the cost of biogas generator, C_{misc} is the miscellaneous cost and n_t is the project lifespan. Eq. (46) can be rearranged to express y_t as a function of τ, Eq. (47) which may be plotted over the AR boundary as contours (for different specified values of n) to determine the point(s) of intersection with the AR boundary. These intersection points represent the optimal operating point (which can interpreted into an optimal reactor structure) required to achieve a specified payback period.

$$y_t(\tau) = \frac{C_{Inv}(1+r)^{n_t} + 3\tau Q}{n_t gVS \times \left(0.9P_{el} \times T_{el} \times b_e \times Pr_{el} - T_{pr} \times Pr_{pf}\right)} \tag{47}$$

Table 3 presents of summary of the parameter sets that are used to perform the economic evaluation of designing a constructing a methane plant.

3. Development of computational model

The design of the graphical user interface (GUI) was done using the Matlab GUIDE (Graphical User Interface Development Environment). This is done using icon-based programing using several objects such as push buttons, static texts, edit texts, pop-up menus and axes handles. GUIDE generates a GUI and the m-file that contains the code to handle the initialization and launching of the GUI. After creation of the GUI, it was programmed by entering the algorithms into the various callback functions in the Matlab m-file. The steps of creating the B-RADeS GUI in Matlab are shown in the flowchart in Fig. 1.

4. Results and discussion

4.1. B-RADeS user interface

Fig. 2 presents the Graphical User Interface of the Biodigester Rapid Analysis and Design System (B-RADeS). This multi-level process design and simulation tool can be used to find the most efficient design of multi-stage anaerobic digester networks to achieve a defined economic and process objective. B-RADeS has several attributes that make it useful for a scientifically and economically objective process design and analysis platform for use by engineers to do their calculations during design and operation of multi-stage digesters. The main features of B-RADeS are as follows:

- ➢ B-RADeS is based on peer-reviewed models that describe growth kinetics of anaerobic digestion microorganisms. It includes ten simple biokinetic models derived based on biogas yield analogy.
- ➢ It does not rely on published kinetic coefficients, but it includes a section where the user determines kinetic coefficients required for digester synthesis from own experiments. Upon input of experimental data, B-RADeS automatically scans through the 10 models and ranks them in order of best fit using both quantitative and qualitative techniques.
- ➢ Data requirements are simple: Only experimental measurements of biogas yield are required to determine kinetic coefficients, construct attainable regions a well as synthesize digester networks.
- ➢ It takes into account the country-specific macroeconomic parameters (interest rate, electricity feed in tariff rate and annual working days) into the design process, which is a key motivation for investors.
- ➢ It is based on a systematic methodological framework for the design of multistage digester networks using the global opti-

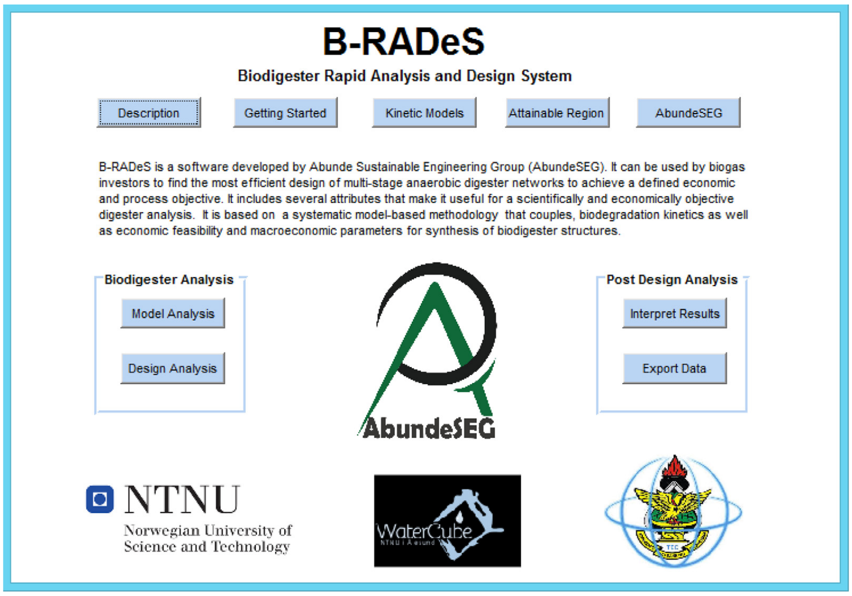

Fig. 2. B-RADeS inferace: Main overview screen.

mization technique of attainable regions. The main advantage of this approach over the use of superstructure optimization is that it enables knowledge of all possible states for all possible digester configurations (even those that have not yet been devised) to be first obtained, considering mixing and biodegradation as the only fundamental process occurring in the digester.

At the level of the main interface, users can get a description of the different biokinetic models, examine the underlying assumptions and approximations of the models, fundamental concepts required to interpret AR boundary in terms of digester structures, select the level of activity (Model fitting and analysis or digerter synthesis and analysis), and export simulations results for documentation. The software interface thus allows easy interactive modeling, design and simulation of multistage anaerobic digesters taking into consideration process kinetics and economic parameters.

4.2. Digester design and analysis with B-RADeS

The following four steps are required to perform complete analysis of an anaerobic treatment process using B-RADeS:

Step 1: Determine biogas yield kinetic model that best describes the organic substrate of interest. This requires data from anaerobic treatability studies using the substrate of interest. Upon input of experimental data, the software performs an automated fitting for all the ten models and ranks them in order of best fit using both numerical and graphical approaches. The numerical approach resides in the computation of a parameter, α (Eq. (48)), which takes into account four statistical coefficients for its computation. These coefficient include: the coefficient of determination (R^2), adjusted coefficient of determination (R^2Adj), root mean square error ($RMSE$) and the reduced chi-square (χ^2) were major validation criteria for model selection. For good quality fit, R^2, and R^2Adj values should be high while $RMSE$ and χ^2 should be low.

$$\alpha = \chi^2 + RMSE + \left(1 - R^2Adj\right) + \left(1 - R^2\right) \tag{48}$$

Models with the lowest value of α are considered more appropriate to describe a given data set if they share similar correlation coefficient. The graphical approach is based on examining the confidence contours which describe the correlation between the model parameter. The following five conditions are necessary for interpreting joint confidence regions

(a) If the region, given by an ellipse is aligned with the any of the coordinate axis (vertically or horizontally), then no correlation exist between the parameters that constitute the region.
(b) The parameter that lies on the coordinate axis with the greatest shadow corresponds to the parameter with the greatest variation.
(c) By definition, the elliptical region is centered at the least square estimate of the model parameters.
(d) If the region is a long narrow rotated ellipses, it indicates there exist significant correlation between parameter estimates.
(e) If values of zero for one or more of the parameter estimates lie in the region, these parameters are plausibly zero and the corresponding terms are not significant in the model.

Models, which show less correlation between the estimated parameters are more reliable. The user can also manually test the fitting of a particular model of interest without necessarily going through the automated fitting procedure (Fig. 3).

Step 2: Specify economic objective to be attained as well as county-specific macroeconomic parameters governing operation of the anaerobic digester system. The economic objective is specified in terms of the number of years required to recover investment following construction of an anaerobic digester for biogas production and electricity generation. The macroeconomic parameters are the interest rate, feed-in tariff for electricity generation from biogas as well as annual working days. B-RADeS the passes the estimated kinetic constants (for the best fitted model) to the design functionality, which together with the specified economic parameters is able

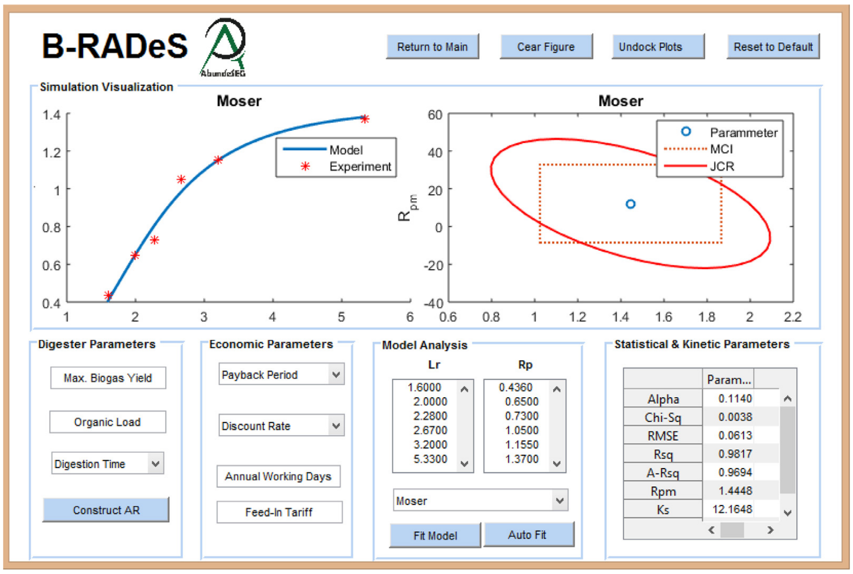

Fig. 3. B-RADeS interface: Here users can input experimental data, and estimate the kinetic constants for a given digestion process.

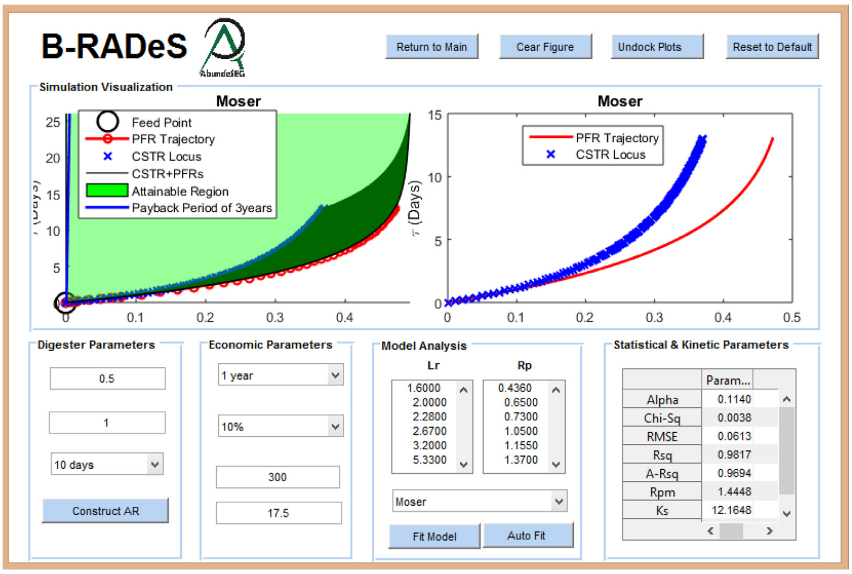

Fig. 4. B-RADeS interface: Here users set economic feasibility targets and country specific macroeconomic parameters, for B-RADeS to perform AR analysis for the user to interpret into digester structures.

to construct candidate attainable regions, and overlay the defined economic objective (payback period) onto the boundary of the attainable region in order to determine the point of intersection (see Fig. 4).

Step 3: Interpret the attainable regions and particularly the intersection point (which represents the optimal operating point) in terms of optimal digester structure. The interpretation of the AR boundary is based on three key fundamental results of two-dimensional AR used in everyday practice (Ming et al., 2016). These include:

➢ The AR is composed of reaction and mixing surfaces only. Reaction surfaces are always convex.
➢ Points that form convex sections of the AR boundary arise from effluent concentrations specifically from PFR trajectories.

Table 4
Statistical validity and kinetic coefficients of biogas models for swine wastewater.

Model	Statistical coefficients					Parameter estimates		
	α	χ^2	*RMSE*	R^2	$AdjR^2$	R_{pm}	K_S	Other
Moser & Bergter	0.0134	0.0000	0.0032	0.9955	0.9943	0.3451	0.7703	$m=1.5000$ $K_i=2.0000$
Moser	0.0288	0.0000	0.0052	0.9911	0.9852	0.1493	0.1663	$m=2.0332$
Tessier	0.0297	0.0000	0.0053	0.9908	0.9847	0.1439	0.4041	$kp=1.3171$
Andrews	0.0748	0.0001	0.0082	0.9704	0.9630	0.4162	1.7980	$K_i=2.0000$
Aiba	0.0797	0.0001	0.0084	0.9684	0.9604	0.6583	2.7423	$K_i=2.0000$
Ierusalimsky	0.1021	0.0001	0.0096	0.9590	0.9487	0.7091	2.7938	$K_i=2.0000$
Haldane	0.1200	0.0001	0.0111	0.9592	0.9320	0.5963	2.3614	$K_i=2.3615$
Dagley & H- inshelwood	0.1431	0.0001	0.0115	0.9416	0.9270	0.2263	0.8273	$K_i=2.0000$
Chen & Hashimoto	0.1464	0.0001	0.0116	0.9402	0.9252	0.1211	0.4507	$K_i=2.2000$
Monod	0.1464	0.0001	0.0116	0.9402	0.9252	0.2205	0.8011	–

➢ Points on the AR boundary that initiate these convex PFR trajectories (from point 2 above) arise from specialized CSTRs for two-dimensional constructions.

These guidelines are provided in the Attainable region section on the main menu of B-RADeSS.

4.3. Biodigester design case studies with B-RADeS

Multi-stage anaerobic digestion in which multiple digesters are operated in a network are designed to optimize each step of the anaerobic digestion process are potentially applicable for all wastewater treatment plants (EPA, 2006). Therefore, although many anaerobic wastewater treatment plants have traditionally performed anaerobic digestion processes as single stage, the use of multistate network digesters would allow these facilities to optimize the various stages of the anaerobic digestion process to meet their need. In fact, multistage digesters provide a great potential for a more efficient and flexible biogas systems that can better integrate into the bioeconomy and help harvest the energetic potential of organic waste while contributing to sustainable nutrient recycling (Cumiskey, 2005; Theuerl et al., 2019). We illustrate the capabilities of B-RADeS considering three anaerobic wastewater treatment case studies as presented in the following section.

Case study 1: Anaerobic digestion of swine wastewater

The objective here was to design a biodigester for treatment of swine wastewater that will yield a return on investment within 6 months (payback period) following start-up. Experimental data for anaerobic digestion of swine wastewater was obtained from Yang et al. (2016). Table 4 presents fitting characteristics and kinetic coefficients for all ten biogas yield models present in B-RADeS. The models are ordered using the automating fitting functionality in B-RADeS (see step 2 in Section 4.1). Amongst the Ten models fitted, the Moser & Bergter, Moser and Tessier based biogas yield models were the top three based on the numerical value of the α-parameter.

However, the selection of most accurate model for description of swine wastewater required consideration of the graphical approach of confidence contours as well. Fig. 5 presents model fits and confidence contours for the first three models.

By examining the confidence contours of the top three models presented in Fig. 5, we see that all the models have long rotated ellipses but that of the Moser-based model is narrowest indicating that there exist significant correlation between its parameter estimates. No major significant difference can be observed between that of the Moser & Bergter and the Tessier but since the former was better in terms of the numerical fitting criteria (alpha parameter), it therefore more accurately describe the experimental data for swine wastewater. Given the kinetic model of the system, the next step was to use the design analysis functionality of B-RADeS to perform an attainable region analysis of the system. Fig. 6 presents the candidate two-dimensional attainable region on which different payback period has been overlaid to indicate optimal operating points (points of intersection between the AR boundary and the objective function).

The objective function formulated is particularly important as it incorporates aspects of both total digester volume (residence time) and biogas yield. Three very interesting observations can be made from Fig. 6. (1) For a given biogas yield, higher payback periods are achievable for larger digester volumes (higher residence times). This is because for a given biogas yield, higher digester volumes will require more investment cost, hence resulting in a longer time to break even in investment. (2) For a given residence time on the AR boundary, shorter payback periods are achievable for higher yields of biogas. The results are highly consistent with practice because if we maintain the volume of the biogas plant constant, then we only require high yields in order to break even in investments for relatively shorter duration. (3) The range of payback periods considered intersect the AR at many points in the region, indicating that there are multiple operating points (multiple optima) for this system. However, since the objective of the design is to find a digester configuration with a payback period of 0.5 year, the reader can observe Fig. 7, which shows how the payback period of 0.5 year has been independently overlaid onto the boundary of the AR. The left plot of Fig. 7 presents the PFR trajectory and the CSTR locus, referred to as the base trajectories.

From Fig. 6, it is also important for readers to note that even though there are multiple intersection points of the objectives, the actual operating point to be chosen will depend on the investor's amount of capital. Points corresponding to smaller digester volume or smaller residence times (points associated with the lower part of the AR) require smaller capital investment while points corresponding to larger digester volumes require huge capital investment. Another very interesting remark in this example is that as the payback period increases, the influence of running cost (digester volume) on the payback period decreases, seen by the close proximity of the 2-, 3- and 4-year payback periods are to each other. This behavior has interesting interpretations on the investment strategy as it implies that it is more favorable to construct a larger digester, with larger operating expenses, with the intention of producing a higher biogas quantity of biogas. Hence, even if the required digester system is more complex, the plant is profitable in shorter a period of time.

Furthermore, notice from Fig. 6 that payback periods of less than 0.3 years are not achievable irrespective of the digester structure employed. Contour lines for payback periods less than 0.3 years turn to approach the horizontal axis and do not intersect the AR boundary at all. However, the attainable region is unique for a given kinetics and feed point, and if any of these change, the limits of achievability by the system may also change and hence payback periods less than 0.3 years could become attainable.

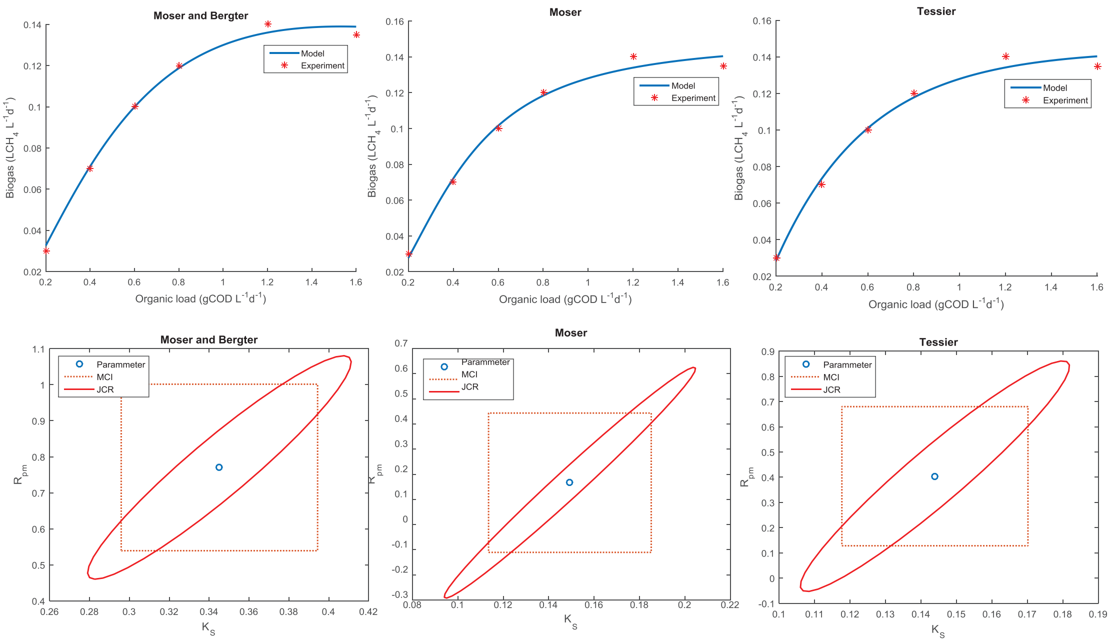

Fig. 5. Model fits and confidence contours for anaerobic digestion of swine wastewater.

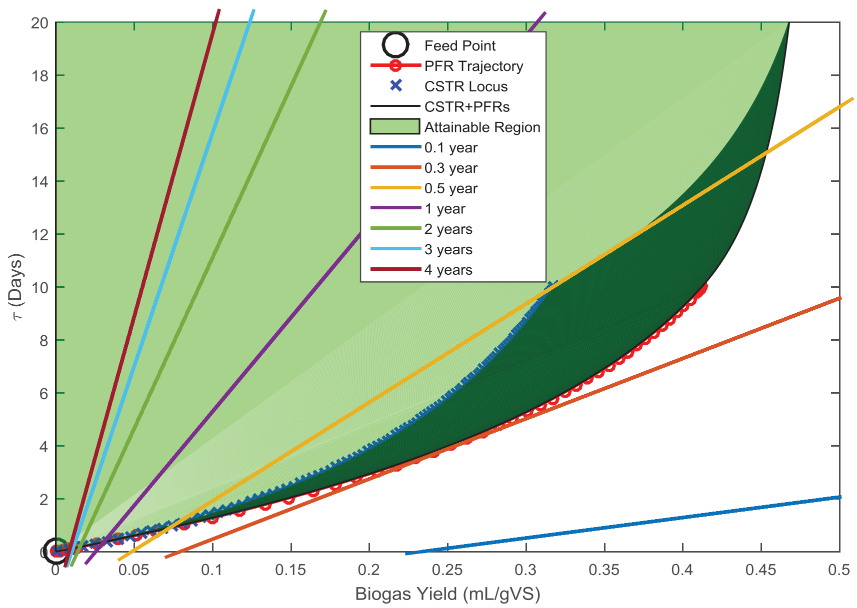

Fig. 6. Candidate attainable region showing contours of different payback periods: Feed-in tariff: 17.5, annual working days: 300, discount rate: 10%, digestion time: 10days, organic load: 0.5 gVS/L, experimental methane yield: 0.5 mL/gVS. The dark green part represents a series of PFR trajectories run from each point on a CSTR locus. They are actually dark line plots but since they are many, it gives an appearance of dark green. Notice on the legend that the CSTR+PFRs appear as a dark line plot.

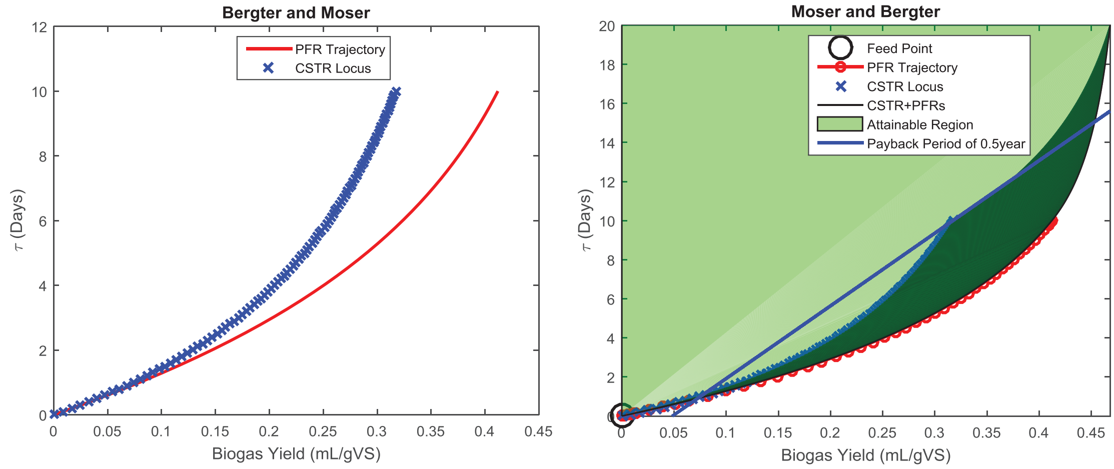

Fig. 7. PFR trajectory and CSTR locus (left figure) and candidate attainable region with overlaid payback period of 0.5year (right plot).

Table 5
Statistical validity and kinetic coefficients of biogas models for POME.

Model	Statistical coefficients					Parameter estimates		
	α	χ^2	*RMSE*	R^2	$AdjR^2$	R_{pm}	K_S	Other
Moser	0.1140	0.0038	0.0613	0.9817	0.9694	1.4448	12.1648	$m = 3.3230$
Tessier	0.1439	0.0051	0.0718	0.9749	0.9581	1.4538	1.3568	$kp = 2.3461$
Moser & Bergter	0.3623	0.0153	0.1236	0.9007	0.8759	25.1932	47.3672	$m = 1.5000$
$K_i = 2.0000$								
Andrews	0.3861	0.0165	0.1284	0.8928	0.8660	16.4562	42.2917	$K_i = 2.0000$
Dagley & H- inshelwood	0.4428	0.0194	0.1393	0.8738	0.8422	4.5000	10.4429	$K_i = 2.0000$
Chen & Hashimoto	0.4488	0.0197	0.1405	0.8717	0.8396	0.1915	0.9498	$K_i = 2.2000$
Monod	0.4488	0.0197	0.1405	0.8717	0.8396	3.8114	8.7594	–
Haldane	0.5162	0.0255	0.1596	0.8758	0.7930	8.7345	20.3733	$K_i = 20.2753$
Ierusalimsky	–	–	–	–	–	–	–	–
Aiba	–	–	–	–	–	–	–	–

It is also interesting for the readers to note that the particular choice of payback period might also influence the optimal reactor structure necessary to achieve it. To attain a payback period of 0.5 year, larger capital investments will require a CSTR followed by a PFR as the optimal digester structure (corresponding to intersection point at the lower part of the AR) while smaller capital investments will require a PFR as the optimal reactor structure (corresponding to intersection point at the upper part of the AR)

Case study 2: Anaerobic digestion of palm oil mill effluent (POME)

Here, we consider the design and optimization of a multistage anaerobic digester for the treatment of palm oil mill wastewater in which the design objective is to attain a payback period of 1 year. We demonstrate the use of B-RADeS by considering experimental data from Faisal and Unno (2001). Table 5 presents fitting characteristics and kinetic coefficients for all ten biogas yield models present in B-RADeS while Fig. 8 presents model fits and confidence contours for the first three models. Considering the numerical and graphical approaches for model selection (as clearly explained in case 1) the Moser based biogas yield model was selected to describe the kinetics of the process. Unlike case study 1, we notice that information for the Ierusalimsky and the Aiba based models is not included. This is because B-RADeS is programmed such that during automatic fitting of all 10 models, models that show any error during the fitting process are assigned a very large alpha value. The user then gets a displayed message stating such models and indicating that they should be deleted from the list. Hence the Ierusalimsky and the Aiba based models were not considered in the fitting experiment for POME.

