

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



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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.

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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 dairy, 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,

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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_{sac}	monod half-saturation constant for acidogenic bacteria (gBVS/L)
K_{sme}	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 ($^{\circ}\text{C}$)
T_{min}	minimum temperature at which growth rate is zero ($^{\circ}\text{C}$)
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_{SBVS}	reaction rate for biodegradable volatile solids (gBVS/L/d)
r_{SVFA}	reaction rate for volatile fatty acids (gVFA/L/d)
r_{Xac}	reaction rate for acidogenic bacteria (gac. /L/d)
r_{Xme}	reaction rate for methanogenic archae (gme. /L/d)
$t_{v,\alpha/2}$	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
μ_{mac}	maximum specific growth rate of acidogenic bacteria (d ⁻¹)
μ_{mme}	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 ($^{\circ}\text{C}^{-1} \text{ h}^{-0.5}$)
C	Ratkowsky parameter C ($^{\circ}\text{C}^{-1}$)
T	reactor temperature ($^{\circ}\text{C}$)
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 archaea, 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

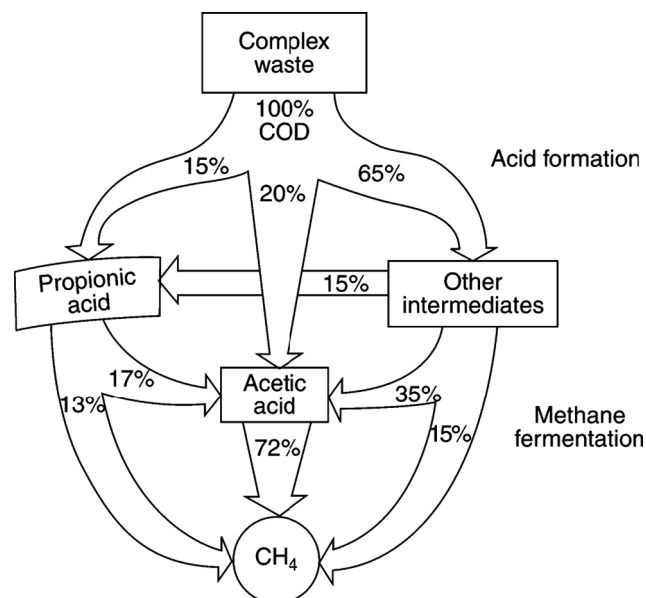


Fig. 1. Biochemical pathways for volatile solids reduction and methane generation [2].

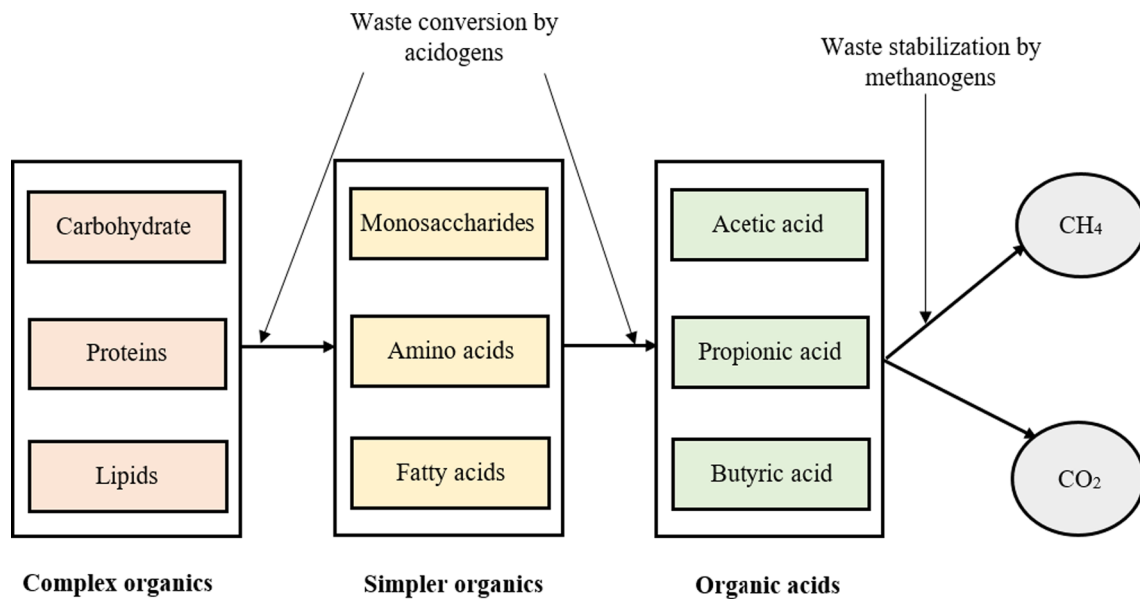


Fig. 2. Simplified two-stage scheme for anaerobic treatment process.

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

$$\mu_{ac} = \mu_{mac} \frac{S_{BVS}}{K_{sac} + S_{BVS}} \tag{5}$$

$$\mu_{me} = \mu_{mme} \frac{S_{VFA}}{K_{sme} + S_{VFA} \left(1 + \frac{S_{VFA}}{K_{ime}}\right)} \tag{6}$$

(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 archaea 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.

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

$$X_{me0} = (1 - \vartheta) X_0 \tag{10}$$

The maximum growth rates of acid-forming (μ_{mac}) and methane forming bacteria (μ_{mme}) 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.

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

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

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

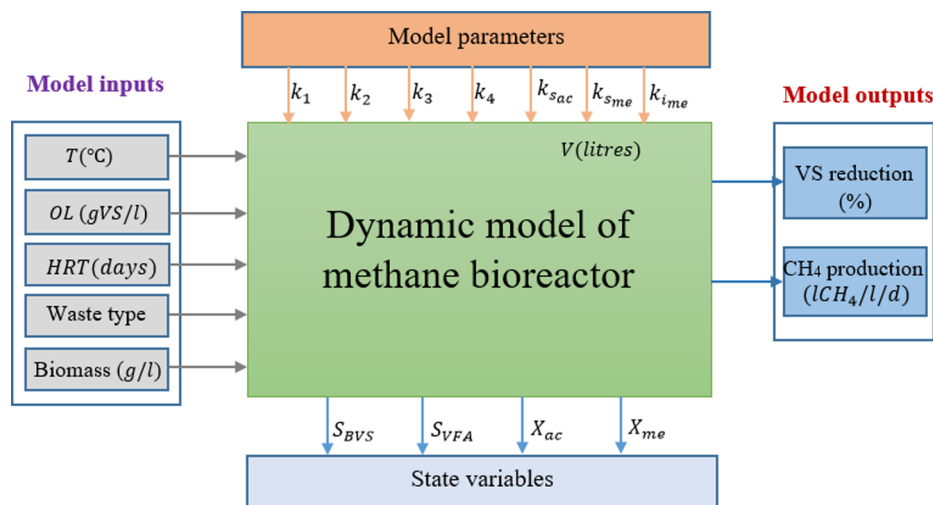


Fig. 3. Model of methane bioreactor showing inputs, outputs, parameters and state variables.

(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].

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

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

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 CO₂ and ammonia gas is not considered because their effect is already lumped into B₀ 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.

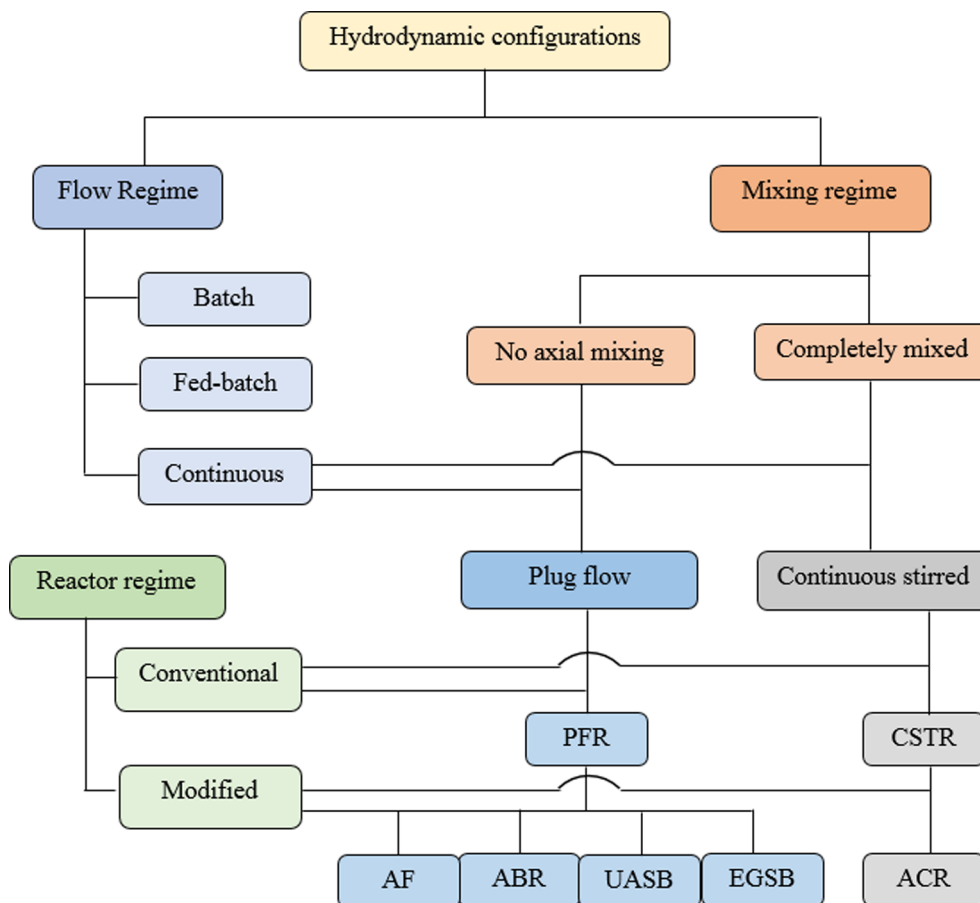


Fig. 4. Classification of hydrodynamic configurations of methane bioreactors.