Fig. 9 (right plot) presents the candidate two-dimensional attainable region on which the 1 year payback period has been overlaid to indicate optimal operating points (points of intersection between the AR boundary and the objective function). The left plot of Fig. 9 presents the PFR trajectory and the CSTR locus.

Unlike case 1, the objective function intersects the lower part of the AR boundary slightly close to the feed point and rather approaches the unbounded section of the AR in the upper part of the curve. It is worth noting that the AR will always be unbounded at the residence time axis owing to the fact that states that are achieved at a given residence time will always be achievable for all later residence times (Ming et al., 2016). Considering the intersection point at the lower part of the AR boundary, an anaerobic PFR is required as an optimal reactor structure to achieve a payback period of 1 year.

Case study 3: Anaerobic digestion of pharmaceutical wastewater

Finally, we demonstrate the usability of B-RADeS for synthesis of a digester structure for treatment of pharmaceutical wastewater in which the objective is to attain a payback period of 2 years. The experimental data has been obtain from the work of Pandian et al. (2011). Table 6 presents fitting characteristics and kinetic coefficients for all ten biogas yield models while Fig. 10 presents model fits and confidence contours for the first three models. Considering the numerical and graphical approaches for

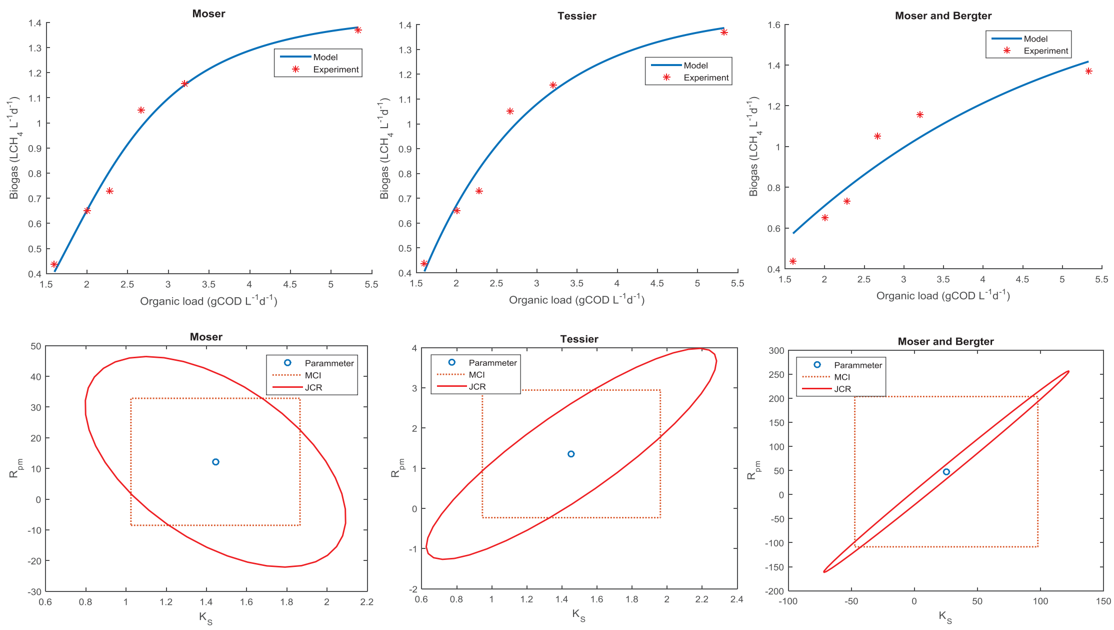

Fig. 8. Model fits and confidence contours for anaerobic digestion of POME.

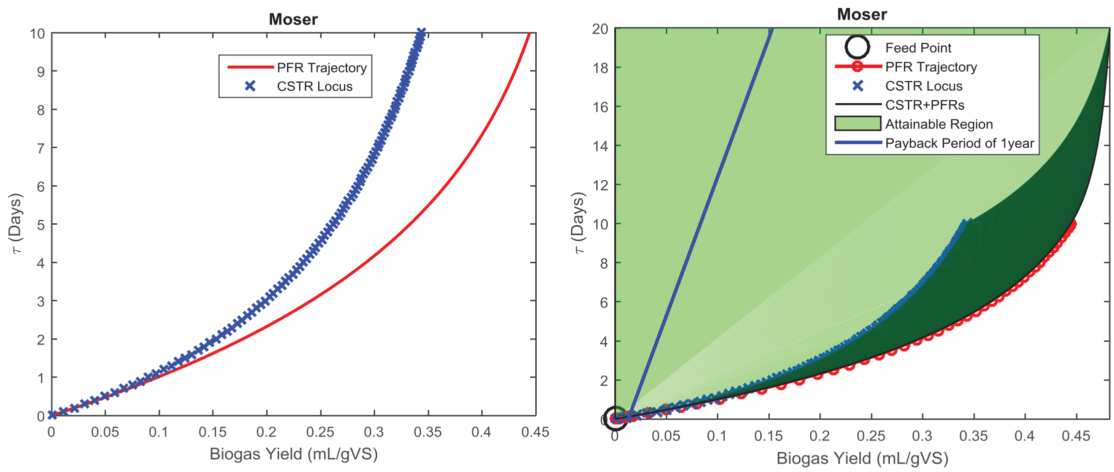

Fig. 9. PFR trajectory and CSTR locus (left figure) and candidate attainable region with overlaid objective function (1-year payback period). Feed-in tariff: 17.5, annual working days: 300, discount rate: 10%, digestion time: 10days, organic load: 1.0 gVS/L, experimental methane yield: 0.5 mL/gVS.

model selection the Tessier based biogas yield model was selected to describe the treatment kinetics of pharmaceutical wastewater.

Fig. 11 (right plot) presents the candidate two-dimensional attainable region on which the 2 year payback period has been overlaid to indicate optimal operating points (points of intersection between the AR boundary and the objective function). The left plot of Fig. 11 presents the PFR trajectory and the CSTR locus.

Unlike cases 1 and 2, the objective function intersects the lower part of the AR boundary at the feed point (0, 0), which is not feasible to operate a system at this point. However, the objective function passes through other points within the AR, any of which could be selected to operate the system. Consider a line A-B drawn such that it cuts the residence time axis as indicated on Fig. 12.

The intersection point (point C) of line AB and the objective function is selected to be the optimal operating point and the digester structure corresponding to this point is the optimal digester design. Since this point lies on line AB, it is attainable my mixing digester effluents from points A and B, Eq. (49) (see illustration in Section 2.2.1). Point B lies on the PFR trajectory and is therefore attainable by running an anaerobic PFR for 3 days (point where line AB intersects the residence time axis). Point A lies on the residence time axis (corresponding to biogas yield of zero) but since a biogas

Table 6
Statistical and kinetic coefficients of biogas models for pharmaceutical wastewater.

Model	Statistical coefficients					Parameter estimate		
	α	χ^2	*RMSE*	R^2	$AdjR^2$	R_{pm}	K_S	Other
Moser	0.3880	0.0357	0.1890	0.9388	0.8980	1.7323	152.4800	$m = 2.4740$
Andrews	0.3962	0.0343	0.1853	0.9215	0.9019	48.4565	384.4733	$K_i = 2.0000$
Tessier	0.4244	0.0402	0.2005	0.9311	0.8852	1.7138	6.9871	$kp = 1.4187$
Dagley & H- inshelwood	0.5460	0.0518	0.2277	0.8816	0.8520	5.3108	35.0258	$K_i = 2.0000$
Moser & Bergter	0.6524	0.0648	0.2545	0.8520	0.8149	105.7607	486.2520	$m = 1.5000$ $K_i = 2.0000$
Monod	0.7000	0.0707	0.2659	0.8385	0.7981	2.6339	15.5124	–
Chen & Hashimoto	0.7000	0.0707	0.2659	0.8385	0.7981	0.0747	0.9716	$K_i = 2.2000$
Haldane	0.7083	0.0773	0.2780	0.8676	0.7793	7.0897	44.5099	$K_i = 44.4790$
Aiba	–	–	–	–	–	–	–	–
Ierusalimsky	–	–	–	–	–	–	–	–

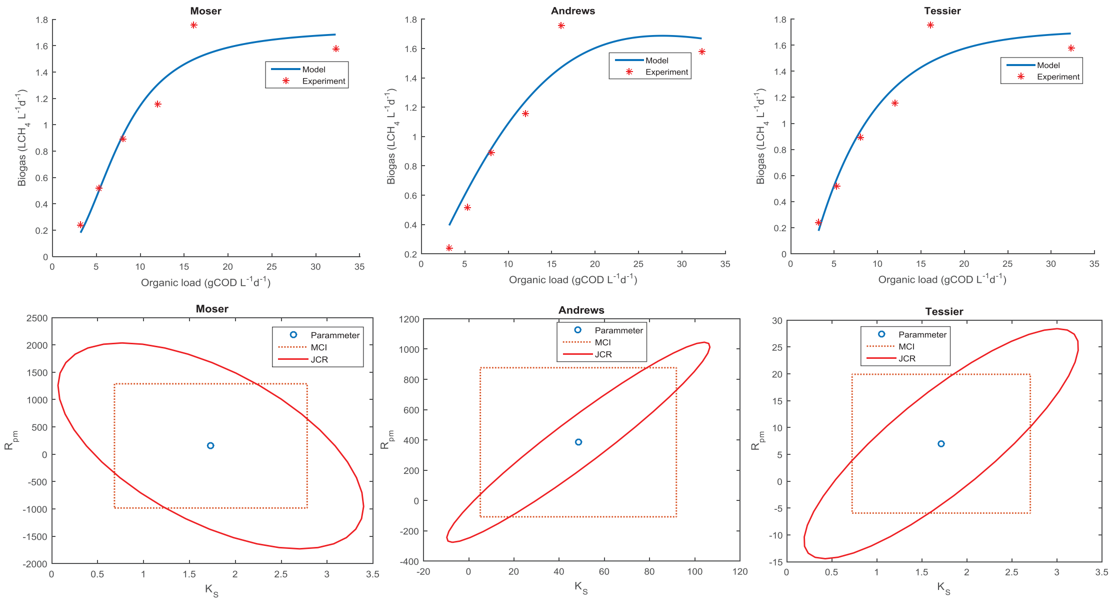

Fig. 10. Model fits and confidence contours for anaerobic digestion of pharmaceutical wastewater.

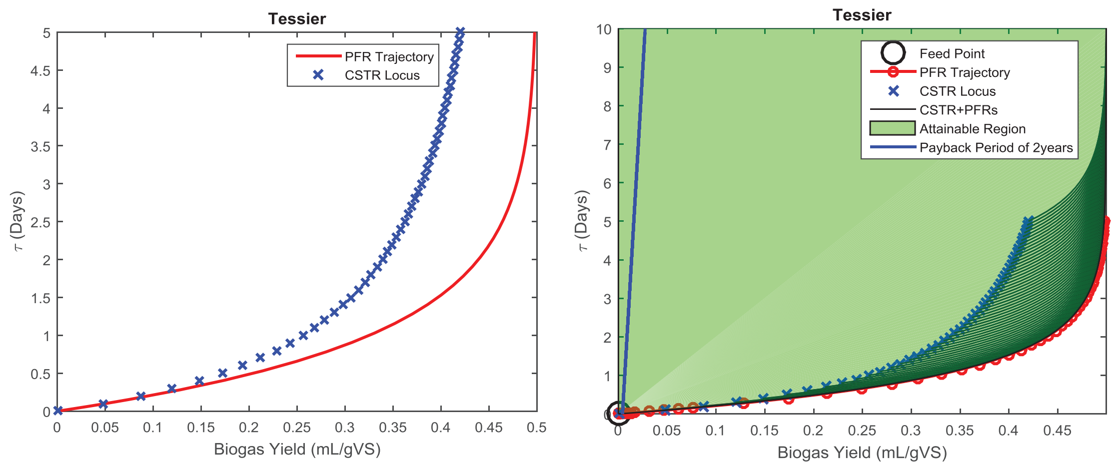

Fig. 11. PFR trajectory and CSTR locus (left figure) and candidate attainable region with overlaid objective function (2-year payback period). Feed-in tariff: 17.5, annual working days: 100, discount rate: 10%, digestion time: 5days, organic load: 5.0 gVS/L, experimental methane yield: 0.5 mL/gVS.

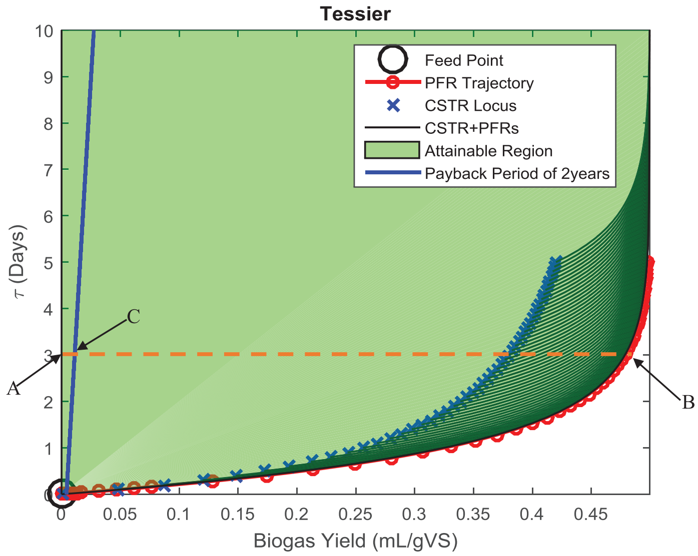

Fig. 12. Selection of operating point for anaerobic digestion of pharmaceutical wastewater.

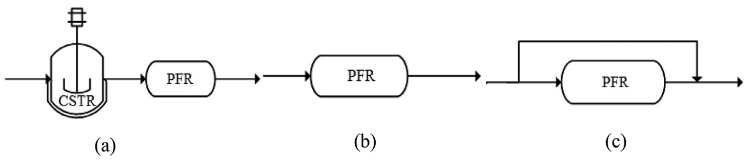

Fig. 13. Optimal digester structures obtained depending on the design operating point for different digested substrates. (a) Swine wastewater: a CSTR followed by a PFR, (b) Palm oil mill wastewater: A PFR, and (c) Pharmaceutical wastewater: PFR with bypass of the feed.

yield of zero is achieved at a lower residence time (feed point) it is also achievable at any later residence time on the residence time axis. The optimal digester structure required to achieve a payback period of 2 year therefore consist of a PFR and a bypass valve from the feed point.

$$y_{tC} = \alpha y_{tA} + (1-\alpha) y_{tB} \quad 0 \le \alpha \le 1 \tag{49}$$

Table 7 provides a summary of the payback periods, intersection points as well as the required digester volumes for the three design case studies. As earlier mentioned, there a several intersection points between the objective function and the AR and the points selected in Table 7 are for illustration. In practice, the actual operating point selected by the designer will depend on other factors such as cost and space constraints. This is because different points will correspond to different digester structures some of which have different space and/or cost requirements.

Fig. 13 present the optimal digester structures corresponding to the selected points of operation (see Table 7) for the three case studies of anaerobic digestion.

The article is of high relevance to designers of biogas digesters as it is first of its kind demonstrating the usefulness of biogas yield measurements for design and optimization of biodigester structures. Biodigester structures, which involve a staged operation of either multiple digesters or a single digester with by-pass or recycle streams has gained increasing importance due to their ability to optimize every step in the anaerobic treatment process. The authors of this study have presented a systematic model-based methodology for synthesis of biodigester structures requiring simple data requirements. The framework is based on the global optimization technique of attainable regions. The main advantage of this approach over other approaches is that it enables knowledge of all possible states for all possible digester structures (even those that have not yet been devised) to be first obtained, considering mixing and biodegradation as the only fundamental process occurring in the digester. The main novelty of the study is that it couples, biodegradation kinetics, economic objectives (payback period) and country specific macroeconomic parameters in the design process. It is also interesting for the readers to note that the particular choice of economic feasibility objective (payback

Table 7
Summary of required design specifications for three case studies.

Case study	Payback period	Operating point	Residence time
Swine wastewater	6 months	[0.45, 15.00]	15 days
Palm oil mill effluent	1 year	[0.03, 0.20]	4.8 h
Pharmaceutical wastewater	2 years	[0.02, 3.00]	3 days

period), as well as macroeconomic parameters (interest rate or feed-in tariff rate) influence the optimal biodigester structure necessary to achieve it. Due to the minimal data requirements, the study offers great promises for widespread application to enhance design of biodigester structures since biogas yield measurements are readily available from treatability studies. Even though the model-based methodolgy has been applied to only been applied to swine wastewater, palm oil mill effluent and pharmaceutical wastewater, other types of wastewaters as well as solid wastes or even wastewater sludges offers a strong research attraction for application of the framework presented in this study. This study bridges the gap between research, development and implementation of digester networks.

It is interesting to compare the results of this study with our recent publication Abunde Neba et al. (2019) using attainable regions to synthesize multistage anaerobic digesters. The study considered a four-state dynamic model of anaerobic treatment process. More states imply more need for experimental measurements making it less applicable to situations where process measurements are limited. In addition, the two-dimensional attainable regions were constructed in concentration space only, and the lack of residence time makes it impossible to size the digester structure. Furthermore, the study focused on process objectives (volumetric methane productivity and gas stabilization) for design meanwhile the current study simultaneously couples process parameters, economic objectives in the construction attainable regions in residence time space, which is a key motivation for investors and makes it possible to size the digester structures.

Considering both studies put together, the results can be applied to design and optimize (based on economic and process objectives) multistage digester structures in cases of available as well as limited experimental measurements.

5. Conclusion

The present study was designed to develop a theoretical framework for using simplified kinetic models based on only biogas yield to model and optimize (based on economic objectives) hydrodynamic configurations of anaerobic digesters. The study has developed two-stage kinetic models based on the biogas yield approach and formulated and economic evaluation model based on simple payback period. Furthermore, the study has shown that by using two-dimensional attainable regions in residence time space, it is possible to design and optimize hydrodynamic configurations for operating low rate anaerobic digesters considering mixing and biodegradation as the only fundamental processes occurring in the digester. Attainable region analysis is a global optimization technique which incorporates elements of geometry and mathematical optimization to synthesize optimal reactor networks to achieve a given objective. For proof-of-concept, we have considered three design case studies and applied the simple payback period as the design objective for modeling optimal digester configurations.

In this article a novel software, which can be used by biodigester design engineers to rapidly model hydrodynamic configurations using experimental measurements of biogas yield. The software package has been successfully employed to model the kinetics and design optimal digester configurations for three different substrates: swine wastewater, palm oil mill effluent and pharmaceutical wastewater. Broad functionalities of B-RADeS is able to address key problems arising in design and optimization of anaerobic digester networks including: (1) modeling of anaerobic digestion kinetics by automatically fitting 10 different biokinetic models and assessing the quality of fit using numerical and graphical approaches, and finally using the selected models to determine kinetic coefficients of the process (2), Construction of two-dimensional attainable regions in residence time space, and (3) optimization of anaerobic digester structures using simple payback period as well as county-specific macroeconomic parameters such as interest rate and renewable energy feed-in tariff rate. This software allows user to animate simulation results and, thereby, present them in more comprehensible and aesthetic mode. The article therefore concludes that, in principle, with only experimental measurements of biogas yield, B-RADeS can be used to generate the attainable region of the process which can be used to propose the optimal digester configuration for the process. This is highly practicable for use in small-scale onsite systems since data requirements are simple: Only experimental measurements of biogas yield are required to complete determination of kinetic coefficients, construction of attainable regions a well as synthesis of digester networks.

Finally, the study has demonstrated that the use of digester structures as opposed to single digesters improves process economics and reduces the time required to break even in investment. This result can be considered as a fundamental framework for design of digester networks using attainable regions when only biogas yield measurements are available. As recommendation for further studies, it would be interesting to apply the Infinite DimEnsionAl State-space (IDEAS) to obtain a general mathematical formulation for the construction of a true NRT-C-AR and compare the optimization performance with what has been obtained in the current study.

Funding

This work was supported by EnPe-NORAD under the project Upgrading Education and Research Capacity in Renewable Energy Technologies (UPERC-RET).

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

Our team expresses gratitude to the following institutions; The Brew-Hammond Energy Centre, KNUST Ghana, The Water and Environmental Engineering Group, NTNU Ålesund and the Abunde Sustainable Engineering Group (AbundeSEG) for its immense technical support.

References

Abunde Neba, F., Asiedu, N.Y., Addo, A., Morken, J., Østerhus, S.W., Seidu, R., 2019. Use of attainable regions for synthesis and optimization of multistage anaerobic digesters. Appl. Energy 242, 334–350.

Alhusseini, Z.I., Manousiouthakis, V.I., 2013. Network residence time constrained attainable region (NRT-C-AR). Chapter 2 global optimization of chemical reactors and kinetic optimization, PhD Thesis, UCLA, pp. 53–86.

Asiedu, N., Hildebrandt, D., Glasser, D., 2015. Experimental simulation of three-dimensional attainable region for the synthesis of exothermic reversible reaction: ethyl acetate synthesis case study. Ind. Eng. Chem. Res. 54, 2619–2626.

Barthakur, A., Bora, M. & Singh, H.D. 1991. Kinetic model for substrate utilization and methane production in the anaerobic digestion of organic feeds. 7, 369–376.

Batstone, D.J., 2006. Mathematical modelling of anaerobic reactors treating domestic wastewater: rational criteria for model use. Rev. Environ. Sci. Bio/Techn. 5, 57–71.

Burri, J.F., Wilson, S.D., Manousiouthakis, V.I., 2002. Infinite dimensional state-space approach to reactor network synthesis: application to attainable region construction. Comput. Chem. Eng. 26, 849–862.

Conner, J.A., Manousiouthakis, V.I., 2014. On the attainable region for process networks. AIChE J. 60, 193–212.

Cumiskey, A. 2005. UK leads the way in advanced digestion technology. Water Wastewater Int..

Davis, B.J., Taylor, L.A., Manousiouthakis, V.I., 2008. Identification of the attainable region for batch reactor networks. Ind. Eng. Chem. Res. 47, 3388–3400.

EPA, U. S. E. P. A. 2006. Biosolids technology fact sheet, multi-stage anaerobic digestion. National Service Center or Environmental Publications (NSCEP).
Faisal, M., Unno, H., 2001. Kinetic analysis of palm oil mill wastewater treatment by a modified anaerobic baffled reactor. Biochem. Eng. J. 9, 25–31.
Fdez.-Güelfo, L.A., Álvarez-Gallego, C., Sales Márquez, D., Romero García, L.I., 2011. The effect of different pretreatments on biomethanation kinetics of industrial organic fraction of municipal solid wastes (OFMSW). Chem. Eng. J. 171, 411–417.
Fdez-Güelfo, L.A., Álvarez-Gallego, C., Sales, D., Romero García, L.I., 2012. Dry-thermophilic anaerobic digestion of organic fraction of municipal solid waste: methane production modeling. Waste Manage. (Oxford) 32, 382–388.
Gittinger, J.P., 1986. Economic Analysis of Agricultural Project. The John Hopkins University Press, Baltimore, USA.
Glasser, D., Crowe, C., Hildebrandt, D., 1987. A geometric approach to steady flow reactors: the attainable region and optimization in concentration space. Ind. Eng. Chem. Res. 26, 1803–1810.
Grau, P., Dohányos, M., Chudoba, J., 1975. Kinetics of multicomponent substrate removal by activated sludge. Water Res. 9, 637–642.
Henze, M., Van Loosdrecht, M.C.M., Ekama, G.A. & Brdjanovic, D. 2008. *Biological wastewater treatment*, IWA Publishing.
Hildebrandt, D., Glasser, D., 1990. The attainable region and optimal reactor structures. Chem. Eng. Sci. 45, 2161–2168.
Hildebrandt, D., Glasser, D., Crowe, C.M., 1990. Geometry of the attainable region generated by reaction and mixing: with and without constraints. Ind. Eng. Chem. Res. 29, 49–58.
Kim, J.K., Oh, B.R., Chun, Y.N., Kim, S.W., 2006. Effects of temperature and hydraulic retention time on anaerobic digestion of food waste. J. Biosci. Bioeng. 102, 328–332.
Kythreotou, N., Florides, G., Tassou, S.A., 2014. A review of simple to scientific models for anaerobic digestion. Renew. Energy 71, 701–714.
Linke, B., 2006. Kinetic study of thermophilic anaerobic digestion of solid wastes from potato processing. Biomass Bioenergy 30, 892–896.
Liu, Y., 2006. A simple thermodynamic approach for derivation of a general Monod equation for microbial growth. Biochem. Eng. J. 31, 102–105.
Manousiouthakis, V.I., Justanieah, A.M., Taylor, L.A., 2004. The Shrink–Wrap algorithm for the construction of the attainable region: an application of the ideas framework. Comput. Chem. Eng. 28, 1563–1575.
Mao, C., Feng, Y., Wang, X., Ren, G., 2015. Review on research achievements of biogas from anaerobic digestion. Renew. Sustain. Energy Rev. 45, 540–555.
Mes, T.Z.D.D., Stams, A.J.M., Reith, J.H., Zeeman, G., 2003. Methane production by anaerobic digestion of wastewater and solid wastes. Bio-methane and Bio-hydrogen: Status and Perspectives of Biological Methane and Hydrogen Production. The Hague, The Netherlands: Dutch Biological Hydrogen Foundation – NOVEM.
Ming, D., Glasser, D., Hildebrandt, D., 2013. Application of attainable region theory to batch reactors. Chem. Eng. Sci. 99, 203–214.
Ming, D., Glasser, D., Hildebrandt, D., Glasser, B., Metzer, M., 2016. Attainable Region Theory: An Introduction to Choosing an Optimal Reactor. John Wiley & Sons, Inc., Hoboken, New Jersey.
Mohammed, M., Egyir, I.S., Donkor, A.K., Amoah, P., Nyarko, S., Boateng, K.K., Ziwu, C., 2017. Feasibility study for biogas integration into waste treatment plants in Ghana. Egypt. J. Pet. 26, 695–703.
Momoh, O.L.Y., Anyata, B.U., Saroj, D.P., 2013. Development of simplified anaerobic digestion models (SADM's) for studying anaerobic biodegradability and kinetics of complex biomass. Biochem. Eng. J. 79, 84–93.
Momoh, O.L.Y., Nwaogazie, I.L., 2011. The effect of waste paper on the kinetics of biogas yield from the co-digestion of cow manure and water hyacinth. Biomass Bioenergy 35, 1345–1351.
Pandian, M., Huu-Hao, N., Pazhaniappan, S., 2011. Substrate removal kinetics of an anaerobic hybrid reactor treating pharmaceutical wastewater. J. Water Sustain. 1, 301–312.
Posada, A., Manousiouthakis, V., 2008. Multi-feed attainable region construction using the Shrink–Wrap algorithm. Chem. Eng. Sci. 63, 5571–5592.
Theuerl, S., Herrmann, C., Heiermann, M., Grundmann, P., Landwehr, N., Kreidenweis, U. & Prochnow, A. 2019. The future agricultural biogas plant in Germany: a vision. 12, 396.
Wang, L.K., Shammas, N.K., Hung, Y.-.T., 2007. Biosolids Treatment Processes. Humana Press Inc, New Jersey.
Yang, H., Deng, L., Liu, G., Yang, D., Liu, Y., Chen, Z., 2016. A model for methane production in anaerobic digestion of swine wastewater. Water Res. 102, 464–474.
Yu, L., Wensel, P.C., Ma, J., Chen, S., 2013. Mathematical modeling in anaerobic digestion (AD). Bioremed. Biodegrad. 2155–6199.
Zhang, Q., Hu, J., Lee, D.-J., 2016. Biogas from anaerobic digestion processes: research updates. Renew. Energy 98, 108–119.
Zhou, W., Manousiouthakis, V.I., 2006. Non-ideal reactor network synthesis through IDEAS: attainable region construction. Chem. Eng. Sci. 61, 6936–6945.
Zhou, W., Manousiouthakis, V.I., 2007a. Pollution prevention through reactor network synthesis: the ideas approach. Int. J. Environ. Pollut. 29, 206–231.
Zhou, W., Manousiouthakis, V.I., 2007b. Variable density fluid reactor network synthesis – Construction of the attainable region through the ideas approach. Chem. Eng. J. 129, 91–103.
Zhou, W., Manousiouthakis, V.I., 2008. On dimensionality of attainable region construction for isothermal reactor networks. Comput. Chem. Eng. 32, 439–450.
Zhou, W., Manousiouthakis, V.I., 2009. Automating the AR construction for non-isothermal reactor networks. Comput. Chem. Eng. 33, 176–180.
Zinatizadeh, A.A.L., Mohamed, A.R., Najafpour, G.D., Hasnain Isa, M., Nasrollahzadeh, H., 2006. Kinetic evaluation of palm oil mill effluent digestion in a high rate up-flow anaerobic sludge fixed film bioreactor. Process Biochem. 41, 1038–1046.

Paper 4:

Attainable regions and fuzzy multi-criteria decisions: Modeling a novel configuration of methane bioreactor using experimental limits of operation

A paper published in ***Bioresource Technology***

Citation:

ABUNDE NEBA, F., ASIEDU, N. Y., ADDO, A. & SEIDU, R. 2020. Attainable regions and fuzzy multi-criteria decisions: Modeling a novel configuration of methane bioreactor using experimental limits of operation. ***Bioresource Technology*,** 295, 122273.

Highlights:

- A framework for simultaneous synthesis of digester structures and selection of digester subunits is presented
- Optimal digester structures can be synthesized only with experimental measurements biogas yield no kinetic model is required
- Laboratory experiments on anaerobic treatability are conducted using abattoir effluent as feed stock
- The AHP-fuzzy TOPSIS technique is introduced for selection of digester subunits based on the characteristic of the individual digesters
- For the feedstocks considered, optimal batch operation involves 2 batch with intermittent semi-batch digesters
- Continuous mode operation requires a continuous stirred tank digester with bypass from feed followed by an anaerobic baffled digester
- A novel attainable-region-inspired digester prototype is modelled for construction and testing

Bioresource Technology 295 (2020) 122273

Contents lists available at ScienceDirect

Bioresource Technology

journal homepage: www.elsevier.com/locate/biortech

Attainable regions and fuzzy multi-criteria decisions: Modeling a novel configuration of methane bioreactor using experimental limits of operation

F. Abunde Neba[c,d,*], Nana Y. Asiedu[b], Ahmad Addo[a], Razak Seidu[d]

[a] Department of Agricultural and Biosystems Engineering, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana
[b] Department of Chemical Engineering, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana
[c] Department of Civil and Environmental Engineering, Norwegian University of Science and Technology, Trondheim, Norway
[d] Institute for Marine Operations and Civil Engineering, Norwegian University of Science and Technology, Ålesund, Norway

ARTICLE INFO

Keywords:
Attainable regions
Multi-criteria decision methods
Multi-stage anaerobic digestion
Novel digester prototype
Abattoir waste

ABSTRACT

This study sets out to develop an approach that couples attainable regions and fuzzy multicriteria decision methods for modeling optimal configurations of multistage digesters without using a kinetic model of the process. The approach is based on geometric analysis of methane curves as their shapes contain valuable insight into substrate biodegradability characteristics during anaerobic digestion. With the case study of abattoir waste, the results indicate that the optimal batch operation policy involves four anaerobic sequencing batch reactors operated in series with fresh feed being added at the second and the four stages (fed-batch systems). For continuous mode operation, the optimal configuration involves a continuous stirred tank digester with bypass from feed followed by an anaerobic baffled digester, which has been used to obtain a novel prototype. The methodological framework presented in this study can be adopted to enhance design of multistage anaerobic digesters especially when reliable kinetic models are unavailable.

1. Introduction

The anaerobic treatment process has increasingly been recognized as an efficient technology for sustainable nutrient recycling, renewable energy generation and waste sanitation, having a strong potential to mitigate current energy resource and climate change challenges. However, the success of an industrial-scale anaerobic digestion is only possible if the following two prerequisite factors are met: (1) Availability of a sustainable supply of organic feedstock and (2) Design of optimal process configurations that are well adapted to the characteristics of the feedstock of interest. Concerning the second requirement, a wide variety of anaerobic digester systems have been developed, which can be classified in to three groups: conventional digesters (e.g. ASBR, CSTR, and PFR), sludge retention digesters (e.g. ACR, UASB, UASSR, ABR and ICR) and membrane digesters (e.g. AF, EGSB and AFBR) (Mao et al., 2015). Recent studies continue to develop new digesters, which either modify the principle of an existing digester technology or present novel features, all geared towards improving process performance (Pan et al., 2019; Terboven et al., 2017; Xiong et al., 2019).

Although various digester systems exist, each with different physical and geometric characteristics, the hydrodynamic configurations of all digesters can be derived from different combinations of three fundamental regimes: flow regime, mixing regime and sludge retention regime. Under flow regime, anaerobic digesters can be operated as batch, fed-batch or continuous; under mixing regime, they can be operated as completely mixed or with no axial mixing and under sludge retention regime, the operation can be with or without sludge retention. For example, a continuous flow regime operated with no axial mixing and no sludge retention gives a plug flow anaerobic digester (PFR) and when operated with sludge retention can result in either AF, ABR, UASB or EGSB. It is also important to mention that batch and fed-batch reactors can be operated as completely mixed or unmixed depending on the practical considerations.

Ming et al. (2016) illuminated critical aspects concerning a plug flow and a continuous stirred tank reactor focusing on mixing and reaction. By their analogy of a plug flow reactor (reactor with no axial mixing along the length) as a series of batch reactors (reacting vessels) travelling on a conveyor belt, the Plug flow reactor can be considered a reaction reactor. The authors also illustrated that CSTR operates directly opposite to the PFR with respect to mixing due to its perfect mixing assumption where conversion of reactants into product is assumed to occur as a result of mixing and dilution rather than from reaction alone. The PFR and CSTR are therefore at the extremes of mixing

* Corresponding author at: Department of Civil and Environmental Engineering, Norwegian University of Science and Technology, Trondheim, Norway.
E-mail address: fabricen@stud.ntnu.no (F. Abunde Neba).

https://doi.org/10.1016/j.biortech.2019.122273
Received 24 July 2019; Received in revised form 14 October 2019; Accepted 15 October 2019
Available online 17 October 2019
0960-8524/

Nomenclature

V_g	volume of biogas produced (mL)
m_S	mass of substrate added to the digester (g)
y_t	cumulative biogas yield (mL/g)
ABR	Anaerobic Baffled Reactor
AF	Anaerobic Filter
AFBR	Anaerobic Fixed Bed Reactor
AHP	Analytical Hierarchy Process
APFR	Anaerobic Plug Flow Reactor
AR	Attainable Regions
ASBR	Anaerobic Sequencing Batch Reactor
CSTR	Continuous Stirred Tank Reactor
DSR	Differential Sidestream Reactor
EGSB	Expanded Granular Sludge Bed
F TOPSIS	Fuzzy Technique for Order of Preference by Similarity to Ideal Solution
FBDR	Fixed Bed Disc Reactor
ICR	Internal Circulating Reactor
MCDM	Multi-criteria decision-making
PFR	Plug Flow Reactor
TOPSIS	Technique for Order of Preference by Similarity to Ideal Solution
UASB	Up flow Anaerobic Sludge Bed
UASSR	Up flow Anaerobic Solid-State Reactor
α	mixing ratio (–)
τ	Residence time (days)

and reaction and different combinations of these digesters will provide different extents of mixing and reaction in a reactor system (or reactor network) made up of both reactor types.

Generally, digester technologies can be broadly classified into high-rate (having separate solids and hydraulic retention times) and low-rate (having coupled solid and hydraulic retention times systems (Mes et al., 2003). In essence, all high rate digesters (sludge retention and membrane reactors) provide a mechanism of sludge separation in addition to the mixing and/or reaction. For example the anaerobic contact reactor provides mixing (due to presence of a CSTR) and separation while the anaerobic baffled reactor provide reaction (because the ABR operates in plug flow mode) and separation (Abunde et al., 2019b). What differentiates the high rate digesters is the mechanism in which sludge separation is performed, which can either be through fixed microbial films on solid surfaces or through an external separation and recycle (Mes et al., 2003). As expected, the different mechanisms result in different extents of separation, each of which is more suited for different substrates and operational characteristics than others. Therefore, irrespective of the type of digester technology, the performance of the anaerobic treatment process depends on three fundamental processes, mixing (performed by CSTR) reaction (performed by PFR) and separation (performed by high rate systems). What this means is that instead of focusing attention to devise new or perhaps novel digesters with the aim of improving the systems performance, it would be more important to focus attention on optimally arranging combinations of PFR, CSTR and/or high rate systems, or integrating more fundamental processes to the anaerobic treatment process (e.g. reversed osmosis + anaerobic digestion). This is referred to as the so called multi-stage anaerobic digestion in which every step of the anaerobic treatment process is optimized by operating digesters in a network in a network (EPA, 2006).

Several studies exist operating anaerobic digestion by coupling two or more types of digesters in multiple stages. Some examples include: AF + UASB (Lew et al., 2004), UASB + AF + AF (Chernicharo and Machado, 1998) CSTR + UASB (Aslanzadeh et al., 2014), ABR + AF (Mang and Li, 2010) CSTR + CSTR (Gaby et al., 2017), etc. The cited studies and many other practical examples of coupled digester systems are usually designed using an empirical approach, where candidate digester configurations are predefined at the start followed by experimental evaluations to select the configuration that yields the best performance. This strategy is not only expensive and time-consuming, but also limited to series combination of digesters, without any systematic way to determine the number and type of digester subunits or how the individual digester subunits should be connected. In addition, other combinations (e.g. parallel or both parallel and series) of the fundamental anaerobic digester types can always be derived, which can have similar or even improved performance than the series combinations (hence problem of multiple solutions). The authors' recent studies, have been first to lay down the theoretical framework for use of attainable regions (AR) in solving the problem of multiple solutions during synthesis of anaerobic digester networks (Abunde et al., 2019b,a). However, the attainable region approach used for modeling configurations of anaerobic digester networks provides a global optimal structure consisting of digesters operated in a plug flow or continuous stirred mode (sometimes involving bypass and or recycle streams) (Ming et al., 2013, Hildebrandt and Glasser, 1990) but provides no information about the type or nature of the individual digesters. This therefore poses another challenge on the choice of plug flow digester to use considering that there exist several digesters that can be considered to have a plug flow mode of operation. This study has therefore been designed to develop a novel methodological framework that couples attainable region theory (for reactor network synthesis) and a fuzzy multicriterial decision making method (for optimal selection of subunits for a digester network configuration) for optimal synthesis of anaerobic digester structures. Another interesting aspect of the study is that unlike previous studies that require a reliable kinetic model before AR can be applied to synthesize anaerobic digesters, the framework presented in the current study only requires experimental data for synthesis of digester network configurations.

2. Methods

2.1. Attainable region synthesis of anaerobic digester networks

The Attainable Region (AR) theory is a technique that incorporates elements of geometry and mathematical optimization, to design and improve operation of chemical reactors (Hildebrandt and Glasser, 1990). The power of the AR approach to process optimization is that the answer to all possible optimization problems, even the ones not considered are first determined, before looking for ways of achieving that answer. In reactor operation knowledge of all possible reactor states for all possible reactor configurations, even those that have not yet been devised, is obtained. The convex hull for the set of all points achievable by all possible combinations of CSTR + PFR defines the attainable region. The convex hull is understood as the smallest subset of a set of points that can be used to generate all other points by reaction and mixing (Ming et al., 2016). Geometrically, a convex hull is a finite convex polytope enclosed by a finite number of hyperplanes, which is interpreted in a two- dimensional space as the smallest polygon enclosed by planar facets such that all of the elements lie on or in the interior of the polygon (Asiedu et al., 2015). Once the AR has been determined, the limits of achievability by the system for the given kinetics and feed point is known, which can then be used to answer different design or optimization questions related to the system.

Given a system, the following needs to be performed to do an AR analysis

- Define the fundamental processes occurring within the system

- Determine the state variables used to construct the AR
- Define the geometry of the fundamental reactor subunits
- Generate the AR using combinations of the fundamental processes
- Interpret the AR boundary in terms of reactor structures
- Define and overlay an objective function onto the AR boundary
- Determine the specific reactor configuration required to achieve the intersection point

The last two bullet points are essential if the attainable region is to be used to determine a specific design or optimization question. It is not the focus of the paper to present a deep theory of the AR concept. Interested readers can consult the cited literature for a more in-depth theoretical background (Ming et al., 2016).

2.2. Selection of plug for anaerobic digesters

2.2.1. Formulation of digester selection problem

As mentioned in Section 1, there are several anaerobic digesters (UASB, EGSB, AF, ABR, etc.) that can be considered to have a plug flow mode of operation (hence no axial mixing), selecting the appropriate plug flow digester becomes a challenging task. After a detailed literature survey, the most common plug flow anaerobic digesters were selected as candidates for the multicriteria decision making. These include: Anaerobic Fluidized Bed Reactor (AFBR), Anaerobic Plug Flow Reactor (APFR), Expanded Granular Sludge Bed (EGSB), Internal Circulating Reactor (ICR), Up flow Anaerobic Sludge Bed (UASB), Anaerobic Baffled Reactor (ABR) and Anaerobic Filter (AF). Therefore, the approach proposed in this paper relies on a modular coupling of the geometric technique of attainable regions followed by the multicriteria decision making tools. It is worth mentioning that the idea is not to explore all the existing types of plug flow digesters, but to present a framework for selection of plug flow digesters that will accompany the optimal structure defined by the attainable regions. Several criteria have been defined for use in evaluating the digester alternatives as described in Table 1. The next section of the paper will present the fuzzy multi-criteria decision-making process for selection of the most appropriate plug flow anaerobic digester using anaerobic treatment of abattoir waste as the case study.

2.2.2. Fuzzy multicriteria decision making process

The use of ordinary Multicriteria decision making (MCDM) tools for ranking of alternatives requires that the performance score of the alternatives with respect to each criterion is quantitative in nature (i.e. can be measured and attributed a crisp numerical value). An example is the work of Karagiannidis and Perkoulidis (Karagiannidis and Perkoulidis, 2009), quantitative characteristics, which can be measured (such as greenhouse gas emitted, recovered energy, recovered nutrients, operating cost, etc.) for selection of anaerobic digester technologies. However, in this study the performance score of the alternatives with respect to each criterion did not have crisp numerical values and the ordinary MCDM cannot therefore be applied. The strength of this study is illustrated by extending the decision-making process to include fuzziness, where by ratings of alternatives versus criteria is done using linguistic variables represented as triangular fuzzy numbers (TFN). The linguistic variables and their corresponding TFN (presented in parenthesis) utilized include: very poor (1,0,0), poor (3,1,1), medium poor (5,3,3), fair (7,5,5), medium good (9,7,7), good (10,9,9) and very good (10,10,10). This provides an opportunity of the decision-making process to be performed even in cases where crisp numerical ratings of the alternatives with respect to the criteria is not available or even in cases of uncertainty.

The selection of the appropriate anaerobic plug flow digester was done using a hybrid of the Fuzzy Technique for Order Preference by Similarity to Ideal Solution (FTOPSIS) and the Analytical Hierarchy Process. At first, AHP is used to compute the criteria weights, which show the relative importance of the different attributes used for digester selection. Afterwards, the FTOPSIS method is applied to prioritize the different alternatives (plug flow digesters) based on the computed criteria weights (Esmaili Dooki et al., 2017, Balioti et al., 2018, Basahel and Taylan, 2016). Assuming there exist m alternatives and n criteria/attributes, the algorithm for the integrated AHP-Fuzzy TOPSIS method utilized in this study is summarized in the following 6 steps:

Step 1: Construct the decision matrix. The performance value of each alternative with respect to each criterion is determined using a fuzzified seven-point scale. The seven-point scale is fuzzified using a triangular membership function, where each linguistic term is expressed in positive triangular fuzzy numbers. In case of multiple decision makers, each decision maker attributes a linguistic label on all alternatives with respect to each criterion and Eq. (1) is used to compute the combined positive triangular fuzzy numbers for all the decision makers.

$$a_{ij} = \min_{k}\{a_{ij}^{k}\} \tag{1a}$$

$$b_{ij} = \frac{1}{K}\sum_{k=1}^{K} b_{ij}^{k} \tag{1b}$$

$$c_{ij} = \max_{k}\{c_{ij}^{k}\} \tag{1c}$$

Table 1
Set of decision criteria to evaluate plug flow anaerobic digesters.

Symbol	Name of Criteria	Objective	Description
C1	COD/VS Reduction Efficiency	Maximize	This measures the ability of an anaerobic digester to reduce organic pollution
C2	Retention of Residual Nutrients	Minimize	High nutrient retention by anaerobic digestate can result in eutrophication when disposed to the environment. The objective is to maximize biogas production and not nutrient recovery
C3	Total Solids content in the Digester	Minimize	This parameter differentiates between wet fermentation (15–25%T) and dry fermentation (> 30%). Wet digesters that are more adapted to minimal TS because the substrate (abattoir waste) fall in the range of wet fermentation (TS = 17.5%)
C4	Organic Loading Capacity	Maximize	This measures the processing rate of organic matter for a given anaerobic digester type. Higher values are economically attractive
C5	Axial Mixing	Minimize	Digesters with plug flow operation (PFRs, UASB, AFs, ABRs, etc.) offer a higher processing capacity to microorganisms and hence higher degree of biodegradation. This is because such systems present little or no axial mixing of the digester content during operation
C6	Biogas Yield	Maximize	Biogas is a renewable energy, which can be used as substitute for fossil-based fuels
C7	Stage of Treatment (Primary or Secondary)	Maximize	Nutrient removal, hygienisation and COD reduction mostly occur in the secondary treatment
C8	Thermal stability of the system	Maximize	Small-scaled digester systems mostly operate under non-isothermal conditions. Hence system that are less sensitive to temperature variations are more attractive

Step 2: Construct a normalized fuzzy decision matrix. This was done by calculating a normalized fuzzy performance value of each alternative with respect to each criterion. For the benefit criteria, a maximum value is desired while for the cost criteria, a minimum value is desired. The normalized fuzzy performance ratings for the alternatives with respect to the benefit and cost criteria was done using Eqs. (2) and (3) respectively.

$$\tilde{r}_{ij} = \left[\frac{a_{ij}}{c_j^*}, \frac{b_{ij}}{c_j^*}, \frac{c_{ij}}{c_j^*}\right], \quad c_j^* = \max_i\{c_{ij}\}(benefit\ criteria) \tag{2}$$

$$\tilde{r}_{ij} = \left[\frac{a_j^-}{c_{ij}}, \frac{a_j^-}{b_{ij}}, \frac{a_j^-}{a_{ij}}\right], \quad a_j^- = \min_i\{a_{ij}\}(cost\ criteria) \tag{3}$$

$where\ j = 1, 2, 3\cdots\cdots..n\ and\ i = 1, 2, 3\cdots\cdots..m$

Step 3: Construct a weighted normalized fuzzy decision matrix by multiplying the normalized TFNs with the weight of each criteria as shown in Eq. (4)

$$\tilde{v}_{ij} = \tilde{r}_{ij} \times w_j \tag{4}$$

The weights of relative importance (w_j) of each of each criterion were determined using the AHP. A pairwise comparison matrix, A using a scale of relative importance was then constructed whereby an attribute compared with itself is always assigned the value 1. The numbers 3, 5, 7, and 9 correspond to the verbal judgments "moderate importance", "strong importance", "very strong importance", and "absolute importance".

The criteria weight vector $W = [W_1, W_2, \ldots W_N]$ was determined using these two steps:

- Normalize the pair-wise comparison matrix, A_{norm} by dividing each entry in A_{norm} column i of A by the sum of the entries in column i.
- The W_i was estimated as the average of the entries in row i of A_{norm}.

The pair-wise comparison matrix is then subjected to consistency check, which involves determination of the maximum Eigen value, Eq. (5) and the consistency Index (CI), Eq. (6).

$$\lambda_{max} = 1/n \sum\nolimits_{i=1}^{n} \frac{i^{th}\ entryin\ AW^T}{i^{th}\ entry\ in\ W^T} \tag{5}$$

where

λ_{max} = maximum Eigen value
n = number of attributes
A = pairwise comparison matrix
W = The estimate of the decision-maker's weight

$$CI = \frac{\lambda_{max} - n}{n - 1} \tag{6}$$

Consistency is checked by comparing the consistency Index to the Random Index (RI) for the appropriate value of n, used in decision-making (Saaty, 2000). If $(CI/RI) < 0.10$, the degree of consistency is satisfactory, but if $(CI/RI) > 0.10$, serious inconsistencies may exist, and the results produced by AHP may not be meaningful.