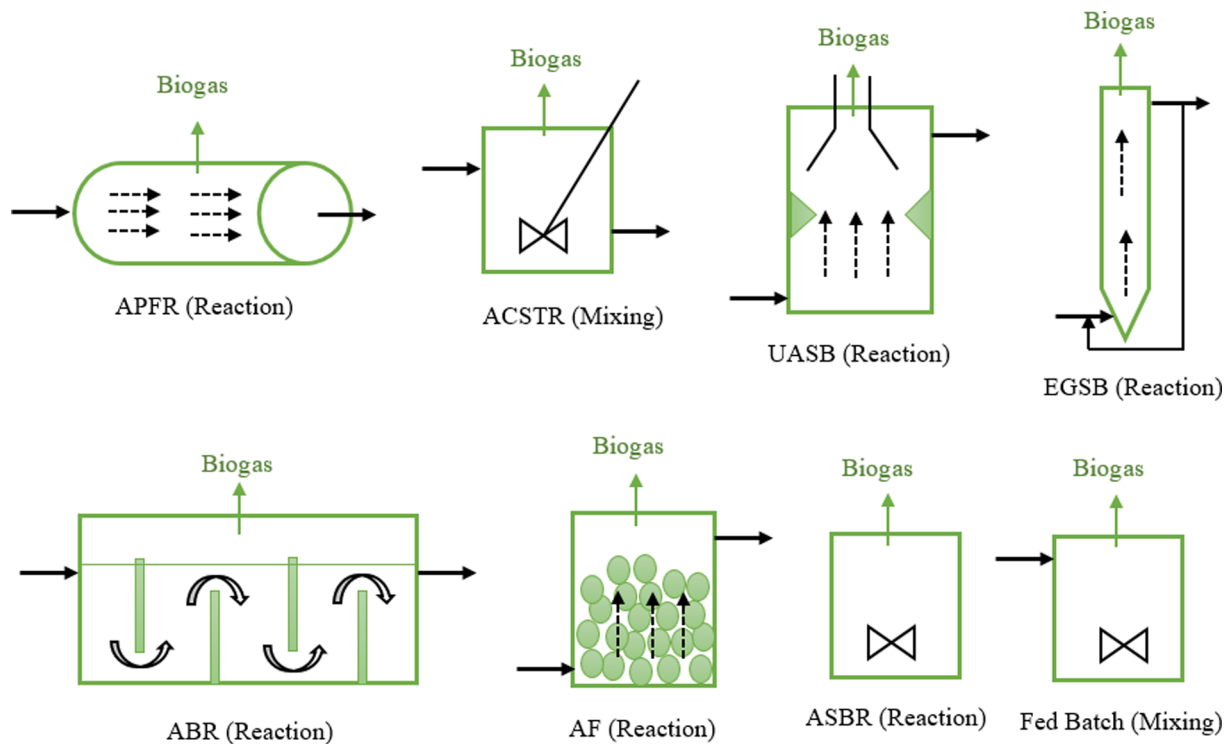


Fig. 5. Grouping of methane bioreactors types into fundamental processes of mixing and reaction.

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].

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

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 ³ d)	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.

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

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

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

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

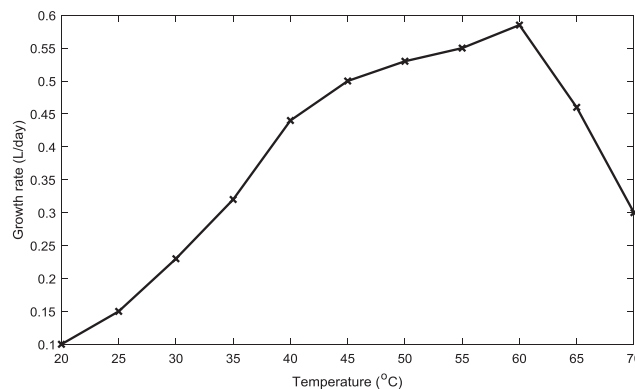


Fig. 6. Chen and Hashimoto curve for temperature dependence of growth rate.

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

(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_{sac} , K_{sme} and K_{ime} 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).