Step 4: Determine the fuzzy positive ideal solution (FPIS) and fuzzy negative ideal solutions (FNIS). For benefit attributes, the ideal best value of all alternatives with respect to a given attribute is the maximum while negative ideal is the minimum weighted normalized fuzzy performance value. The FPIS and FNIS are computed by Eqs. (7) and (8) respectively.

$$A^* = (\tilde{v}_1^*, \tilde{v}_2^*, \ldots.\tilde{v}_n^*) \quad where\ \ \tilde{v}_j^* = \max_i\{v_{ij3}\} \tag{7}$$

$$A^- = (\tilde{v}_1^-, \tilde{v}_2^-, \ldots.\tilde{v}_n^-\ \ ) \quad where\ \tilde{v}_j^- = \min_i\{v_{ij1}\} \tag{8}$$

Step 5: Calculate the separation measurement Euclidean distance of each alternative from the FPIS and FNIS. The distance from FPIS (S_i^+) is computed using Eq. (9) while the distance from FNIS (S_i^-) is computed using Eq. (10). The Euclidean distance between two triangular fuzzy numbers, $\tilde{a}_{ij} = (a_{1ij},\ a_{2ij},\ a_{3ij})$ and $\tilde{b}_{ij} = (b_{1ij}, b_{2ij}, b_{3ij})$ is given by Eq. (11)

$$S_i^+ = \sum_{j=1}^{n} d(\tilde{v}_{ij}, \tilde{v}_j^*) \tag{9}$$

$$S_i^- = \sum_{j=1}^{n} d(\tilde{v}_{ij}, \tilde{v}_j^-) \tag{10}$$

$$d(\tilde{a}_{ij}, \tilde{b}_{ij}\ ) = \sqrt{\frac{1}{3}(a_{1ij} - b_{1ij})^2 + (a_{2ij} - b_{2ij})^2 + (a_{3ij} - b_{3ij})^2} \tag{11}$$

Step 6: Determine the relative closeness or performance score (CC^*) of each alternative, Eq. (12). The alternatives are then ranked based on their performance score with respect to the ideal solution.

$$CC^* = \frac{S^-}{S^+ + S^-} \tag{12}$$

2.3. Experimental edge

2.3.1. Substrate sampling and characterization

The Anaerobic Digestion experiment was conducted at the Department of Agricultural and Biosystems Engineering, Kwame Nkrumah University of Science and Technology (KNUST), Ashanti Region of Ghana. It is located within 06° 41′ 5.67″ N 01° 34′ 13.87″ W. The abattoir waste was obtained from the Kumasi central abattoir. Seed

Table 2
Characteristics of abattoir effluent used for anaerobic treatability studies.

Elemental characteristics (ppm or mg/L)										
Ca	Mg	S	P	Fe	Cu	Zn	Ni	Mn	K	N
0.10	0.74	0.50	0.4	114.6	9.1	39.19	0.04	22.9	1.25	2.02

Biochemical characteristics					
Protein (% DM)	Crude fiber (%DM)	Carbohydrates (% DM)	Total Ash (%DM)	Fats (% DM)	BOD (mg/L)
27.6	13.96	44.48	3.926	2.25	520

Physicochemical characteristics					
Volatile solids (%)	Total solids (%)	Moisture (%)	Total alkalinity (mg$CaCO_3$/L)	Total dissolve solids (mg/L)	COD (mg/L)
87.41	17.515	82.49	1650	220	740

sludge used to facilitate start-up of the digestion process was obtained from a 40 m^3 fixed dome digester fed with faecal matter and located at the Kumasi Institute of Tropical Agriculture. To better understand the intrinsic nature of the abattoir waste, the elemental, biochemical and physiochemical characteristics were determined as presented in Table 2. Moisture content was determined using oven drying method 105 °C while the Total solids (TS) and volatile solids (VS), total dissolved solids (TDS) chemical oxygen demand (COD) biological oxygen demand (BOD) and total alkalinity were determined following the standard methods (APHA, 1998). The analyses of crude fiber (CF), crude protein (CP), crude fat (ether extract) (TF), ash and nitrogen free extract (NFE) were performed following the methods detailed in (AOAC, 1990). Total carbohydrate (TC) was calculated by using values obtained for CF and NFE (TC = CF + NFE). The quantification of heavy metals was done using an absorption spectrophotometer located at the crop research institute, Kumasi, Ghana.

2.3.2. Experimental setup and procedure

4.5 kg of substrate and 0.5 kg of inoculum and 1 L of water was mixed using a paddle and fed into the digester. Anaerobic digestion was performed in a 5 L batch reactor with a total digestion time of 30 days. The digester was insulated with a black polyethene sheet and the system was operated under an average room temperature of 31 °C. A 0.5 L changeable gas collection bag was connected to the digester using a drip set and a silicone sealant was used to make the connection airtight in order to ensure anaerobic conditions exist in the system. The digester was agitated everyday by shaking in order to prevent the formation of surface crust which may prevent contact between the anaerobic microorganisms and the substrate. The daily volumetric gas production was measured everyday using the water displacement method.

3. Results and discussion

3.1. Experimental studies and attainable region construction

Fig. 1 presents the dynamics of cumulative biogas yield obtained from anaerobic treatment of abattoir waste. The curve has a sigmoidal shape, which is characteristic of easily degradable substrates that are prone to some degree of inhibition (Labatut et al., 2011). The interest is not necessarily on the shape of the biogas yield curve, but on how the authors use the curve to synthesize digester structures to minimize digestion time. The design of the optimal digester structure to minimize digestion time involves three main aspects: (1) Construction of attainable regions using geometric techniques, (2) scheduling of batch operation from the attainable regions, and (3) interpretation of continuous mode operation structures from the batch operation.

3.1.1. Construction of attainable regions

The optimization of digestion time using the attainable region technique is done using three major steps and Fig. 2 presents the plots obtained at the different stages of the construction process.

Step 1: Construction of base trajectory

In AR convention, when dealing with data involving residence time space, it is often conventional to plot residence time on the vertical axis while concentration or yield is plotted on the horizontal axis. Fig. 2(a) presents the cumulative biogas yield curve plotted in AR convention and the curve ABCD is called the base anaerobic digestion trajectory.

Step 2: Determine and bypass concavity using a mixing line

Observe that the base anaerobic digestion trajectory given by curve ABCD, is concave with respect to residence time axis, which may be filled by joining points A and C with a mixing line as shown in Fig. 2(b). The location of ABCD on the curve is done as follows: Firstly, determine point A (usually the starting point or feed point. Secondly, identify the region of concavity (on the lower side of the residence time axis) and locate another point, C such that a line drawn from A to the point C fills the concavity. Thirdly, the segment of the curve between A and C is called B.

The straight-line AC has a very significant property. A key criteria for selecting variables in AR is that they must obey the linear mixing law (Hildebrandt et al., 1990). It can be shown that the residence time of a system must lie in a straight line between the residence times of the individual reactors, τ_1 and τ_2 comprising the system (Ming et al., 2016). This implies the residence time obeys the linear mixing law, Eq. (13)

$$\tau_{mix} = \alpha\tau_1 + (1 - \alpha)\tau_2 \tag{13}$$

The cumulative biogas yield (y_t) is given by the volume of biogas produced (mL) per mass of substrate added to the digester (g). $y_t = V_g/m_S$. Consider two digesters of known biogas yield, the actual volume of biogas produced for digesters 1 and 2 can be obtained by $V_{g1} = y_{t1}m_{S1}$ and $V_{g2} = y_{t2}m_{S2}$ respectively. Conservation of mass may be used to calculate the total cumulative biogas yield for both digesters. Conservation of mass ensures that the total mass of substrate in the mixture is equal to the sum of the individual substrate masses contained in digesters 1 and 2, which is given by $m_{ST} = m_{S1} + m_{S2}$. Computing the biogas yield of the entire system is equivalent to determining the biogas yield for a mixture of digesters 1 and 2 because the density of the liquid phase of the digester can be assumed constant. The biogas yield of the mixture is given by the ratio of the total volume of biogas produced to the total mass of organic substrate added as shown by Eq. (14).

$$y_{tM} = \frac{y_{t1}m_{S1} + y_{t2}m_{S2}}{m_{ST}} \tag{14}$$

By setting $\alpha = m_{S1}/m_{ST}$ then Eq. (14) can be written as Eq. (15), which is similar to the linear mixing law. What this means practically is that by mixing the contents of the liquid phase of two digesters, each of which contains a given mass of organic substrate, then the total cumulative biogas yield of the mixture will lie in a straight line joining that of both digesters.

$$y_{tM} = \alpha y_{t1} + (1 - \alpha)y_{t2} \tag{15}$$

This is known as the lever-arm rule and the process of combining the contents of two parallel digesters of different substrate masses results in a linear mixing law (where α is known as the mixing ratio) measured in term of cumulative biogas yield.

The point A on the curve represents a digester condition where a fresh mass of substrate has just been added and no biogas has been produced. The straight-line AC therefore represents a batch digester, which is run up to a certain residence time then the content is mixed with fresh substrate. Because the base anaerobic trajectory lies higher up on the residence time axis than the mixing line AC, bypassing fresh

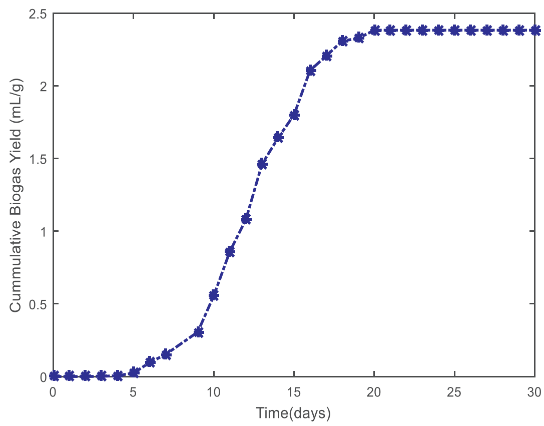

Fig. 1. Cumulative biogas yield curve for anaerobic digestion of abattoir waste.

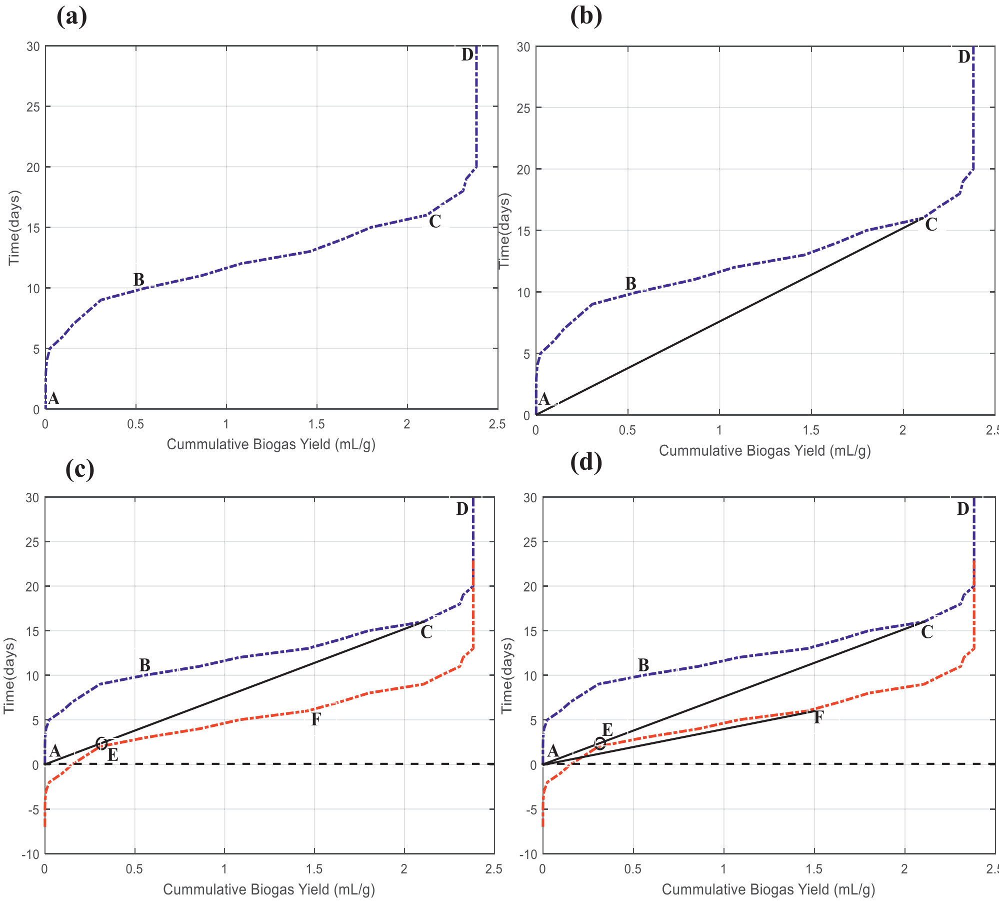

Fig. 2. Attainable region construction process (a) Base anaerobic digestion trajectory in AR convention. (b) Base anaerobic digestion trajectory showing mixing line. (c) Moving down the based trajectory until it touches the mixing line. (d) Generating a candidate AR using only PFRs and a base trajectory.

organic substrate reduces the overall residence for the same cumulative biogas yield (this is only for yields between points A and C). For example, on the initial anaerobic digestion trajectory, observe that a residence time of 10 days is needed to achieve a cumulative biogas yield of 0.5 mL/g, meanwhile the same yield can be achieved at 5 days using the mixing line. This is possible by operating the batch digester up to point C and then mixing fresh substrate with this stream to obtain the desired overall yield. Note that this optimization is only possible because of the concavity in the original anaerobic digestion trajectory, and hence regions of low digestion rate in the digester are to be bypassed by the use of mixing. This phenomenon can be attributed to the fact that adding fresh substrate increases nutrient bioavailability for the anaerobic microorganisms thereby increasing growth and hence production of the desired biogas

Step 3: Expansion of candidate attainable regions using batch trajectory and the mixing line

Notice from step 2 how graphical techniques have been applied to expand the total set biogas yields that is achievable in the anaerobic digester by making use of concavities in cumulative biogas yield curves. Furthermore, from the principles of differential algebra, process trajectories from batch reactors are directional. Geometrically, the reaction rate vectors of batch processes have a unique nature, which ensures that different batch trajectories progress in a manner that they do not cross one another (Asiedu et al., 2014). For a given feed point there exists a unique trajectory for a process operated in batch mode. The overall residence time for anaerobic digestion can be decreased by using the base anaerobic digestion trajectory. This is done by moving the trajectory down until it touches the mixing line at a unique point (point E) as shown in Fig. 2(c). This point of contact (point E) has a significant geometric and practical meaning for further optimization of the anaerobic digestion process. Geometrically, it represents the point where the reaction rate vectors point out of the boundary of the attainable region, which means the region can further be expanded from that point in order to meet the necessary condition of convexity (Glasser et al., 1993, Hildebrandt et al., 1990). Practically, it represents the lowest digestion time on the boundary of the candidate attainable region where from an additional batch digester can be initiated to further expand the region and minimize the residence time. By translating the curve downwards, the direction of the reaction rate vectors vary along the length of the mixing line. Observe that the shifted trajectory this has some small concavity with respect to residence time axis, which may be filled by joining points A and F with a mixing line as shown in Fig. 2(d). By translating the curve downwards, the direction of the reaction rate vectors varies along the length of the mixing line. When the attainable region becomes convex, it implies there is no part on the boundary of the attainable region where the rate vectors point outward, and this implies that the true attainable region has been obtained. The convex curve AFD represents therefore represents attainable region for the anaerobic treatment process. The attainable region represents all possible outputs that can be achieved for all possible reactor designs by interpreting chemical processes as geometric objects. Geometrically, it represents the convex hull for the set of points achievable by a given system.

3.1.2. Modeling process configurations

Fig. 3 presents the optimal process configurations of the anaerobic treatment process for both batch and continuous mode operation Sections 3.1.2.1 and 3.1.2.2 provides a detailed description of how the process structures have been obtained.

3.1.2.1. Scheduling of batch operation policy. After the attainable region has been obtained, the boundary of the attainable region can be used the schedule an operating policy for batch anaerobic digestion, which can be used to achieve the limits define by the system Point E is obtained by running a batch with fresh feed, up to the point C (stage 1) then mixing the content into another batch digester, which is then mixed with fresh organic waste (stage 1). This batch operation policy is illustrated in Fig. 3(a). Another batch is run with the contents of stage 2, stage 3 (from point E) to obtain the point F found on the EFD trajectory then mixed with fresh organic waste (stage 4) to obtain points located on line AF.

3.1.2.2. Continuous mode operation. The boundary of the attainable region can also be interpreted into continuous process configurations, which can be used to attain the same achievable limits defined by the attainable region (Hildebrandt and Glasser, 1990). This will be based on the final structure of the attainable region boundary defined by the curve AFD. The interpretation of the AR boundary is based on three key fundamental results of two-dimensional AR used in everyday practice (Ming et al., 2016). (1) The AR is composed of reaction and mixing surfaces only. Reaction surfaces are always convex. (2) Points that form convex sections of the AR boundary arise from effluent concentrations specifically from PFR trajectories. (3) Points on the AR boundary that initiate these convex PFR trajectories (from point 2 above) arise from specialized CSTRs for two-dimensional constructions. This implies that the point (F), arise from a CSTR and is used to imitate convex PFR trajectories to form the AFD trajectory. The mixing line AF, which eliminates the concavity in the system is represented structurally by a CSTR with a bypass from point A. The final structure of the continuous digester structure is shown in Fig. 3(b).

It is interesting to compare the results of this study with that of the authors' recent studies using attainable regions to synthesize anaerobic digester structures (Abunde et al., 2019a,b). The studies developed a simplified kinetic model of the anaerobic treatment process and applied the kinetic models to construct the attainable regions. The reliability of this approach depends on the availability of a suitable kinetic model as well as kinetic coefficients of the process. In the current study, the construction of attainable regions using only experimental data has been presented. This implies that even without having a kinetic model of the process, it is still possible to design optimal digester systems.

3.2. Selection of digester subunits and definition of optimal process configurations

As mentioned in Section 3.1, the attainable region defines optimal digester structures in terms of the mode of operation, which can be plug flow (with no axial mixing) or continuous (with mixing). Also, Fig. 1 illustrates that there exist several anaerobic digesters, which can be considered to have a plug flow operation. A list of criteria used to select the appropriate plug flow digester to enhance biogas generation has been presented in Table 1. Fig. 4 presents a spider web diagram showing the weights of the criteria (obtained using the analytical hierarchy process), which indicates the extent to which each criterion has on the selection of an anaerobic plug flow digester. Compared to other multi-criteria decision-making methods, the AHP is well known for its strength of weighting criteria, which is why the authors chose it for criteria weighting. The AHP determines the weight of importance of each criterion by using pair-wise comparison matrix that uses the scale of relative importance proposed by Saaty.

It should be noted that out of the three main applications of anaerobic digestion (waste hygienisation, renewable generation and nutrient recycling), the weighting has been focused on maximizing renewable energy generation potential of the process. Fig. 4 shows that biogas yield and stage of treatment carry the highest weights, followed

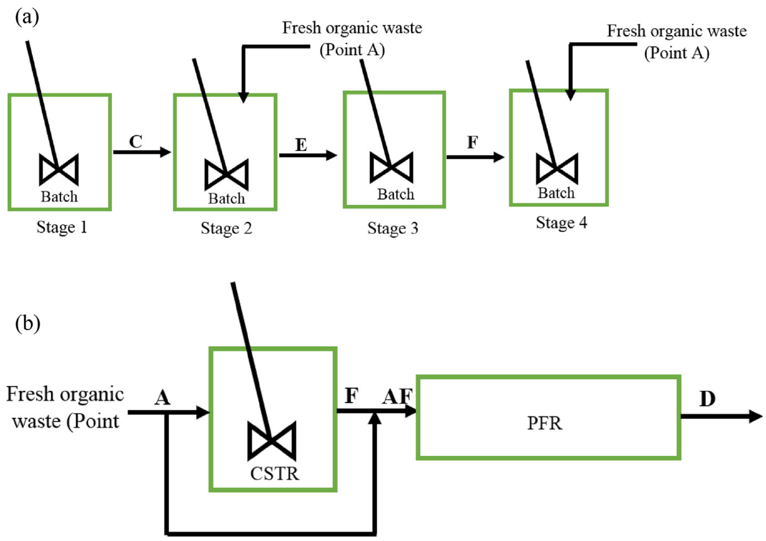

Fig. 3. Optimal process configurations of anaerobic treatment process (a) Optimal scheduling of operating policy for anaerobic batch digesters. (b) Optimal continuous digester structure of treatment of abattoir waste.

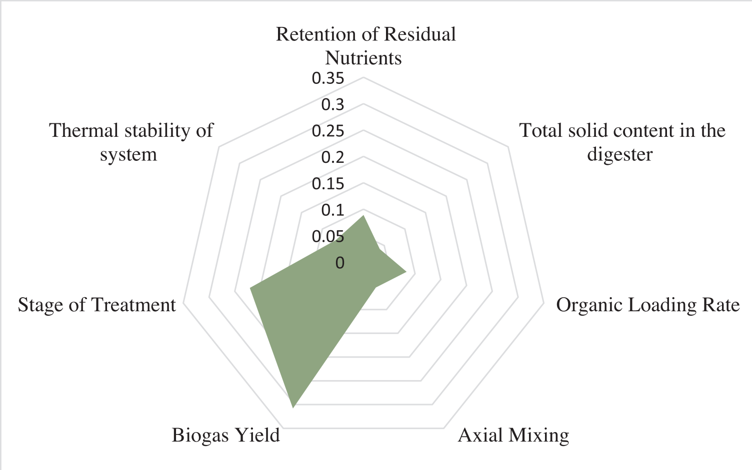

Fig. 4. Criteria weights for selection of anaerobic plug flow digester.

by organic loading rate and retention of residual nutrients while axial mixing, thermal stability and total solids content carry the smallest weights. This can be explained as follows: biogas yield is highest since the objective is to generate renewable energy and hence plug flow digesters more adapted to produce more biogas per gram of substrate are given priority. Regarding stage of treatment, anaerobic digestion can be operated as a primary or secondary treatment system and the primary systems are more adapted to biogas generation while the secondary systems are more adapted to waste hygienisation of nutrient recovery (Mang and Li, 2010). It is therefore important to select digesters that are more adapted to primary treatment. Organic loading rate carries relatively less weight because when anaerobic digestion is used for renewable energy generation, the system is dimensioned based on the energy requirements of the users and not on the flowrate of effluent available. The goal is not to digest all the effluent, but to digest the effluent that will produce the required quantity of energy. However, if the system is to be designed mainly for treatment of effluent, the dimensioning would base on the flow rate of effluent available, which must be treated to meet a given discharge standard. In the same light, the retention of residual nutrients is relatively less important as the goal is to maximise biogas generation. For the case of thermal stability, the relatively low weight is attributed to the fact that the digester will be operated under isothermal conditions and for and axial mixing, all the plug flow digesters are assumed to have no axial mixing, with little variations. Finally, for the case of total solids content, the digester is to be used for the treatment of effluent from abattoir with a defined solids content. It is important for readers to note that the authors are not saying some of the criteria are not important but are just explaining why some of the criteria are considered more important that others in the selection of the appropriate plug flow digester.

Therefore, according to the criteria weights, the ranking order of all the plug anaerobic digesters according to importance was determined using the fuzzy TOPSIS approach and the best alternative was selected among seven alternative anaerobic digesters. The ranking order of the anaerobic digesters based on the closeness coefficient to the ideal solution is given in Table 3. Table 3 indicates that out of the seven anaerobic plug flow digesters considered, the Anaerobic Baffled Reactor (ABR) had the best performance. The results are further strengthened by the findings presented by other researchers concerning the operational characteristics of the ABR. The ABR is a high rate anaerobic plug flow digester having a decoupled sludge and hydraulic retention times enabled by a series of vertical baffles through which effluent flows (Mao et al., 2015). The baffles divide the reactor into a series of compartments and forces incoming effluent to flow axially through a series of blanketed sludge trapped in each compartment. The ABR is therefore considered to be a multi-stage system consisting of several UASB connected in series. The separation of the biological steps within the system ensures overall improvement in performance, as each of the steps can be allowed to operate at their optimal conditions there by minimizing issues of toxicity (Bachmann et al., 1985, Barber and Stuckey, 1999). Recall that the optimal digester structure (Fig. 4b) obtained for the treatment of abattoir effluent consist of a CSTR with bypass of feed followed by a PFR, which will now be a CSTR with bypass of feed followed by an ABR. The optimal reactor configuration has been modelled in 3D as presented in Fig. 5(a). Fig. 5(b) shows a cross sectional view of the system indicating how the baffles have been designed while Fig. 5(c) shows a transparent view of the novel system.

Studies have shown that the optimal application of ABR is post-treatment after a primary treatment step (Mang and Li, 2010). On the other hand, the operation of a single CSTR is less efficient in terms of the biogas yield and hence effluent quality (Boe and Angelidaki, 2009). This further supports why the ABR coming after a primary digestion step using a CSTR is an optimal reactor structure. The novel prototype combines the advantages of a continuous stirred tank anaerobic reactor and an anaerobic baffled reactor.

The system is envisaged to operate as in three stages as follows: In stage 1, effluent is mixed and homogenized in a continuous stirred tank for a given retention time. This first stage has the advantage of rapid acidification due to mixing from continuous stirring, resulting in the production of high quantities of volatile fatty acids. The second stage involves bypass of fresh effluent to mix with the effluent from CSTR. As

Table 3
Importance ranks of anaerobic plug flow digesters fuzzy AHP-TOPSIS method.

	Positive ideal Solution (S^+)	Negative Ideal solution (S^-)	Relative closeness to ideal solution $S^-/(S^- + S^+)$	Rank	Digesters
A_1	2.301	1.910	0.453	6	AFBR
A_2	0.718	2.888	0.800	2	UPFR
A_3	2.254	1.304	0.366	7	EGSB
A_4	1.292	3.543	0.732	3	ICR
A_5	1.895	2.459	0.564	5	UASB
A_6	0.146	3.855	0.952	1	ABR
A_7	1.307	3.675	0.677	4	AF

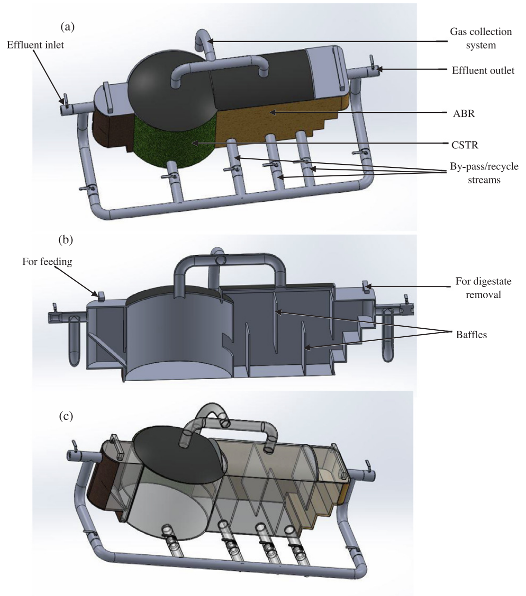

Fig. 5. Attainable region inspired novel prototype of an anaerobic digester.

demonstrated in Section 3.1, the bypass valve from feed has been systematically added based on the attainable region process to bypass regions of slow biodegradation increases the overall efficiency of the process. The third stage (ABR), which retains high amounts of sludge rapidly converts the volatile acids in to biogas. Observe from Fig. 5 that other valves have been included in the system meanwhile the optimal system for treatment of abattoir effluent includes only a single bypass stream. The authors' previous study on attainable regions indicated that the attainable region and hence the optimal digester structure is unique for each digested substrate (Abunde et al., 2019b). Hence a different organic substrate might require a change in the position of the bypass stream or the addition of a recycle stream. In such cases, the network configuration can be changed by simply opening and closing certain valves, which ensure robustness in the reactor structure for different substrates. This will be very helpful during experimental testing of the system where multiple substrates can be tested using the same prototype without the need to redesign a completely new system in cases where the configuration changes due to change in substrate. It is interesting to compare the conceptual operation of the novel prototype presented in this study with some of the multistage studies presented in the literature involving CSTR in the primary stage. Boe and Angelidaki (2009) confirmed that using a multi-stage system involving two CSTRs, an improvement in biodegradation efficiency and biogas generation is seen mainly after addition of the second stage. In another study using a CSTR as a primary stage and an up-flow anaerobic sludge blanket reactor as the second stage, the results showed that the two stage system is more stable at higher organic loading rates compared to a single stage involving only a CSTR (Aslanzadeh et al., 2014). Observe that in both cases, CSTR performs optimally when used as a first stage. However, a major drawback with the aforementioned and many other studies involving multistage digestion is that the digester configuration is often predefined at the start of the study with no systematic rule for answering the following key questions: (1) what type of digesters subunits to include in the network (2) how many individual digester subunits should be included (3) should the subunits be connected in series, parallel or both (4) should bypass or recycle streams be included and if yes where within the system? The main advantage of the presented prototype compared to other multistage systems in that it has been designed based on a systematic framework that uses experimental data, which contains necessary information about the kinetics of the process. In addition, by being a compact multistage system, the prototype can separate the acidogenic and methanogenic phases axially within the reactor (as with other multistage systems) but without the high cost and control problems normally associated with multistage systems.

Although the prototype is still to be subjected to experimental validation, it can be theorised to have the following advantages: simple design (relative to other multistage systems), low sludge generation (as much of the sludge is retained in the system), no requirement of biomass with special settling properties, no requirement of a special gas or sludge separation system as well as stability to organic shocks. A natural progression of the study will be to subject the prototype to experimental testing whereby it will be constructed and operated simultaneously with a conventional fixed dome system under similar experimental conditions. This will allow for the determination of optimal flow rates for the feed stream, bypass stream and effluent stream from the primary treatment stage.

A very interesting continuation of the current study with respect to the fuzzy decision-making aspect will be to consider other scenarios for use of anaerobic digestion technologies. The anaerobic digestion technology can be used for three main applications: Renewable energy generation, sustainable nutrient recycling as well as waste sanitation and different digester technologies are more adapted for one application than the other. This implies the ranking of the digester technologies using the fuzzy method will be different if the application of anaerobic digestion technology changes. This study has only focused on use of anaerobic digestion for renewable energy generation. It will be interesting if further studies could expand the fuzzy multicriteria decision to the other two applications of anaerobic digestion and compare the results for all three cases. More interestingly, because the method is novel and not very common in the field of anaerobic digestion, the ultimate research goal should be to integrate the methodological framework presented in this study into a web-based application, which can serve industry practitioners and researchers involved in design of anaerobic digester systems

4. Conclusion

A framework that couples attainable regions and fuzzy multicriteria decision making for modeling configurations of anaerobic digesters without use of a kinetic model has been developed. Taking a case study of anaerobic treatment of abattoir effluent, the optimal batch policy involves four anaerobic sequencing batch reactors operated in series with fresh feed being added at the second and the fourth stages (fed-batch systems). In the case of a continuous mode operation, the optimal digester structure involves a continuous stirred tank digester with bypass from feed followed by an anaerobic baffled digester, which has been modelled as a compact three-dimensional prototype.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

Our team expresses gratitude to the following institutions; The Brew-Hammond Energy Centre, KNUST Ghana, The Water and Environmental Engineering Group, NTNU Ålesund and the Abunde Sustainable Engineering Group (AbundeSEG) for its immense technical support.

Funding: This work was supported by EnPe-NORAD under the project Upgrading Education and Research Capacity in Renewable

Energy Technologies (UPERC-RET).

References

Abunde Neba, F., Asiedu, N.Y., Addo, A., Morken, J., Østerhus, S.W., Seidu, R., 2019a. Simulation of two-dimensional attainable regions and its application to model digester structures for maximum stability of anaerobic treatment process. Water Res., 114891.

Abunde Neba, F., Asiedu, N.Y., Addo, A., Morken, J., Østerhus, S.W., Seidu, R., 2019b. Use of attainable regions for synthesis and optimization of multistage anaerobic digesters. Appl. Energy 242, 334–350.

AOAC, 1990. Official Methods of Analysis, 15th ed. Association of Official Analytical Chemists (AOAC), Washington, DC.

APHA, 1998. Standard Methods for the Examination of Water and Wastewater, 20th ed. American Public Health Association (APHA), Washington, DC.

Asiedu, N., Hildebrandt, D., Glasser, D., 2014. Experimental simulation of a two-dimensional attainable region and its application in the optimization of production rate and process time of an adiabatic batch reactor. Ind. Eng. Chem. Res. 53, 13308–13319.

Asiedu, N., Hildebrandt, D., Glasser, D., 2015. Experimental simulation of three-dimensional attainable region for the synthesis of exothermic reversible reaction: ethyl acetate synthesis case study. Ind. Eng. Chem. Res. 54, 2619–2626.

Aslanzadeh, S., Rajendran, K., Taherzadeh, M.J., 2014. A comparative study between single- and two-stage anaerobic digestion processes: Effects of organic loading rate and hydraulic retention time. Int. Biodeterior. Biodegrad. 95, 181–188.

Bachmann, A., Beard, V.L., McCarty, P.L., 1985. Performance characteristics of the anaerobic baffled reactor. Water Res. 19, 99–106.

Balioti, V., Tzimopoulos, C., Evangelides, C., 2018. Multi-criteria decision making using TOPSIS method under fuzzy environment. Appl. Spillway Sel. 2, 637.

Barber, W.P., Stuckey, D.C., 1999. The use of the anaerobic baffled reactor (ABR) for wastewater treatment: a review. Water Res. 33, 1559–1578.

Basahel, A., Taylan, O., 2016. Using fuzzy AHP and fuzzy TOPSIS approaches for assessing safety conditions at worksites in construction industry. Int. J. Secur. Eng. 6, 728–745.

Boe, K., Angelidaki, I., 2009. Serial CSTR digester configuration for improving biogas production from manure. Water Res. 43, 166–172.

Chernicharo, C.A.L., Machado, R.M.G., 1998. Feasibility of the UASB/AF system for domestic sewage treatment in developing countries. Water Sci. Technol. 38, 325–332.

EPA U.S.E.P.A., 2006. Biosolids technology fact sheet, multi-stage anaerobic digestion. National Service Center or Environmental Publications (NSCEP).

Esmaili Dooki, A., Bolhasani, P., Fallah, M., 2017. An integrated fuzzy AHP and fuzzy TOPSIS approach for ranking and selecting the chief inspectors of bank: a case study. J. Appl. Res. Ind. Eng. 4, 8–23.

Gaby, J.C., Zamanzadeh, M., Horn, S.J.J.B.F.B., 2017. The effect of temperature and retention time on methane production and microbial community composition in staged anaerobic digesters fed with food waste. Biotechnol. Biofuels 10, 302.

Glasser, D., Hildebrandt, D., Godorr, S., Jobson, M., 1993. A geometric approach to variational optimization: finding the attainable region. IFAC Proc. Vol. 26, 451–457.

Hildebrandt, D., Glasser, D., 1990. The attainable region and optimal reactor structures. Chem. Eng. Sci. 45, 2161–2168.

Hildebrandt, D., Glasser, D., Crowe, C.M., 1990. Geometry of the attainable region generated by reaction and mixing: with and without constraints. Ind. Eng. Chem. Res. 29, 49–58.

Karagiannidis, A., Perkoulidis, G., 2009. A multi-criteria ranking of different technologies for the anaerobic digestion for energy recovery of the organic fraction of municipal solid wastes. Bioresour. Technol. 100, 2355–2360.

Labatut, R.A., Angenent, L.T., Scott, N.R., 2011. Biochemical methane potential and biodegradability of complex organic substrates. Bioresour. Technol. 102, 2255–2264.

Lew, B., Tarre, S., Belavski, M., Green, M., 2004. UASB reactor for domestic wastewater treatment at low temperatures: a comparison between a classical UASB and hybrid UASB-filter reactor. Water Sci. Technol. 49, 295–301.

Mang, H.P., Li, Z., 2010. Technology Review of Biogas Sanitation (Draft). GIZ, Eschborn.

Mao, C., Feng, Y., Wang, X., Ren, G., 2015. Review on research achievements of biogas from anaerobic digestion. Renew. Sustain. Energy Rev. 45, 540–555.

Mes, T.Z.D.D., Stams, A.J.M., Reith, J.H., Zeeman, G., 2003. Methane Production by Anaerobic Digestion of Wastewater and Solid Wastes. Bio-methane and bio-hydrogen: status and perspectives of biological methane and hydrogen production. Dutch Biological Hydrogen Foundation – NOVEM, The Hague, The Netherlands.

Ming, D., Glasser, D., Hildebrandt, D., 2013. Application of attainable region theory to batch reactors. Chem. Eng. Sci. 99, 203–214.

Ming, D., Glasser, D., Hildebrandt, D., Glasser, B., Metzer, M., 2016. Attainable Region Theory: An Introduction to Choosing an Optimal Reactor. John Wiley & Sons Inc, Hoboken, New Jersey.

Pan, S., Wen, C., Liu, Q., Chi, Y., Mi, H., Li, Z., Du, L., Huang, R., Wei, Y., 2019. A novel hydraulic biogas digester controlling the scum formation in batch and semi-continuous tests using banana stems. Bioresour. Technol., 121372.

Terboven, C., Ramm, P., Herrmann, C., 2017. Demand-driven biogas production from sugar beet silage in a novel fixed bed disc reactor under mesophilic and thermophilic conditions. Bioresour. Technol. 241, 582–592.

Xiong, Z., Hussain, A., Lee, H.-S., 2019. Food waste treatment with a leachate bed reactor: Effects of inoculum to substrate ratio and reactor design. Bioresour. Technol. 285, 121350.

Paper 5:

Self-optimizing attainable regions of the anaerobic treatment process: Modeling performance targets under kinetic uncertainty

A paper published in ***Water Research***

Citation:

ABUNDE NEBA, F., TORNYEVIADZI, H. M., ØSTERHUS, S. W. & SEIDU, R. 2020b. Self-optimizing attainable regions of the anaerobic treatment process: Modeling performance targets under kinetic uncertainty. ***Water Research***, 171, 115377.

Highlights:

- The concept of self-optimizing attainable regions is introduced, by modeling and propagating kinetic uncertainty onto the attainable regions
- Practical identifiability is used to improve model reliability by determined the subset of kinetic parameters that can be reliably estimated
- Monte-Carlo simulation procedure is used to model state uncertainty resulting from uncertainty in the settings for the unidentifiable kinetic coefficients
- Incorporating uncertainty reduces performance targets, defined by the attainable region but increases robustness
- The concept is useful for making economic decisions can now be made under conditions of uncertainty
- The framework can be applied to model other sources of process uncertainty such as temperature, inhibitions, substrate, etc.

Water Research 171 (2020) 115377

Contents lists available at ScienceDirect

Water Research

journal homepage: www.elsevier.com/locate/watres

Self-optimizing attainable regions of the anaerobic treatment process: Modeling performance targets under kinetic uncertainty

F. Abunde Neba [a, b, c, *], Hoese M. Tornyeviadzi [a, c], Stein W. Østerhus [b], Razak Seidu [c]

[a] Abunde Sustainable Engineering Group, AbundeSEG, Ghana
[b] Department of Civil and Environmental Engineering, Norwegian University of Science and Technology, Trondheim, Norway
[c] Department of Marine Operations and Civil Engineering, Norwegian University of Science and Technology, Ålesund, Norway

ARTICLE INFO

Article history:
Received 2 August 2019
Received in revised form
5 December 2019
Accepted 6 December 2019
Available online 10 December 2019

Keywords:
Self-optimizing attainable regions
Practical identifiability
Uncertainty quantification
Anaerobic digester synthesis

ABSTRACT

Despite the advantage of model-based design, anaerobic digesters are seldom designed using biokinetic models due to lack of reliable kinetic coefficients and/or systematic approaches for incorporating kinetic models into digester design. This study presents a systematic framework, which couples practical identifiability, uncertainty quantification and attainable region (AR) concepts for defining process performance targets, especially when reliable kinetic coefficients are unavailable. Within the framework, we introduce the concept of self-optimizing ARs, which define performance targets that results in near optimal operation in spite of variations in kinetic coefficients. Using the case of modified Hill model, only 3 out of the 6 model parameters (unidentifiable set) are responsible for the model prediction uncertainty. The uncertainty bands (mean, 10th percentile and 90th percentile) on the model states has been computed using the Monte Carlo Simulation procedure and attainable regions for the different levels of uncertainty has been constructed and the boundaries interpreted into digester structures. The self-optimizing attainable regions have been defined as the intersection region of the attainable regions corresponding to the mean, 10th percentile and 90th percentile. Incorporating uncertainty significantly reduces performance targets of the process but increases self-optimality in defining performance targets. Unlike the attainable region, which represents the limits of achievability for defined kinetics, the self-optimizing attainable region represents the set of all possible states attainable by the system even in cases of kinetic uncertainty. In summary, the concept of self-optimizing ARs provides a systematic way of defining process performance targets and making economic decisions under conditions of uncertainty.

1. Introduction

Anaerobic digestion (AD) of animal manure is of great importance to the waste treatment and bioenergy industries, since the biomethane produced is a promising renewable energy alternative to fossil fuels. The modelling of anaerobic treatment process is a mature research area, now with a strong shift from model development towards application development, aimed at solving various design and operational challenges. Various models have been constructed to describe the anaerobic treatment process and the key motivations for model development have mainly been operational analysis, technology development, as well as digester design (Batstone, 2006). The model-based design is particularly important as the capital cost for anaerobic digesters determined from design is a key motivation for implementers. The kinetics captured by AD models is highly important for an optimal digester design since operating conditions, volumetric gas production, process stability (Finn et al., 2013; Yu et al., 2013; Batstone, 2006; Kythreotou et al., 2014), as well as effluent quality can be predicted (Kythreotou et al., 2014; Yu et al., 2013). Despite the advantage of model-based design, anaerobic digesters are seldom designed using biokinetic models but rather based on a combination of hydraulic and organic loading, where the digester capacity is determined for a given loading rate, temperature regime, mixing, etc. (Wang et al., 2007). This is because the use of biokinetic models is highly dependent upon availability of kinetic coefficients (Batstone, 2006; Wang et al., 2007), but it is often difficult to get reliable kinetic parameters in practical operation, which results in kinetic uncertainty (and hence uncertain process performance) if the models are used for digester

* Corresponding author. Department of Civil and Environmental Engineering, Norwegian University of Science and Technology, Trondheim, Norway.
E-mail address: fabricen@stud.ntnu.no (F. Abunde Neba).

https://doi.org/10.1016/j.watres.2019.115377

design. Summarily, three main challenges can be mentioned with the use of models to design anaerobic digesters: (1) Lack of systematic approaches for incorporating process kinetics in digester design. (2) The reliability of some of the studies using kinetic models to guide design of anaerobic digesters is undermined by uncertainty existing in the in kinetic coefficients. (3) Existing model-based studies are limited to single stage digesters and operating the process as a single stage generally limits overall performance. This is supported by the fact that AD involves multiple bioreactions steps (each step catalyzed by a specific group of microorganisms) and when operated as a single stage, it limits the possible combination of pathways since the process conditions are only suitable for all microorganisms with no reaction being optimized (EPA, 2006). However, most of the modeling studies on AD have focused on the model development approach, techniques for parameter estimation, with less effort devoted to assessing model reliability (identifiability and uncertainty) or how to incorporate uncertainty in digester design and operation. A systematic approach for handling kinetic uncertainty in design of anaerobic digesters with focus on multi-stage anaerobic digesters networks as opposed to single stage systems will be a breakthrough in advancing model-based optimization of anaerobic digestion.

This study is therefore designed to develop a systematic model-based framework (Fig. 1) for performance targeting and synthesis of anaerobic digester networks, when reliable kinetic models are not available. The framework (Fig. 1) is realized in two phases, which may involve feedback checks at specific steps depending on the system performance after every step. In the first phase (model reliability assessment), one selects a kinetic model appropriate for a digested substrate of interest (e.g. solid waste, sludge, wastewater, etc.), assesses the model's reliability and quantifies the model prediction uncertainty resulting from kinetic uncertainty. In the second phase (self-optimizing design), one mainly defines the robust performance targets of the system considering the uncertainty bounds computed in phase 1. Phase 2 is based on the concept of attainable regions, which is a geometric optimization technique that is used for both performance targeting and reactor network synthesis (Hildebrandt et al., 1990). The AR is a collection of all possible output for all possible reactor designs by interpreting chemical processes as geometric objects that define a region of achievability without having to explicitly enumerate all possible design combinations (Hildebrandt and Glasser, 1990). Central to the AR concept is the availability of reliable kinetic models of all fundamental processes (e.g. biochemical, physicochemical, physical in the case of AD) occurring within the system. In particular, simplified kinetic process models are emphasized as the AR theory involves mixing and attainability of states through a relatively complex geometric and hydrodynamic analysis (Hildebrandt et al., 1990; Ming et al., 2016).

The novel idea presented in this study is that instead of using AR to define an optimal performance target, which can only be achieved some of the times (due to kinetic uncertainty), the authors define a near optimal performance target, which can be attained all the time. This however involves an acceptable loss in process performance resulting from the kinetic uncertainty. Anaerobic digester systems designed with such an acceptable loss in performance resulting from uncertainty in kinetic coefficients are referred to as self-optimizing. Self-optimizing operation also referred to as self-optimizing design/systems (Permin et al., 2016; Gausemeier et al., 2006) is when we can achieve an acceptable loss by using constant setpoint values for design/operation variables (e.g. temperature, kinetics, substrate characteristics, etc., for the case of anaerobic digestion) without the need to reoptimize when variations occur. In the case of this study, we define self-optimizing

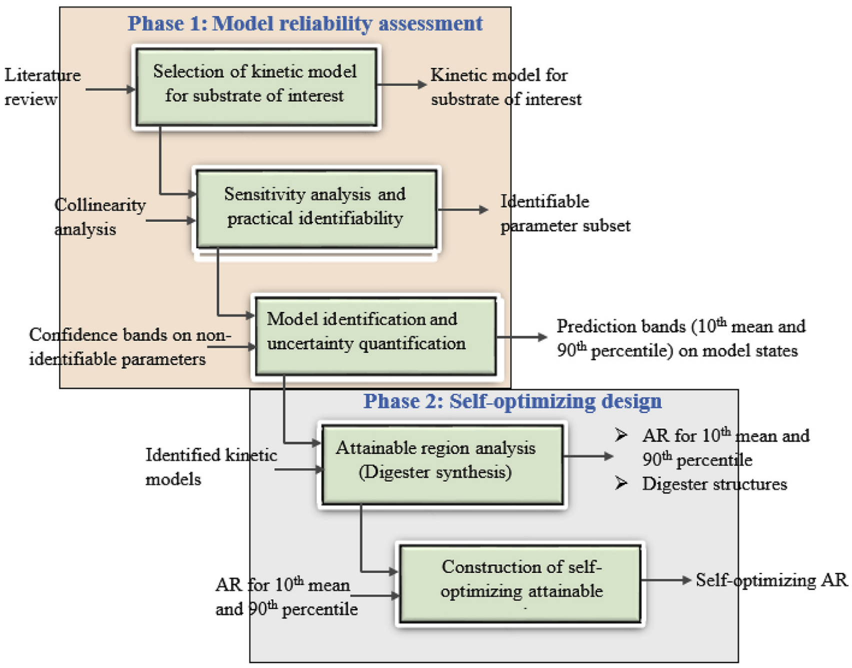

Fig. 1. Two-phase framework for model-based synthesis of methane bioreactors.

operation as an attainable region or performance target that results in near optimal operation despite variations in kinetic coefficients of the process. In the context of process engineering, a similar concept has been applied to plant wide control, known as self-optimizing control, characterized by the choice of self-optimizing controlled variables (Skogestad, 2000; Jäschke et al., 2017).

In order to illustrate the applicability of the framework presented in Fig. 1, the modified Hill model published by (Finn et al., 2013) was selected as a case study. The model considers substrate effects and applies to anaerobic digestion of animal manure (diary, poultry, beef or swine wastes) and predicts acidogens, methanogens, organic substrate and volatile acids. In addition, the model eliminates the need for factors such as alkalinity, concentration of cation, dissolved CO_2 and ammonia gas because their effect is already lumped into two important parameters found in the model, the biodegradability constant (B_0) and acidity factor (A_f).Even though the modified Hill model has been selected, it is important for readers to note that the major contribution of this study is the development of a systematic framework, which couples practical identifiability, uncertainty quantification and attainable region (AR) concepts for defining process performance targets and synthesizing anaerobic digester networks, especially when reliable kinetic coefficients are unavailable. The framework can be used for any other dynamic model selected to describe the kinetics of the anerobic treatment process.

Our recent studies have been first to illustrate the usefulness of AR to define performance targets and model digester configurations that optimize methane productivity and volatile solids reduction (Abunde Neba et al., 2019c), as well as stability of methanogenic archaea (Abunde Neba et al., 2019b). Both studies put together have illustrated that a change in the kinetic model structure or value of kinetic coefficients, induced by differences in substrate and inoculum characteristics significantly influences the performance target as well as the optimal digester configuration required to achieve the target. In another recent study by the authors, a framework was developed and embedded into a software for using simplified microbial kinetic models for AR analysis in cases where data requirements are limited (Abunde Neba et al., 2020a). The integration of economic feasibility indicators (such as payback period, benefit cost ratio, net present value and internal rate of returns) with attainable region analysis has also been presented by the authors, which is very interesting for synthesizing digester structures based on economic objectives (Abunde Neba et al., 2019a). Finally, another approach, which only relies on experimental data (no model required) is developed by coupling attainable regions and fuzzy multicriteria decision making for selection of digester subunits and synthesis of digester network configurations (Abunde Neba et al., 2020b). It is interesting to mention at this point that unlike other model-based studies on AR, which assume that the kinetic coefficients of a model are known before constructing the attainable regions, this study is novel in that it rather quantifies the uncertainty in the kinetic coefficients and propagates it onto the attainable regions. The authors call such regions 'self-optimizing attainable regions', because they will always be attained even if variations occur in kinetic coefficients. Once the AR is obtained, its boundary can always be interpreted into digester structures, which can be used for industrial operation in order to achieve the performance target defined by the region.

2. Theoretical concepts and methods description

2.1. Model reliability assessment

The reliability of a mechanistic model has to do with the degree of uncertainty (the confidence band) of its model parameters and it is influenced by three main factors (Sin et al., 2009, 2010a): (1) the mathematical structure of the model, (2) the nature of the experimental data used for identification, and (3) the set of model parameters used in the identification process. In this paper, the focus is on analyzing the relation amongst model structure (factor 1), identifiable set of parameters (factor 2) and reliability of anaerobic digestion model although the discussion of the results is extended to also reflect on the impact of the information content in the experimental data (factor 2).

Given a kinetic model for a process, we define the following three key steps needed to completely assess the reliability and usage of the model:

Step 1: Perform a sensitivity-based identifiability to determine the identifiable set of model parameters
Step 2: Estimate the identifiable set of model parameters and quantify the confidence band
Step 3: Quantify the model prediction (output) uncertainty using the unidentifiable parameter set as inputs

For studying the identifiability of the biokinetic models, the sensitivity and collinearity analysis are used. For parameter estimation, the method of first order gradients, with gradients computed using the discrete adjoint method, was used and 95% joint and marginal confidence regions were used to assess the identifiability following parameter estimation. For the input-output uncertainty analysis, the Monte Carlo simulation procedure was used.

The objective of this section is to analyze the aforementioned necessary steps with respect to its application to the anaerobic treatment process. However, the analysis requires that the process model is known, and we therefore begin by describing the model of the anaerobic treatment process.

2.1.1. Model selection and description

A number of simplified state-space dynamic models for the anaerobic digestion process have been reviewed by Finn et al. (2013). The modified Hill model which was developed for anaerobic digestion of animal manure (diary, poultry, beef and swine wastes) was selected for this study. The model lumps the effect of hydrolysis, alkalinity, cation concentration, dissolved carbon dioxide and ammonia into two important constants, the biodegradability constant (Bo) and acidity factor (AF) present in the modified Hill model. The Hill model is a mechanistic (model parameters have a physical meaning), which makes it interesting to understand the identifiability characteristics of the model. The identifiability characteristics of a model relates to set of parameters to be estimated in order to accurately describe the observed mechanisms described by the model (Donoso-Bravo et al., 2013) An "over-calibrated" model would accurately describe/fit experimental data but would lose its capability to predict(Donoso-Bravo et al., 2011), which weakens the model's reliability and hence applicability for design purposes.

Fig. 2 presents an illustration of the model by showing the flow of information between four compartments in the methane bioreactor, which include inoculum, substrate, liquid phase and gas phase.

The species conservation and biogas production equations for the modified Hills model is presented as follows

a) Biodegradable volatile solids (S_1) in the liquid phase of the bioreactor

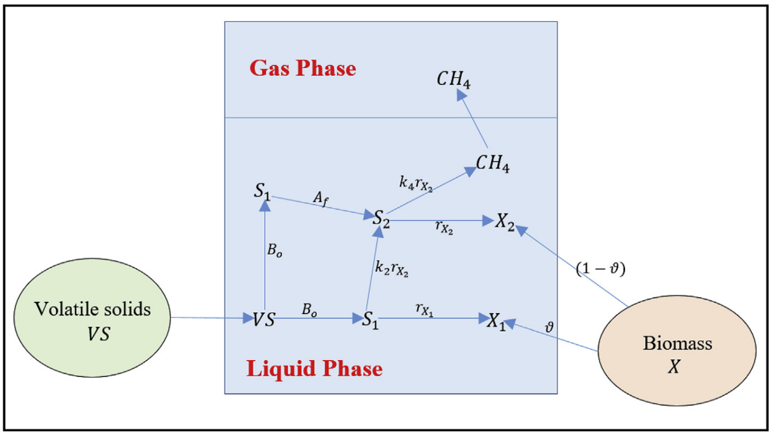

Fig. 2. Information flow of the modified Hill model.

$$\frac{dS_1}{dt} = (S_{1_{in}} - S_1)D - k_1\mu_1 X_1 \tag{1}$$

b) Volatile fatty acids (S_2) in the liquid phase of the bioreactor

$$\frac{dS_2}{dt} = (S_{2_{in}} - S_2)D + k_2\mu_1 X_1 - k_3\mu_2 X_2 \tag{2}$$

c) Acidogens (X_1) in the liquid phase of the bioreactor

$$\frac{dX_1}{dt} = (\mu_1 - K_{d_1} - D)X_1 \tag{3}$$

d) Methanogens (X_2) in the liquid phase of the bioreactor

$$\frac{dX_2}{dt} = (\mu_2 - K_{d_2} - D)X_2 \tag{4}$$

e) Methane gas flow rate

$$Q_{CH_4} = V\mu_2 k_4 X_2 \tag{5}$$

The organic waste is characterized by using the two parameters, which are biodegradability (B_o), Eq. (6) and acidity (A_f), Eq. (7). In the modified Hill model, B_o measures the ease with which the organic substrate can be broken down and stabilized by anaerobic bacteria while A_f of a substrate can be defined as the amount of volatile fatty acids contained in the substrate per unit mass of biodegradable volatile solids

$$S_{1_{in}} = B_0 S_{in} \tag{6}$$

$$S_{2_{in}} = A_f S_{1_{in}} \tag{7}$$

In the modified Hill model the anaerobic biodegradability can be computed via Eq. (8) while the acidity factor is computed using Eq. (9).

$$B_0 = \frac{g\ VS_{destroyed}}{g\ VS_{added}} \quad as\ HRT \to \infty \tag{8}$$

$$A_f = \frac{VFA_{in}}{B_0 \times VSL} \tag{9}$$

The modified Hill's model considers temperature dependence of the anaerobic treatment process through an empirical model, Eq. (10) and since the death rates are set to one tenth of the maximum reaction rates, Eq. (11) they are also show temperature dependent.

$$\mu_{1m}(T) = \mu_{2m}(T) = 0.012T - 0.086 \tag{10}$$

$$K_{d1} = K_{d2} = 0.1\mu_{1m} \tag{11}$$

$10°C < T < 60°C$

In the modified Hill's model, the Monod function, Eq. (12) is used to describe the growth rates of acidogenic and methanogenic microorganisms.

$$\mu_1 = \frac{\mu_{m1} S_1}{K_{s1} + S_1} \tag{12}$$

It is known that anaerobic digestion is sensitive to a wide-range of inhibitory conditions either from toxic substrates or by-products of microbial metabolism (Chen et al., 2014). Since the methanogenic archaea are most sensitive to inhibition than any other group of anaerobic microorganisms (Chen et al., 2008), the Monod function used to describe the growth rate of methanogenic archaea will be replaced by an inhibition counterpart, the Haldane model, Eq. (13). The Haldane model is suited for growth processes affected by the allosteric effectors present in the acidified substrate, non-competitive inhibition (Kythreotou et al., 2014).

$$\mu_2 = \frac{\mu_{m2} S_2}{(K_{s2} + S_2)\left(1 + S_2/K_i\right)} \tag{13}$$

After having defined the kinetic model, we now proceed with assessing the model's reliability for synthesis of anaerobic digesters.

2.1.2. Sensitivity-based identifiability

Model sensitivity analysis provides dynamic information on how the states of a process vary with changes in the model parameters. This information can be used to identify time intervals where experimental data points carry more or less importance for the parameter estimation process. For instance, if the sensitivity of a model state to a given parameter is zero or close to zero in some time interval, then variations in that parameter would have a little influence on that state variable. What this means in practical operation is that having a more accurate experimental measurement of the state variable at that insensitive time interval will not serve to improve the reliability of the parameter estimate. The sensitivity-based identifiability consist of analyzing the sensitivity of the model states to the model parameters, and using these sensitivities to screen for parameter significance ranking by calculating a sensitivity measure, δ_k^{msqr} and for analyzing the near-linear dependency between parameters by a measure called the collinearity index, K.

Given a model for a process, the following five key steps needs to be performed in order to completely assess the reliability and usage of the model (Brun et al., 2002).

Step 1: Compute the absolute sensitivity

Since we do not have an explicit solution to the differential equation model, the absolute sensitivities must be computed using the sensitivity equations. For an n-dimensional system given by Eq. (14)

$$\dot{Y} = f(t, Y; \beta), \quad Y(0) = Y_0 \tag{14}$$

With state variable $Y \in \mathbb{R}^n$, the parameter $\beta \in \mathbb{R}^p$ and Y_0 the initial condition, the matrix of sensitivities $\partial Y / \partial \beta$ satisfy

$$\frac{d}{dt}\frac{\partial Y}{\partial \beta} = \frac{\partial F}{\partial Y}\frac{\partial Y}{\partial \beta} + \frac{\partial F}{\partial \beta} \tag{15}$$

With initial conditions

$$\frac{\partial Y(0)}{\partial \beta} = 0_{n \times p} \tag{16}$$

$\partial Y / \partial \beta$ is the Jacobian of the system. The sensitivity equations are coupled with the original model differential equations and solved to obtain the parameter sensitivities for the necessary time points. The resulting matrix of absolute sensitivities at time point t $S_a(t) = \partial Y / \partial \beta$ will be of the form shown by Eq. (17).

$$S_a(t) = \begin{bmatrix} S_{a,11} & S_{a,12} & \cdots & S_{a,1p} \\ S_{a,21} & S_{a,22} & \cdots & S_{a,2p} \\ \vdots & \vdots & \vdots & \vdots \\ S_{a,n1} & S_{a,12} & \cdots & S_{a,np} \end{bmatrix} \tag{17}$$

Step 2: Compute the non-dimensional sensitivity

The sensitivities of the observables are scaled using the same weights as in Eq. (18), resulting in scaled sensitivities for an output j and a parameter i:

$$S_{nd} = S_a(t) \cdot W \tag{18}$$

The non-dimensional scaling/weighting matrix W is of the form shown by Eq. (19) while the resulting non-dimensional sensitivity of the form given by Eq. (20).

$$W = \begin{bmatrix} \beta_1/S_{c1} & \beta_2/S_{c1} & \cdots & \beta_p/S_{c1} \\ \beta_1/S_{c2} & \beta_2/S_{c2} & \cdots & \beta_p/S_{c2} \\ \vdots & \vdots & \vdots & \vdots \\ \beta_1/S_{c3} & \beta_2/S_{c3} & \cdots & \beta_p/S_{cn} \end{bmatrix} \tag{19}$$

$$S_{nd}(t) = \begin{bmatrix} S_{nd,11} & S_{nd,12} & \cdots & S_{nd,1p} \\ S_{nd,21} & S_{nd,22} & \cdots & S_{nd,2p} \\ \vdots & \vdots & \vdots & \vdots \\ S_{nd,n1} & S_{nd,12} & \cdots & S_{nd,np} \end{bmatrix} \tag{20}$$

Step 3: Compute the sensitivity measure

From the matrix of non-dimensional sensitivities, we compute an overall coring for each parameter, called root mean squared sensitivity, δ_k^{msqr}, to consider changes in time or across experiments. The root mean squared sensitivity is computed using Eq. (21)

$$\delta_k^{msqr} = \sqrt{\frac{1}{N}\sum_{l=1}^{N} \left(S_{nd,lk}\right)^2} \tag{21}$$

N is the number of state variables and $k = 1, 2 \ldots p$where p is the number of model parameters. A vector of root mean squared sensitivities of the different model parameters is created of the form given by Eq. (22)

$$\delta^{msqr} = \begin{bmatrix} \delta_1^{msqr} & \delta_2^{msqr} & \ldots & \delta_p^{msqr} \end{bmatrix} \tag{22}$$

The sensitivity measure (δ^{msqr}) measures the relative importance of the parameters with respect to how the influence the model outputs (states). The higher the magnitude of the sensitivity measure the more important the influence of the parameter on the states.

Step 4: Compute the normalized sensitivity

From the matrix of non-dimensional sensitivity, we compute the normalized sensitivity for each parameter using Eq. (23), re-written as Eq. (24).

$$S_{norm} = \frac{S_{nd,lk}}{S_{nd}(t)} \tag{23}$$

$$S_{norm} = \frac{S_{nd,(l,k)}}{\sqrt{\sum_{k=1}^{n} S_{nd,(l,k)}^2}} \quad l = 1, 2, \ \ldots, n; k = 1, 2, \ \ldots, p \tag{24}$$

Step 5: Compute the collinearity index

Finally, step five consist of computing the collinearity index γ_K using Eq. (25)

$$\gamma_K = \frac{1}{\sqrt{\min \lambda_K}} \tag{25}$$

$$\lambda_K = eigen\left(S_{norm,\ K}^T S_{norm,\ K}\right)$$

K stands for the index of the parameter subset, which is a combinatorial function of the parameter vector β.

If the sensitivity functions of two or more parameters are orthogonal (implying parameters are independent), the index of that parameter subset (K) is equal to unity, but if the parameters are linearly dependent, the index approaches infinity. In order to find an identifiable parameter subset, a threshold value (1-15) is usually used (Brun et al., 2002; Sin and Vanrolleghem, 2007) where by any parameter subset having an index (K) greater than the threshold is said to be unidentifiable.

2.1.3. Parameter estimation: confidence bounds and correlation analysis

In this section, we describe the adjoint-based gradient method for parameter estimation. The method is selected rather than the standard finite difference method because it takes less computing time and is less sensitive to round-off and truncation errors, which becomes very attractive for optimization problems with large number of variables (Benítez et al., 2017). To facilitate mathematical developments in subsequent sections, we redefine the model states and parameters as follows;

$$Y_1 = S_1, \quad Y_2 = S_2, \quad Y_3 = X_1 \quad Y_4 = X_2\,, \quad Y_5 = Q_{CH_4}$$

$$\beta_1 = k_1, \quad \beta_2 = k_2, \quad \beta_3 = k_3, \quad \beta_4 = k_4, \quad \beta_5 = K_{i1}, \quad \beta_6 = K_{i2}$$

$$\textit{Minimize} \quad J(\beta) = \frac{1}{2} Y_i^{obs} - HM\left(t, Y^i; \beta\right)^2 \tag{26}$$

subject to

$$\frac{dY_1}{dt} = \left(Y_{1_{in}} - Y_1\right)D - \beta_1 \mu_1 Y_3 \tag{26a}$$

$$\frac{dY_2}{dt} = \left(Y_{2_{in}} - Y_2\right)D + \beta_2 \mu_1 Y_3 - \beta_3 \mu_2 Y_4 \tag{26b}$$

$$\frac{dY_3}{dt} = \left(\mu_1 - K_{d_1} - D\right)Y_3 \tag{26c}$$

$$\frac{dY_4}{dt} = \left(\mu_2 - K_{d_2} - D\right)Y_4 \tag{26d}$$

$$Y_5 = V\mu_2 \beta_4 Y_4 \tag{26e}$$

$$\beta_1 < \beta_3; \quad \beta_5 < \beta_6 \quad \beta_1, \beta_2, \beta_3, \beta_4 > 0 \quad \beta_5, \beta_6 \geq 0$$

$$\mu_i = \mu_i(\beta_{i+4}), \quad i = 1, 2$$

Eq. (26) presents a constraint nonlinear optimization problem, where the constraints are differential algebraic equations. In order to find the numerical solution of the problem there exist indirect and direct methods of minimization of the objective function. In the direct method, the state equations are influenced only by the parameters, and the minimization of the function is done by direct adjustment of the model parameters. The simplest approach to a direct method is that of first order gradients in which the state and co-state equations remain separated. The system of continuous equations is regarded as a limiting case of a system of discrete equations as the time of a subinterval approaches zero. The optimization problem is solved using the method of conjugate gradients with the gradient computed by the adjoint method. The conjugate gradient algorithm is illustrated as follows

Given $J : \mathbb{R}^n \to \mathbb{R}$ and $\nabla J(\beta)$. Let $\beta^{(0)}$ be the initial guess and set $w^{(0)} = -\nabla_\beta J(\beta^{(0)})$

For $k = 0, 1, 2, 3, \ldots$

Step 1: Perform a line search in the direction of to compute $\gamma = \textit{Arg min}\Phi(\rho)$, which minimizes the scalar function $\Phi(\rho) = f(\beta^{(k)}, \rho w^{(k)})$
Step 2: Compute $\beta^{(k+1)} = \beta^{(k)} + \gamma^{(k)} w^{(k)}$
Step 3: Test for convergence. If satisfied Exit, else go to Step 4.
Step 4: Define $\tau^{(k)} = \frac{\nabla J(\beta^{(k)})^2}{\nabla J(\beta^{(k-1)})^2}$
Step 5: Compute $w^{(k+1)} = -\nabla_\beta J(\beta^{(k+1)}) + \tau^{(k)} w^{(k)}$ and go to step (1).

We notice that from the computational point of view a discrete adjoint approach is the one needed to accurately compute the gradient. The model equations are discretized using the Runge-Kutta 4th order scheme as shown by Eq. (27)

$$Y^{(k+1)} = M\left(Y^{(k)}, \beta\right) \tag{27}$$

$$f(t, Y(t);\ \beta) = \begin{bmatrix} \left(Y_{1_{in}} - Y_1\right)D - \beta_1 \mu_1 Y_3 \\ \left(Y_{2_{in}} - Y_2\right)D + \beta_2 \mu_1 Y_3 - \beta_3 \mu_2 Y_4 \\ \left(\mu_1 - K_{d_1} - D\right)Y_3 \\ \left(\mu_2 - K_{d_2} - D\right)Y_4 \\ V\mu_2 \beta_4 Y_4 \end{bmatrix} \tag{27a}$$

$$K_1 = f\left(t^{(k)}, Y^{(k)}; \beta\right) \tag{27b}$$

$$K_2 = f\left(t^{(k)} + 0.5h, Y^{(k)} + 0.5K_1; \beta\right) \tag{27c}$$

$$K_3 = f\left(t^{(k)} + 0.5h, Y^{(k)} + 0.5K_2; \beta\right) \tag{27d}$$

$$K_4 = f\left(t^{(k)} + h, Y^{(k)} + K_3; \beta\right) \tag{27e}$$

$$M\left(Y^{(k)}, \beta\right) = Y^{(k)} + \frac{1}{6}(K_1 + 2K_2 + 2K_3 + K_4) \tag{27f}$$

The optimization problem can then be simply written in a discrete and compressed form as in Eq. (28)

$$\textit{Minimize} \ \ J(\beta) = \frac{1}{2}\sum_{k=0}^{N}\left(Y_k^{obs} - HY^{(k)}\right)^2 \tag{28}$$

subject to

$$Y^{(k+1)} - M\left(Y^{(k)}, \beta\right) = 0 \tag{28a}$$

The adjoint method consist of transforming a constraint optimization problem into an unconstrained problem by defining the Lagrangian, Eq. (29)

$$L(Y,\ \beta,\ \lambda) = J(\beta) + \sum_{k=0}^{N} \lambda_k \left[Y^{(k+1)} - M\left(Y^{(k)}, \beta\right)\right] \tag{29}$$

From the Lagrangian, we can then derive the state equations, Eq. (30) and gradient of the optimization problem, Eq. (31). The Adjoint model is given by Eq. (32) (Roulston, 1999).

$$\frac{\partial L(Y, \beta, \lambda)}{\partial Y^{(k)}} = \frac{\partial J_k(\beta)}{\partial Y^{(k)}} + \lambda_{k-1} - \lambda_k \left[\frac{\partial M\left(Y^{(k)}, \beta\right)}{\partial Y^{(k)}}\right]^T \tag{30}$$

$$\nabla_\beta J(\beta) = \frac{\partial L(Y,\beta,\lambda)}{\partial \beta} = -\sum_{k=0}^{N} \lambda_k M_\beta^T \left(Y^{(k)},\beta\right) \tag{31}$$

$$\lambda_{k-1} - M_Y^T\left(Y^{(k)},\beta\right)\lambda_k = e_k \quad \lambda_N = 0 \tag{32}$$

Step 1: Choose an initial guess $\beta^{(0)}$ and set counter $k = 0$
Step 2: Solve the forward model $Y^{(k+1)} = M\,(Y^{(k)},\beta)$ and compute the criterion $J(\beta)$
Step 3: Solve the Adjoint model $\lambda_{k-1} - M_Y^T(Y^{(k)},\beta)\lambda_k = e_k$ and compute the gradient

$$\nabla_\beta J(\beta) = -\sum_{k=0}^{N} \lambda_k M_\beta^T\left(Y^{(k)},\beta\right)$$

Step 4: Determine the descent direction

if $k = 0$

$$w^{(k)} = -\nabla_\beta J\left(\beta^{(k)}\right)$$

else

$$w^{(k)} = -\nabla_\beta J\left(\beta^{(k)}\right) + \frac{\nabla J\left(\beta^{(k)}\right)^2}{\nabla J\left(\beta^{(k-1)}\right)^2} w^{(k-1)}$$

Step 5: Perform a line search in the direction of to compute $\gamma = Arg\ min\Phi(\rho)$, which minimizes the scalar function $\Phi(\rho) = f(\beta^{(k)}, \rho w^{(k)})$
Step 6: Compute a new state vector estimate $\beta^{(k+1)} = \beta^{(k)} + \gamma^{(k)} w^{(k)}$
Step 7: Set $k =: k+1$ and return to step 2 until a termination condition is reached

All the work on the computer was carried out using Matlab R2017b (Mathworks Natick) using i7-6600U, 2.6 GHz CPU PC with 16 GB RAM and 64bits operating system.

2.1.4. Uncertainty quantification in model predictions

As mentioned in section 1, self-optimizing operation of anaerobic digesters is when we have an acceptable loss in performance as a result of kinetic uncertainty in the model. In order to therefore use the model to model the self-optimizing performance target, one needs to quantify the model prediction uncertainty resulting from uncertainty in kinetic coefficients. In order to quantify the model prediction uncertainty, the Monte Carlo simulation procedure, presented in Fig. 3 was applied in a similar way as in Sin et al. (2010b).

Input-output uncertainty analysis is highly dependent on the input uncertainty range (confidence bounds) as well as correlation coefficients. The variance metrics and correlation coefficients of the unidentifiable set of model parameters for the different biokinetic models were obtained by estimating the complete set of parameters (identifiable and unidentifiable) using the estimation procedure presented in section 2.1.3.

2.2. Self-optimizing performance targeting

Given a set of reactions and associated kinetics, the following five key steps needs to be performed in order to define the performance target of a process using attainable region analysis (Ming et al., 2016):

- ➢ Define the reaction, dimension and feed set
- ➢ Define the fundamental processes occurring in the system
- ➢ Generate the AR using combinations of the fundamental processes
- ➢ Interpret the AR boundary in terms of reactor equipment
- ➢ Define the objective function and overlay this onto the AR to determine point of intersection with the AR boundary
- ➢ Determine the specific reactor configuration required to achieve the intersection point

The previous two bullet points are important if the attainable region is to be used to answer a specific design or optimization question.

Some necessary conditions for AR can be summarized as follows (Hildebrandt and Glasser, 1990; Hildebrandt et al., 1990):

- ➢ The AR includes all feeds to the system.
- ➢ The AR is convex.
- ➢ No process vector point out of the AR boundary.
- ➢ No rate vectors in the complement of the AR when extended backward intersects the AR.

The objective of this section is to analyze the aforementioned necessary requirements with respect to its application to the anaerobic treatment process.

2.2.1. Reaction scheme and process kinetics

Using the information flow diagram of the kinetic model presented in Fig. 2, a stoichiometric scheme of the bioreaction occurring in the anaerobic digester consist of two main reactions catalyzed by acid-forming bacteria, Eq. (33) and methane-forming bacteria Eq. (34)

$$k_1 S_1 \xrightarrow{r_{X_1}} X_1 + k_2 S_2 \tag{33}$$

$$k_3 S_2 \xrightarrow{r_{X_2}} X_2 + k_4 CH_4 \tag{34}$$

If we assume the specific death rate to be negligible compared to the specific growth rate of both microbial populations, the rate expressions for the different reaction species is defined by Eq. (35) − (38)

$$r_{X_1} = \mu_1 X_1 \tag{35}$$

$$r_{X_2} = \mu_2 X_2 \tag{36}$$

$$r_{S_1} = -k_1 \mu_1 X_1 \tag{37}$$

$$r_{S_2} = k_2 \mu_1 X_1 - k_3 \mu_2 X_2 \tag{38}$$

2.2.2. Fundamental processes

Various fundamental processes can occur within a system, which for bioreactors may include: mass transfer, mixing, bioreaction (biodegradation, bioconversion), adsorption, heat transfer, etc. The AR approach requires the fundamental processes taking place in the system be identified. The following two main fundamental processes are identified to be associated with the anaerobic treatment process: Biodegradation and mixing. The attainable

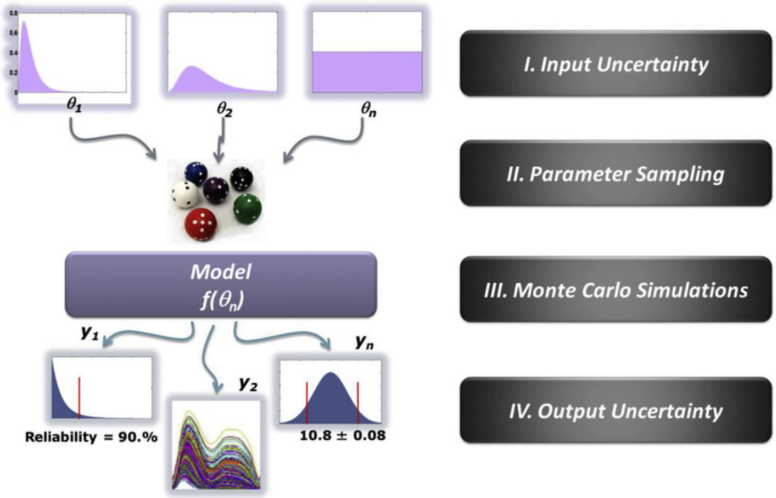

Fig. 3. Monte-Carlo simulation method for uncertainty propagation (Morales-Rodriguez et al., 2012).

region (AR) for the anaerobic treatment process therefore represents the set of all possible states that can be achieved by a combination the two fundamental processes, biodegradation and mixing. In AR theory, mixing is performed by a continuous stirred tank reactor (CSTR) while reaction (biodegradation) is achieved in a plug flow reactor (PFR), since the operation of both reactors respectively mimic the two fundamental processes. At steady state operation, the general mathematical representation of a CSTR and PFR are given by Eqs. (39) and (40) respectively.

$$C = C_f + \tau r(C) \tag{39}$$

$$\frac{dC}{d\tau} = r(C) \tag{40}$$

C is the state vector while $r(C)$ is the reaction rate vector as shown by Eqs. (41) and (42) respectively.

$$\mathbf{C} = [X_1 \quad X_2 \quad S_1 \quad S_2]^T \tag{41}$$

$$\mathbf{r}(\mathbf{C}) = [r_{X_1} \quad r_{X_2} \quad r_{S_1} \quad r_{S_2}]^T \tag{42}$$

2.2.3. Dimensionality analysis and model reduction

The reaction stoichiometry of the system can be used to determine the dimension of the system. The dimension of the AR is determined from the number of independent reactions occurring in the reactor system, which defines the dimension of the stoichiometric subspace (the rank of the stoichiometric coefficient matrix A), in which the AR must reside. Since there are two independent reactions occurring in the system, the set of points generated by the anaerobic treatment process must reside in a two-dimensional subspace in $\mathbb{R}^5$ (Ming et al., 2016). The reduced state and reaction rate vectors are therefore presented by Eqs. (43) and (44).

$$\mathbf{C} = [S_2 \quad X_2]^T \tag{43}$$

$$\mathbf{r}(\mathbf{C}) = [r_{S_2} \quad r_{X_2}]^T \tag{44}$$

The reduction involved expressing S_1 and X_1, as a function of S_2 and X_2, as shown by Eqs. (45) and (46).

$$S_1 = S_{1_{in}} - k_1(X_1 - X_{1_{in}}) \tag{45}$$

$$X_1 = X_{1_{in}} + \frac{1}{k_2}[S_2 - S_{2_{in}} + k_3(X_2 - X_{2_{in}})] \tag{46}$$

This reduction in the dimensions of the state and rate vectors was done using the approach illustrate in our recent study using attainable regions for synthesis and optimization of methane bioreactors (Ref). The model reduction assumes that the specific death rates of acidogens and methanogens is negligible compared to their respective specific growth rates.

2.2.4. AR construction and defining performance target for the system

After stating the process kinetics, the AR construction process is initiated by defining feed point and process conditions that influence the system. In anaerobic treatment, the digester is normally maintained at constant temperature (isothermal process) throughout retention time, which makes the AR dependent on the particular temperature in the system. The anaerobic digestion was carried out under mesophilic conditions at a temperature of 35°C. Using the specified feed, kinetics and temperature conditions, the set of points generated by solving the PFR equation are called the PFR trajectory and those generated by solving the CSTR equation are called the CSTR locus. The convex hull of the set of points generated by the system defines the attainable region, which represents the limits of achievability by the system.

3. Results and discussion

In this section, data from a real experiment is utilized to illustrate the theories presented in the previous sections. The case study is based on a batch methane bioreactor operated with diary manure, where experimental measurements of volatile fatty acids and methane gas flowrate (which, can be used to get the concentration of methanogenic archae) were obtained (Zaher et al., 2009).

3.1. Parameter identifiability measures

3.1.1. Sensitivity analysis

In this section, the objective was to determine, which set of parameters should be estimated to accurately describe the mechanisms of the anaerobic digestion process. This depends on analyzing the sensitivity function of the model parameters with respect to with respect to the states. Fig. 4 presents the sensitivity functions (dynamic sensitivities) of the states for the parameters of the biokinetic model. From the shape of the sensitivity functions, the authors made the following remarks: (1) All the states show some sensitivity to the model parameters, which can either be a negative or positive sensitivity. (2) The anaerobic microorganisms mostly show negative sensitivity while the substrates show both negative and positive sensitivities to the model parameters. Table 1 presents the numerical characteristics sensitivity analysis, which include: the nominal values and scale of the model parameters; the mean, minimum and maximum values of the dynamic sensitivities; the sensitivity measures (L1 and L2) as well as the number of data points (N). The 130 data points corresponds to the small time step of 0.0769 that was used to integrate the sensitivity equations from 0 to 10days.

Particularly, it is worth mentioning that for the substrates, the biodegradable volatile solids is most sensitive to the acid yield coefficient (k_2) while volatile fatty acids are most sensitivity to the Monod saturation constant for volatile acids (K_{s2}) and inhibition constant (K_i). For the anaerobic microorganisms, the acidogenic bacteria is highly sensitive to the Monod saturation constant (K_{s2}), while the methanogenic archae are highly sensitive to inhibition constant (K_i). These outcome accurately describe the underlying theories of the anaerobic treatment process, which include: breakdown of volatile solids into volatile fatty acids by acidogenic bacteria, utilization of volatile fatty acids for growth of methanogenic archaea as well as high sensitivity of methanogenic archaea to inhibitions (Henze et al., 2008; Wang et al., 2007). Hence the results clearly illustrate the ability of the model to describe the anaerobic digestion process.

Fig. 5 presents the use of the sensitivity measure (sum of sensitivity functions of the available measurements with respect to

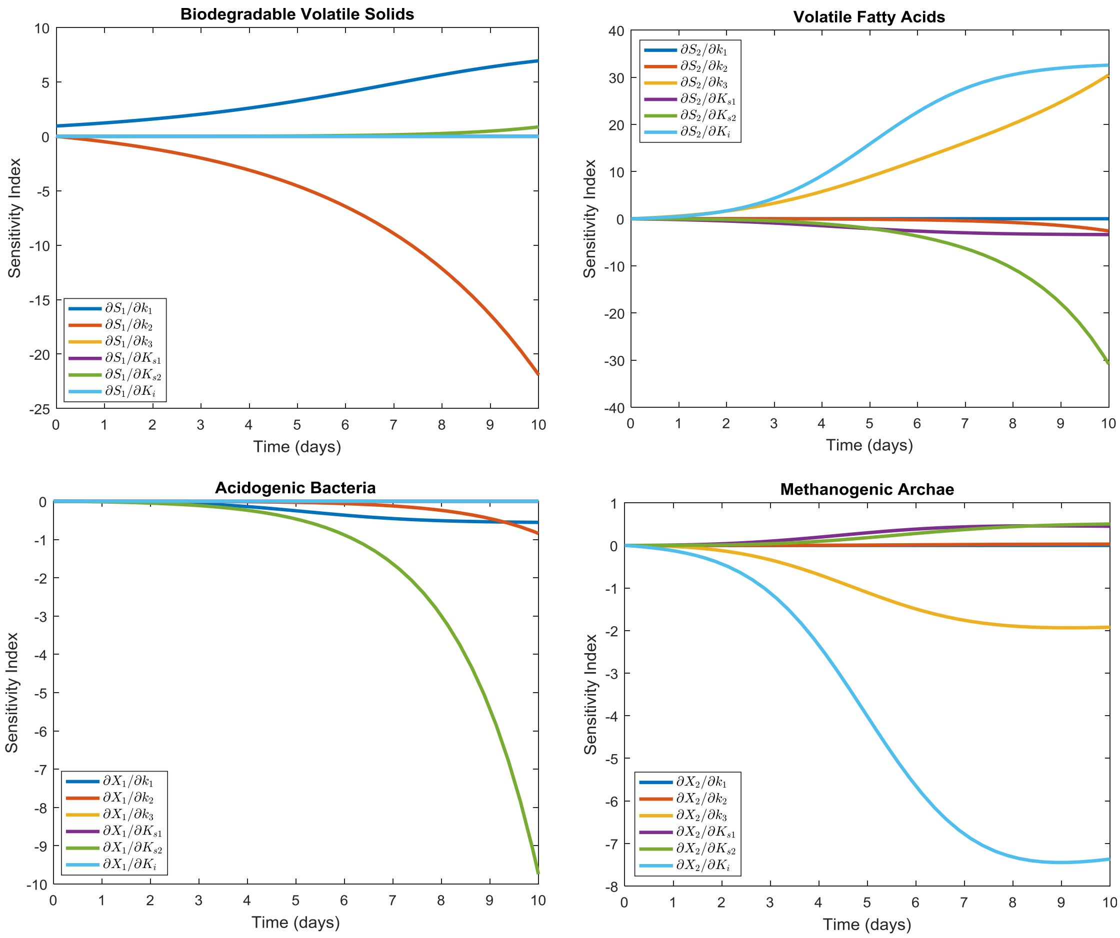

Fig. 4. Sensitivity of model states to model parameters.

Table 1
Sensitivity measures of the observable states to the model parameters.

Parameters	Value	Scale	L1	L2	Mean	Min	Max	N
$\boldsymbol{k_1}$	0.1920	0.1920	0.0000	0.0000	0.0000	0.0000	0.0000	130
$\boldsymbol{k_2}$	0.5029	0.5029	0.2131	0.6086	−0.2042	−2.5915	0.0302	130
$\boldsymbol{k_3}$	0.1920	0.1920	4.6606	9.4970	3.9058	−1.9351	30.5131	130
$\boldsymbol{K_{s1}}$	25.0687	25.0687	0.7843	1.3894	−0.5965	−3.3613	0.4591	130
$\boldsymbol{K_{s2}}$	0.0899	0.0899	2.7457	7.4232	−2.5820	−30.8306	0.5022	130
$\boldsymbol{K_i}$	1088.8324	1088.8324	7.3896	12.9875	4.5331	−7.4461	32.5866	130

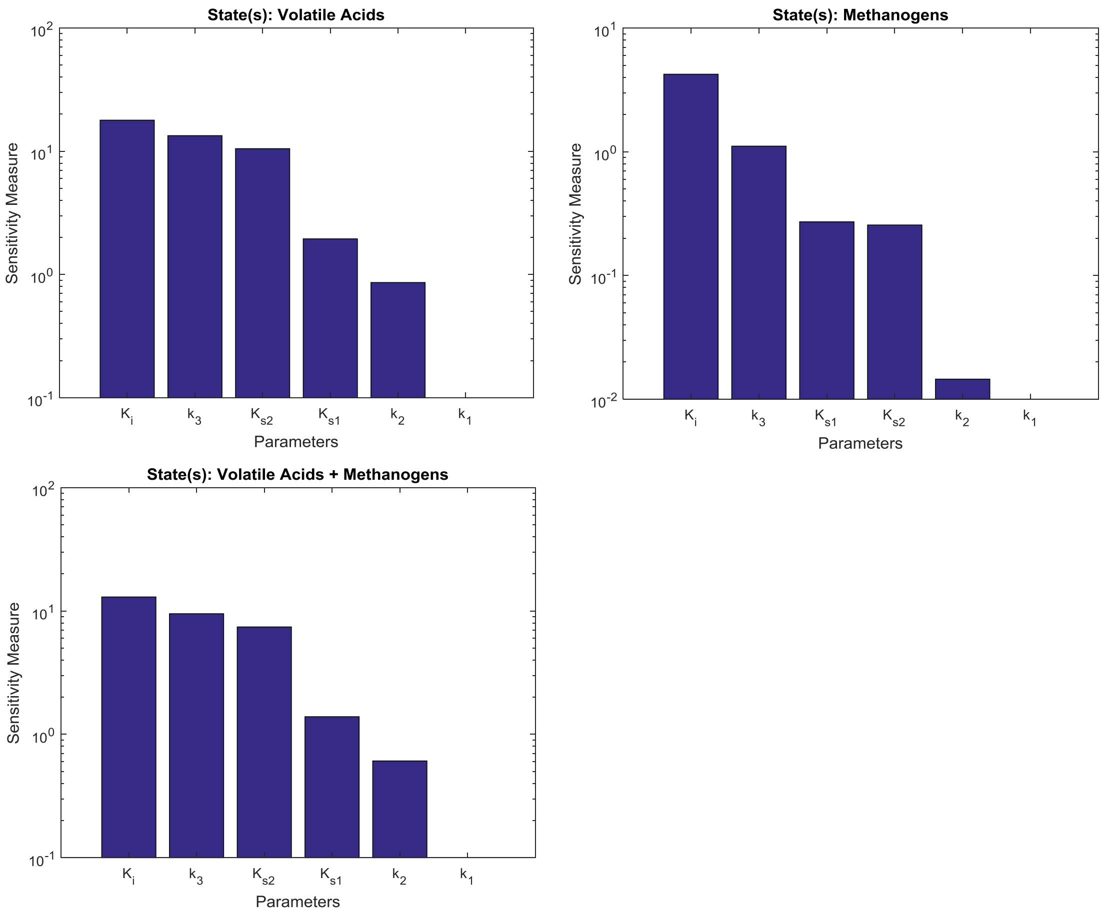

Fig. 5. Parameter significance ranking for observable states based on sensitivity measure.

parameters) to rank the model parameters, which reveals the relative significance of the parameters with respect to the observable states. Only the relative ranking for volatile acids, methanogenic archaea as well as volatile acids + methanogenic archae as model as model outputs are considered. This is because the experimental data, which is used as our case study only contains measurements for volatile acids methanogenic archae (calculated form methane flowrate) and we therefore determine the set of model parameters that are identifiable considering these two observable states.

It is noteworthy that the yield coefficient (k_1) is found completely not sensitive to any of the observable states. The inhibition constant (K_i) was found most significant for all the observable states followed by the yield coefficient (k_3). The Monod constants (K_{s1} and K_{s2}) and the yield coefficient (k_2) were found significant albeit to a relatively lower degree. The practical relevance of the parameter significance ranking is that only those parameters with significant sensitivity measure with respect to the observable states can be identified. Therefore, considering volatile fatty acids and methanogenic archaea as the only observable states in the system, the yield coefficient k_1 cannot be identified from the available data since it has a sensitivity measure of zero. This implies only 5 out of the 6 model parameters can be candidates for parameter estimation.

3.1.2. Collinearity analysis

This section of the identifiability analysis only considers those set of parameters (5 out of the 6 model parameters were significant), which have a significant effect on the observable states. The collinearity analysis screens all possible subsets of the potential candidate parameters to determine the identifiable subsets using a collinearity index. The five potential candidates for parameter estimation gives a total of approximately 31 parameter subset combinations, with a maximum subset size of five parameters

Fig. 6a presents the collinearity analysis for all possible combinations of parameter subsets while Fig. 6b presents collinearity analysis for the potentially identifiable subsets. From Fig. 6, it can be observed that of the 31 possible subset combinations, only 18 are potentially identifiable (those having collinearity index less than 15) and with a maximum identifiable subset size of three parameters

These findings suggest that for a given set of observable states (experimental measurements), there exist many identifiable subset combinations of model parameters having a maximum number of parameters that can be estimated uniquely. Unique estimation means that by using an identifiable subset, the estimated parameters should have a relatively lower correlation values and/or confidence intervals. The results corroborate the theoretical premise that subjecting an overparameterized model to limited quality/quantity of data limits the number of parameter that can be estimated to uniquely and accurately describe the system (Brun et al., 2002; Sin and Vanrolleghem, 2007).

3.2. Model fits and parameter uncertainty

Two cases of parameter estimation were considered: one with an identifiable subset (specifically k_2, K_{s2} and K_i) and one with all the model parameters (known as the nominal case) so that the effect of identifiability analysis can be ascertained. The model fits for both cases are presented in Fig. 7 while parameter estimates together with their 95% marginal confidence intervals are shown in Table 2. Visually, both cases show a good fit between the experimental measurements and model predictions with no observable difference in both cases. However, from a numerical perspective (see Table 2), the parameter estimates from the nominal case shows a much higher degree of uncertainty (given as the standard deviation, which relates to the 95% marginal confidence interval) than that of the identifiable case. Put it in another way, the identifiability analysis has served to reduce the degree of uncertainty in model parameter estimates.

The results indicate that despite variation in parameter uncertainty, the quality of the model fit to experimental data is not compromised and using an identifiable subset of model parameters serves to improve the quality of the model parameters. It is worth mentioning that the identifiable parameter subset utilized for parameter estimation is just one of the three identifiable subsets available with size of 3. Other three-parameter combinations of identifiable subsets can still be selected as candidates for the parameter estimation. The focus of this study is not to consider all the identifiable subset, but to illustrate how these identifiability issues should be incorporated in digester synthesis.

3.3. Uncertainty quantification on model states

Recall from step 3 of section 2, which stated the need to quantify the model prediction (state) uncertainty using the unidentifiable parameter set as inputs. From section 3.2, we have demonstrated the use of an identifiable parameter subset (k_2,K_{s2} and K_i) to reduce uncertainty in model parameters. Even though the use of an identifiable subset reduces parameter uncertainty, it causes another problem, which is that of model uncertainty. This is because those parameters that are not identifiable (k_1, k_2 and K_{s1}) need to be kept constant (probably using values estimated from previous studies or independent experiments), which influences the reliability of the model. Since the geometric optimization technique of attainable regions presented in this study for synthesis of anaerobic digesters is unique for a given kinetic model, an unreliable model will therefore result in an unreliable digester system, which can easily lead to operational failure. Hence before using the model to construct the attainable regions (which can be interpreted into digester structures), we quantify and incorporate the model prediction uncertainty into the limits of achievability of the system, which is defined by the attainable regions.

Fig. 8 presents the results obtained from the Monte Carlo simulations. From a general perspective, the results indicate that each of the model states have a time varying uncertainty band defined by the 10th and the 90th percentile. The methanogenic archaea shows insignificant uncertainty band to the model inputs at certain times instants, where the mean, 10th and 90th percentile are equal. The width of the band (difference between the 10th and the 90th

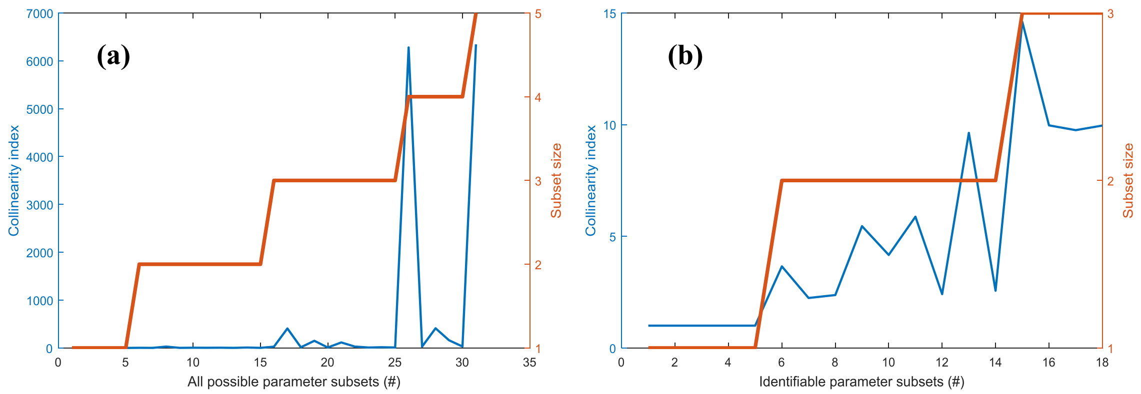

Fig. 6. Collinearity analysis for: (a) all possible combination of parameter subsets (b) potentially identifiable parameter subsets.

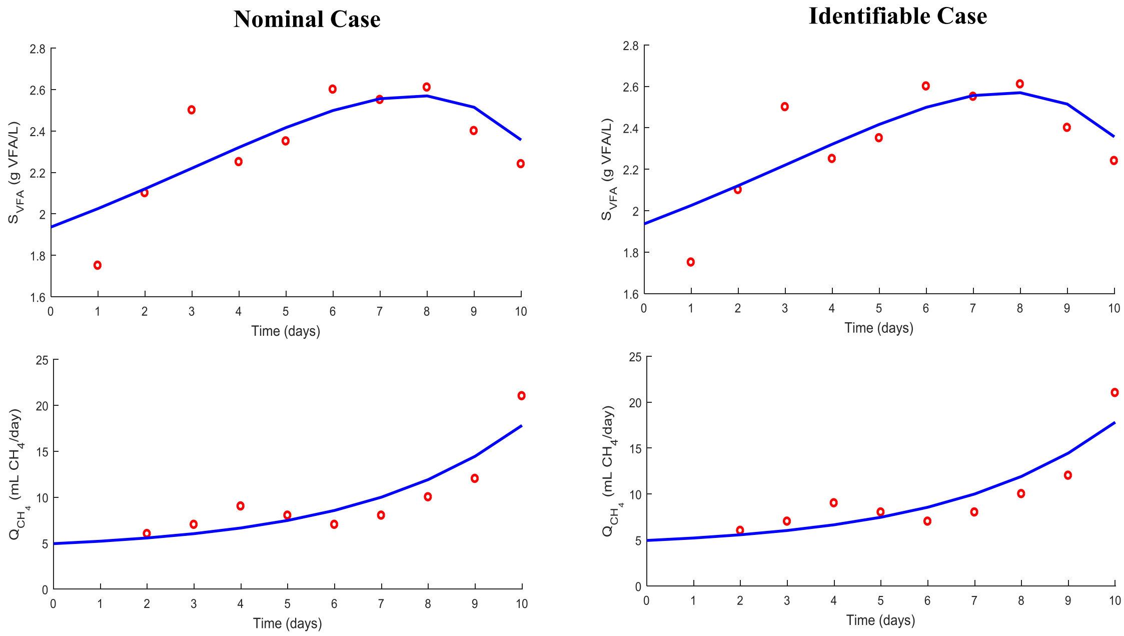

Fig. 7. Model fits to experimental data for both nominal and identifiable cases.

Table 2
Parameter estimates and uncertainty given as the standard deviation.

Model Parameters	Parameter Estimates		Standard Deviation	
	Nominal case	Identifiable case	Nominal case	Identifiable case
$\boldsymbol{k_1}$	0.1920	0.1920	45.8498	N/A
$\boldsymbol{k_2}$	0.5029	0.514889	3.2951	0.62322
$\boldsymbol{k_3}$	0.1920	0.1920	3.6133	N/A
$\boldsymbol{K_{s1}}$	25.0687	25.0687	386.8622	N/A
$\boldsymbol{K_{s2}}$	0.0899	0.852436	0.2822	0.78416
$\boldsymbol{K_i}$	1088.8324	1088.5435	22887.6440	12.1010

percentile) describes the spread of the distribution of the model states resulting from parameter uncertainty and the larger the width, the higher the degree of model output uncertainty. This is often called mapping/propagating parameter (input) uncertainty onto states (output) uncertainty.

The state uncertainty bands presented in Fig. 8 are highly dependent on the uncertainty range of the model parameters. The study used the joint confidence region (Fig. 9) of the sampled parameters, which takes into consideration the correlation amongst model parameters and eliminates the need to define the correlation amongst model parameters during the Monte Carlo procedure.

The interpretation of these results is based on the relationship between uncertainty band and model quality: the higher the uncertainty band, the lower the model quality. Hence model predictions for biodegradable volatile solids followed by volatile fatty acids and acidogenic bacteria are deemed of low quality (large uncertainty bands) while that of methanogenic archaea can be deemed acceptable.

3.4. Self-optimizing attainable regions

Surely, whether of acceptable quality or not, the prediction uncertainty around the model states affects the limits of achievability of the anaerobic digestion process and hence the nature of the optimal digester structures. This is because for synthesis of methane bioreactors using attainable region analysis, the predicted performance target or limits of achievability by the system is computed by the area of the convex hull for the set of states (outputs) achievable by the system.

Hence, when using attainable regions for performance targeting and digester network synthesis, we suggest that it should be mandatory to incorporate uncertainty of model prediction during construction of the attainable regions. The approach here relies on constructing the attainable regions using the three key points of the state's prediction (mean, 10th and 90th percentile) and superposing the regions to obtain a robust region which considers the effect of uncertainty. Fig. 10 presents the AR for the 10th percentile, mean and 90th percentile state predictions on to which the digester structures required to attain points on the AR boundary has been overlaid. A detailed explanation of how the AR boundary has been interpreted into digester structures is presented in our recent publication (Abunde Neba et al., 2019c). It can be observed that for all the cases where the AR boundary is convex, the optimal digester structure involves a plug flow digester in order to attain points on

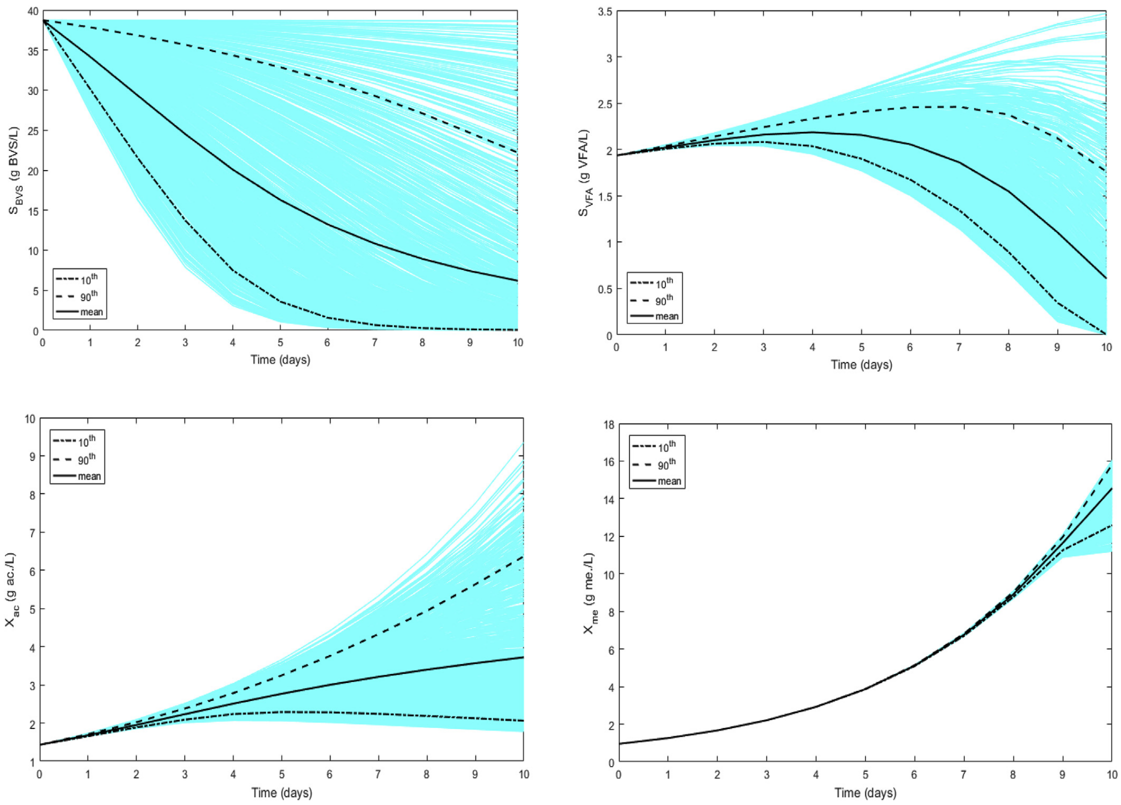

Fig. 8. Model uncertainty quantification (the mean, the 10th and 90th) using 1000 Monte Carlo simulations.

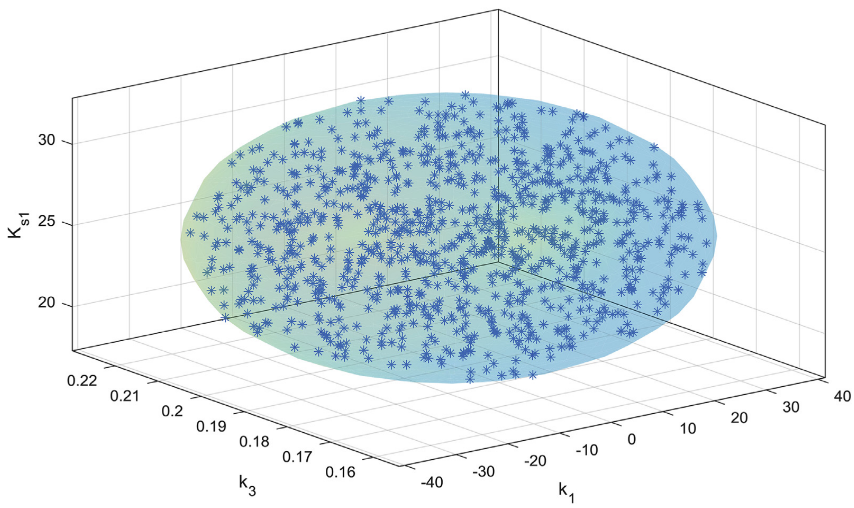

Fig. 9. Joint confidence region of unidentifiable parameters showing parameters sample for 1000 Monte Carlo simulations.

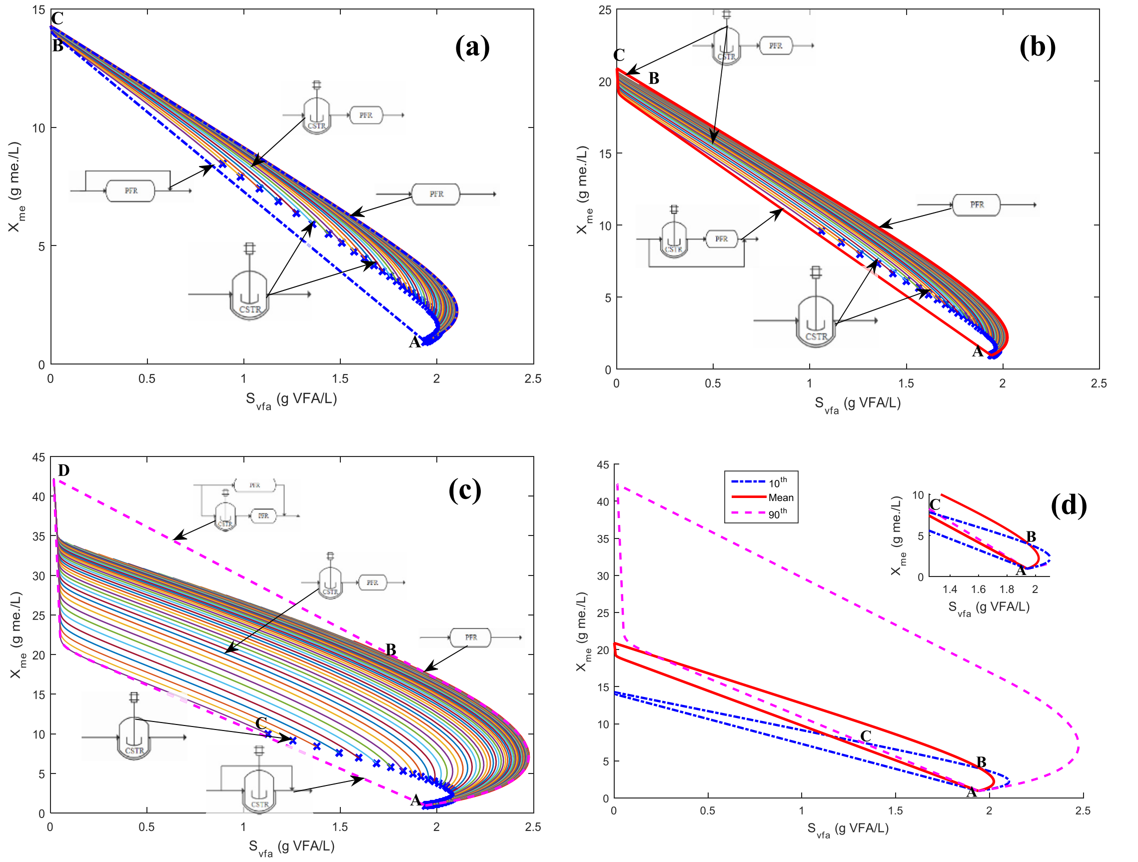

Fig. 10. Illustration of the self-optimizing attainable region: (a) Attainable region for 10th percentile uncertainty bound. (b) Attainable region for mean parameter values. (c) Attainable region for 90th percentile uncertainty bound. (d) Self-optimizing attainable regions of the anaerobic treatment process.

the AR boundary. This can be explained by the intrinsic geometric characteristics of this type of reactor in relation to the properties of the AR boundary. The AR boundary is composed entirely of reaction and mixing surfaces only. Reaction surfaces are always convex and the points that form convex sections of the AR boundary arise specifically from points on PFR trajectories (Ming et al., 2016). This is so because governing equations of a PFR is a system of first order ordinary differential equations Eq. (17), where a phase plane presentation of the solution of the system for a given organic load and digestion time is called PFR trajectory. Geometrically, the rate vector evaluated at points on the PFR trajectory is tangent to all points on the trajectory (Hildebrandt and Glasser, 1990; Ming et al., 2016). This implies the boundary of a true AR will always contain points originating from PFR trajectory, otherwise it becomes a candidate AR. Fig. 10d present the intersection of the three regions to define the self-optimizing attainable region. It can be observed that the region looks smaller than any of the three individual regions (10th percentile, mean and 90th percentile). This illustrates the accept loss in process operation mentioned in section 1. It is necessary here to re-clarify exactly what is meant by self-optimizing attainable regions. Unlike the attainable region, which represents the set of all possible states that is attainable by the system for a defined kinetics and initial condition (feed point), the self-optimizing attainable region represents the set of all possible states attainable by the system even in cases of kinetic uncertainty. The size of the self-optimizing attainable region is related to the domain of uncertainty defined for the unidentifiable set of model parameters used for the uncertainty propagation. The presence of uncertainty reduces the size of the self-optimizing AR and if the domain of uncertainty is reduced, the size increases. As mentioned in section 1, the attainable region defines the limits of achievability (performance targets) by a system. This implies that considering uncertainty has greatly reduced the limits of achievability by the system even though we have benefited from increased robustness. The authors will also like to clarify at this point that by performance targets, the authors refer to the totality concentration of microorganisms and substrates that can be output by the different digester combinations using the fundamental processes occurring in the system. This is defined by the attainable region of the system for a given kinetics and by the self-optimizing attainable region for different kinetic variations within a defined domain.

The findings from this study are therefore highly important in making economic feasibility decisions about the performance of biogas plants especially in cases where accuracy is very necessary. Put it in another way, when assessing the economic feasibility of the anaerobic treatment process, one can now consider the economic performance of the process even in cases of uncertainty and reliably compare it with other process alternatives. In summary coupling uncertainty analysis and attainable region theory provides a systematic methodological framework for dealing with kinetic uncertainty during design of biogas digesters and hence allows biogas engineers to benefit from the advantages of model-based design. These advantages include easy digester scale-up, less experimental runs (hence less cost), as well as obtain optimal design parameters and digester configurations. This approach is therefore recommended as a reliable strategy for design of biogas plants in cases of kinetic uncertainty, which is very common with biokinetic models for anaerobic digestion.

4. Conclusion

A systematic model-based framework for the synthesis of biogas reactors under cases of kinetic uncertainty has been developed. Using the case of the modified Hill model for anaerobic digestion, the following conclusions are made:

- ➢ Identifiability analysis reveals that only 5 out of the 6 model parameters can be candidates for parameter estimation. The 5 potential candidates for parameter estimation gives a total of approximately 31 parameter subset combinations, with a maximum subset size of 5. Of the 31 possible subset combinations, only 18 are potentially identifiable and with a maximum identifiable subset size of 3.
- ➢ Parameter estimation indicates that despite variation in parameter uncertainty, the quality of the model fit to experimental data is not compromised and using an identifiable subset of model parameters serves reduce the degree of uncertainty (confidence interval) in model parameter estimates.
- ➢ Following sensitivity analysis, the biodegradable volatile solids are most sensitive to the acid yield coefficient (k_2) while volatile fatty acids are most sensitivity to the Monod saturation constant for volatile acids (K_{s2}) and inhibition constant (K_i). For the anaerobic microorganisms, the acidogenic bacteria is highly sensitive to the Monod saturation constant (K_{s2}), while the methanogenic archae are highly sensitive to inhibition constant (K_i).
- ➢ Uncertainty quantification reveals that of the four model states, the methanogenic archaea, shows an insignificant uncertainty band to the model inputs at certain times instants, while all the other sates show a degree of significant uncertainty to the model inputs at all times instants
- ➢ The systematic model-based framework proposed in this study has been based on the concept of attainable regions. Hence, when using attainable regions for performance targeting and digester network synthesis, we suggest that it should be mandatory to incorporate uncertainty of model prediction during construction of the attainable regions. The attainable region obtained in such cases is referred to as a self-optimizing attainable region, which is generally smaller than the attainable region. It is concluded that incorporating kinetic uncertainty onto attainable regions has greatly reduces the limits of achievability by the system even though we have benefited from increased robustness. When the AR is obtained, the boundary of the AR can be interpreted into digester structures, whereby the optimal digester structure always involves a plug flow digester in combination with either a CSTR and/or bypass streams.

In summary coupling identifiability analysis, uncertainty quantification and the attainable region theory provides a systematic methodological framework for defining the performance targets of the anaerobic treatment process under conditions of uncertainty. It is also worth mentioning that even though the study is based on the anaerobic treatment process, the framework can be applied to optimally design other environmental chemical processes, which can be described with a kinetic model.

More research is needed to extend the concept of self-optimizing attainable regions in the field of anaerobic digestion. This study has focused on kinetic uncertainty and it would be interesting to assess the effects of other potential sources of uncertainty (such as substrate characteristics, presence of inhibitions or temperature variations) on the performance targets (defined by the self-optimizing attainable regions) of the anaerobic treatment process.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

Our team expresses gratitude to the following institutions; The Brew-Hammond Energy Centre, KNUST Ghana, The Water and Environmental Engineering Group, NTNU Ålesund and the Abunde Sustainable Engineering Group (AbundeSEG) for its immense technical support.

Nomenclature

X_{1in}	Influent concentration of acidogenic bacteria ($g\ ac./L$)
A_f	Acidity factor ($g\ VFA/L)/(g\ BVS/L$)
B_0	Biodegradability constant ($g\ BVS/L)/(g\ VS/L$)
K_{d_1}	Specific death rate of acidogenic bacteria (d^{-1})
K_{d_2}	Specific death rate of methanogenic bacteria (d^{-1})
K_i	VFA inhibition constant ($g\ VFA/L$)
K_{i1}	VFA inhibition constant for acidogenic bacteria ($g\ VFA/L$)
K_{i2}	VFA inhibition constant for methanogenic bacteria ($g\ VFA/L$)
K_s	Monod half-saturation constant (g/L)
K_{s1}	Monod half-saturation constant for acidogenic bacteria ($g\ BVS/L$)
K_{s2}	Monod half-saturation constant for acidogenic bacteria ($g\ VFA/L$)
Q_{CH_4}	Methane gas flow rate ($L\ CH_4/d$)
$S_{1_{in}}$	Influent concentration of biodegradable volatile solids ($g\ BVS/L$)
$S_{2_{in}}$	Influent concentration of volatile fatty acids ($g\ VFA/L$)
S_1	Concentration of biodegradable volatile solids in bioreactor ($g\ BVS/L$)
S_2	Concentration of volatile fatty acids in bioreactor ($g\ VFA/L$)
$S_a(t)$	Matrix of absolute sensitivities
S_{in}	Influent concentration of volatile solids ($g\ VS/L$)
S_{nd}	Non-dimensional sensitivity
$X_{2_{in}}$	Influent concentration of methanogenic bacteria ($g\ me./L$)
X_1	Concentration of acidogenic bacteria in bioreactor ($g\ ac./L$)
X_2	Concentration of methanogenic bacteria in bioreactor ($g\ me./L$)
X_{in}	Influent biomass concentration (g/L)
k_1	Yield constant ($g\ BVS/g\ ac./L$)
k_2	Yield constant ($g\ VFA/g\ ac./L$)
k_3	Yield constant ($gVFA/g\ me./L$)

$t_{v,\alpha/2}$	Student t-distribution parameter
δ_k^{msqr}	Root mean squared sensitivity
μ_1	Specific growth rate of acidogenic bacteria (d^{-1})
μ_{1m}	Maximum specific growth rate of acidogenic bacteria (d^{-1})
μ_2	Specific growth rate of methanogenic bacteria (d^{-1})
μ_{2m}	Maximum specific growth rate of methanogenic bacteria (d^{-1})
μ_m	Specific growth rate of bacteria (d^{-1})
D	Dilution rate (d^{-1})
HRT	Hydraulic retention time (d)
S	Substrate concentration (g/L)
T	Reactor temperature ($^{\circ}C$)
V	Volume of methane bioreactor (L)
VFA_{in}	Influent concentration of volatile fatty acids ($g\ VFA/\ L$)
VS	Volatile solids
VSL	Volatile solids loading ($g\ VS/L$)
W	non-dimensional scaling/weighting matrix
f	Inhibition factor
ϑ	Acidogenic fraction
Y	Model states
β	Parameter set
Y_0	Initial Condition
N	Number of state variables
n	Number of data points
p	Number of parameters
S_{norm}	Normalized sensitivity measure
γ_k	Collinearity index
λ_k	Eigen values of normalized sensitivity matrix
$J(\beta)$	Least Square Criterion
$\nabla J(\beta)$	Gradient of Least Square Criterion
Y_i^{obs}	Observable states
H	Observation matrix
$M(t,Y,\beta)$	Discretized model
$L(Y,\beta,\lambda)$	Lagrangian
w	Descent direction
ρ	Step length

Funding

This work was supported by EnPe-NORAD under the project Upgrading Education and Research Capacity in Renewable Energy Technologies (UPERC-RET).

References

Abunde Neba, F., Asiedu, N.Y., Addo, A., Morken, J., Østerhus, S.W., Seidu, R., 2019a. A coupled modeling of design and investment parameters for optimal operation of methane bioreactors: attainable region concept approach. Renew. Energy.

Abunde Neba, F., Asiedu, N.Y., Addo, A., Morken, J., Østerhus, S.W., Seidu, R., 2019b. Simulation of Two-Dimensional Attainable Regions and its Application to Model Digester Structures for Maximum Stability of Anaerobic Treatment Process. Water Research, p. 114891.

Abunde Neba, F., Asiedu, N.Y., Addo, A., Morken, J., Østerhus, S.W., Seidu, R., 2019c. Use of attainable regions for synthesis and optimization of multistage anaerobic digesters. Appl. Energy 242, 334–350.

Abunde Neba, F., Asiedu, N.Y., Addo, A., Morken, J., Østerhus, S.W., Seidu, R., 2020a. Biodigester rapid analysis and design system (B-Rades): a candidate attainable region-based simulator for the synthesis of biogas reactor structures. Comput. Chem. Eng. 132, 106607.

Abunde Neba, F., Asiedu, N.Y., Addo, A., Seidu, R., 2020b. Attainable regions and fuzzy multi-criteria decisions: modeling a novel configuration of methane bioreactor using experimental limits of operation. Bioresour. Technol. 295, 122273.

Batstone, D.J., 2006. Mathematical modelling of anaerobic reactors treating domestic wastewater: rational criteria for model use. Rev. Environ. Sci. Biotechnol. 5, 57–71.

Benítez, M., Bermúdez, A., Rodríguez-Calo, J.F., 2017. Adjoint method for parameter identification problems in models of stirred tank chemical reactors. Chem. Eng. Res. Des. 123, 214–229.

Brun, R., Kühni, M., Siegrist, H., Gujer, W., Reichert, P., 2002. Practical identifiability of Asm2d parameters—systematic selection and tuning of parameter subsets. Water Res. 36, 4113–4127.

Chen, J.L., Ortiz, R., Steele, T.W.J., Stuckey, D.C., 2014. Toxicants inhibiting anaerobic digestion: a review. Biotechnol. Adv. 32, 1523–1534.

Chen, Y., Cheng, J.J., Creamer, K.S., 2008. Inhibition of anaerobic digestion process: a review. Bioresour. Technol. 99, 4044–4064.

Donoso-Bravo, A., Mailier, J., Martin, C., Rodríguez, J., Aceves-Lara, C.A., Wouwer, A.V., 2011. Model selection, identification and validation in anaerobic digestion: a review. Water Res. 45, 5347–5364.

Donoso-Bravo, A., Mailier, J., Ruiz-Filippi, G., Wouwer, A.V., 2013. Identification in an anaerobic batch system: global sensitivity analysis, multi-start strategy and optimization criterion selection. Bioproc. Biosyst. Eng. 36, 35–43.

EPA, U. S. E. P. A, 2006. Biosolids Technology Fact Sheet, Multi-Stage Anaerobic Digestion. National Service Center or Environmental Publications (NSCEP).

Finn, H., Rune, B., Bernt, L., 2013. Adapting dynamic mathematical models to a pilot anaerobic digestion reactor. Model. Identif. Control 34, 35–54.

Gausemeier, J., Frank, U., Schmidt, A., Steffen, D., 2006. Towards a design methodology for self-optimizing systems. In: Elmaraghy, H.A., Elmaraghy, W.H. (Eds.), Advances in Design. Springer London, London.

Henze, M., Van Loosdrecht, M.C.M., Ekama, G.A., Brdjanovic, D., 2008. Biological Wastewater Treatment. Iwa Publishing.

Hildebrandt, D., Glasser, D., 1990. The attainable region and optimal reactor structures. Chem. Eng. Sci. 45, 2161–2168.

Hildebrandt, D., Glasser, D., Crowe, C.M., 1990. Geometry of the attainable region generated by reaction and mixing: with and without constraints. Ind. Eng. Chem. Res. 29, 49–58.

Jäschke, J., Cao, Y., Kariwala, V., 2017. Self-optimizing control – a survey. Annu. Rev. Contr. 43, 199–223.

Kythreotou, N., Florides, G., Tassou, S.A., 2014. A review of simple to scientific models for anaerobic digestion. Renew. Energy 71, 701–714.

Ming, D., Glasser, D., Hildebrandt, D., Glasser, B., Metzer, M., 2016. Attainable Region Theory: an Introduction to Choosing an Optimal Reactor. John Wiley & Sons, Inc, Hoboken, New Jersey.

Morales-Rodriguez, R., Meyer, A.S., Gernaey, K.V., Sin, G., 2012. A framework for model-based optimization of bioprocesses under uncertainty: lignocellulosic ethanol production case. Comput. Chem. Eng. 42, 115–129.

Permin, E., Bertelsmeier, F., Blum, M., Bützler, J., Haag, S., Kuz, S., Özdemir, D., Stemmler, S., Thombansen, U., Schmitt, R., Brecher, C., Schlick, C., Abel, D., Poprawe, R., Loosen, P., Schulz, W., Schuh, G., 2016. Self-optimizing production systems. Procedia Cirp 41, 417–422.

Roulston, M.S., 1999. Data Assimilation in Chaotic systems,Astrophysical and Geophysical Flows as Dynamical Systems 1998 Summer Study Program in Geophysical Fluid Dynamics.

Sin, G., Gernaey, K.V., Lantz, A.E., 2009. Good Modeling Practice for Pat Applications: Propagation of Input Uncertainty and Sensitivity Analysis, vol 25, pp. 1043–1053.

Sin, G., Meyer, A.S., Gernaey, K.V., 2010a. Assessing reliability of cellulose hydrolysis models to support biofuel process design—identifiability and uncertainty analysis. Comput. Chem. Eng. 34, 1385–1392.

Sin, G., Meyer, A.S., Gernaey, K.V., 2010b. Assessing reliability of cellulose hydrolysis models to support biofuel process design—identifiability and uncertainty analysis. Comput. Chem. Eng. 34, 1385–1392.

Sin, G., Vanrolleghem, P.A., 2007. Extensions to modeling aerobic carbon degradation using combined respirometric–titrimetric measurements in view of activated sludge model calibration. Water Res. 41, 3345–3358.

Skogestad, S., 2000. Plantwide control: the search for the self-optimizing control structure. J. Process Control 10, 487–507.

Wang, L.K., Shammas, N.K., Hung, Y.-T., 2007. Biosolids Treatment Processes. Humana Press Inc, New Jersey.

Yu, L., Wensel, P.C., Ma, J., Chen, S., 2013. Mathematical modeling in anaerobic digestion (Ad). Bioremediation & Biodegradation, pp. 2155–6199.

Zaher, U., Pandey, P., Chen, S., 2009. A simple elemental continuity based model application to study the anaerobic microbial activity for the treatment of dairy manure. Appl. Math. Model. 33, 3553–3564.

List of Appendices

A1: Anaerobic treatability experiment with abattoir waste

The table below shows experimental measurements of cumulative biogas, obtained from anaerobic digestion experiments of abattoir effluent and used for AR construction (Paper 4)

Retention time (days)	Cumulative biogas production
1	0
2	0
3	0.005
4	0.03
5	0.12
6	0.48
7	0.76
9	1.53
10	2.78
11	4.3
12	5.4
13	7.3
14	8.2
15	9
16	10.53
17	11.02
18	11.54
19	11.63
20	11.91
21	11.91
22	11.91
23	11.91
24	11.91
25	11.91
26	11.91
27	11.91
28	11.91
29	11.91
30	13.38