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

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).



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).

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

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).

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

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

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).

$$X_{ac} = X_{ac0} + \frac{1}{k_2}[S_{VFA} - S_{VFA0} + k_3(X_{me} - X_{me0})] \quad (28)$$

$$S_{BVS} = S_{BVS0} - \frac{k_1}{k_2}[S_{VFA} - S_{VFA0} + k_3(X_{me} - X_{me0})] \quad (29)$$

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 hull 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.

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

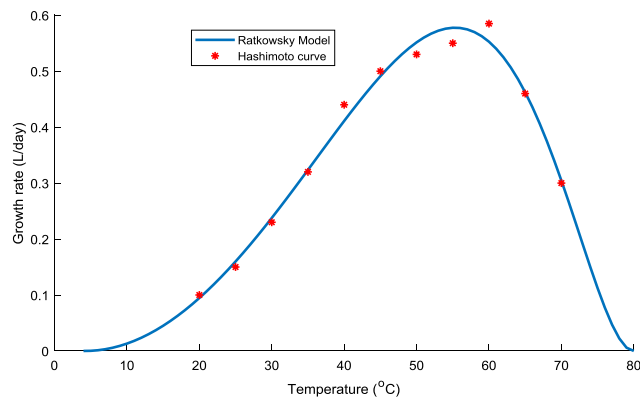


Fig. 7. Fitting of temperature dependence model to test data.

$$X_{me} = \frac{VSR \times S_{VS0}}{k_3\mu_{me} \times 100} \quad (31)$$

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 it 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

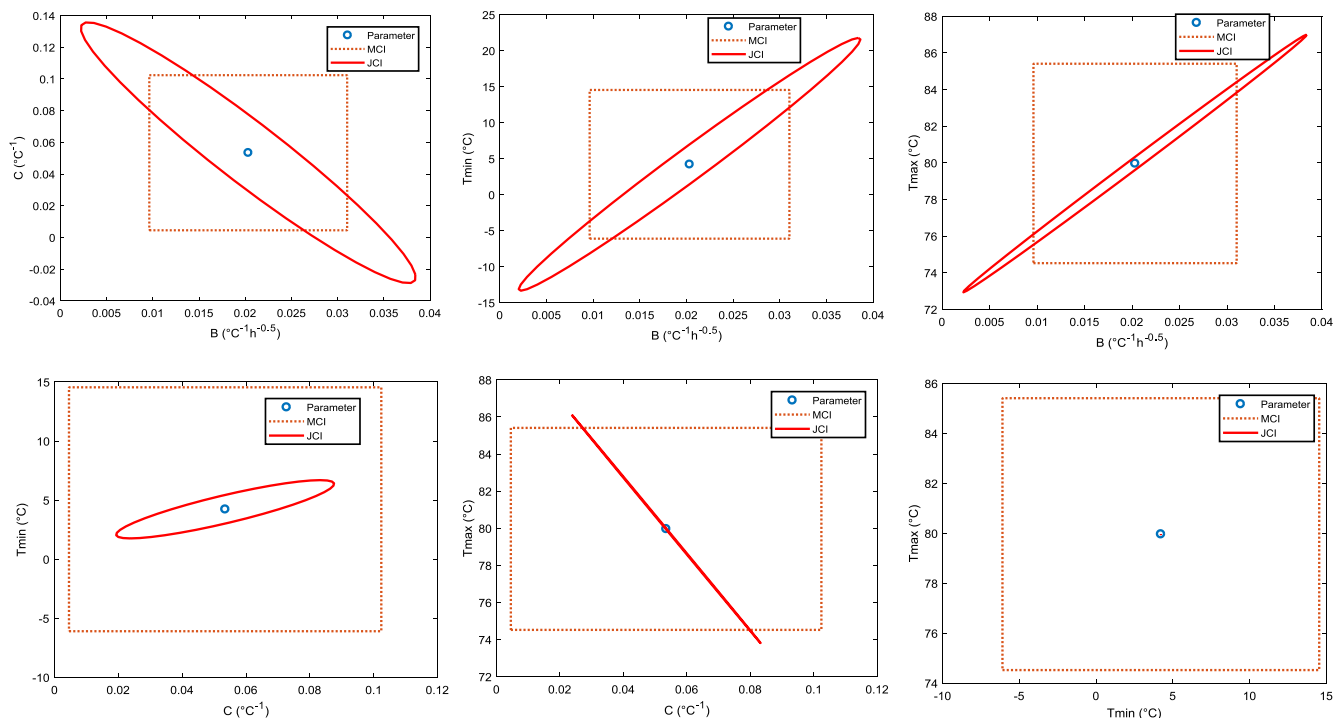


Fig. 8. Confidence contours of parameter estimates for temperature dependence model.

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

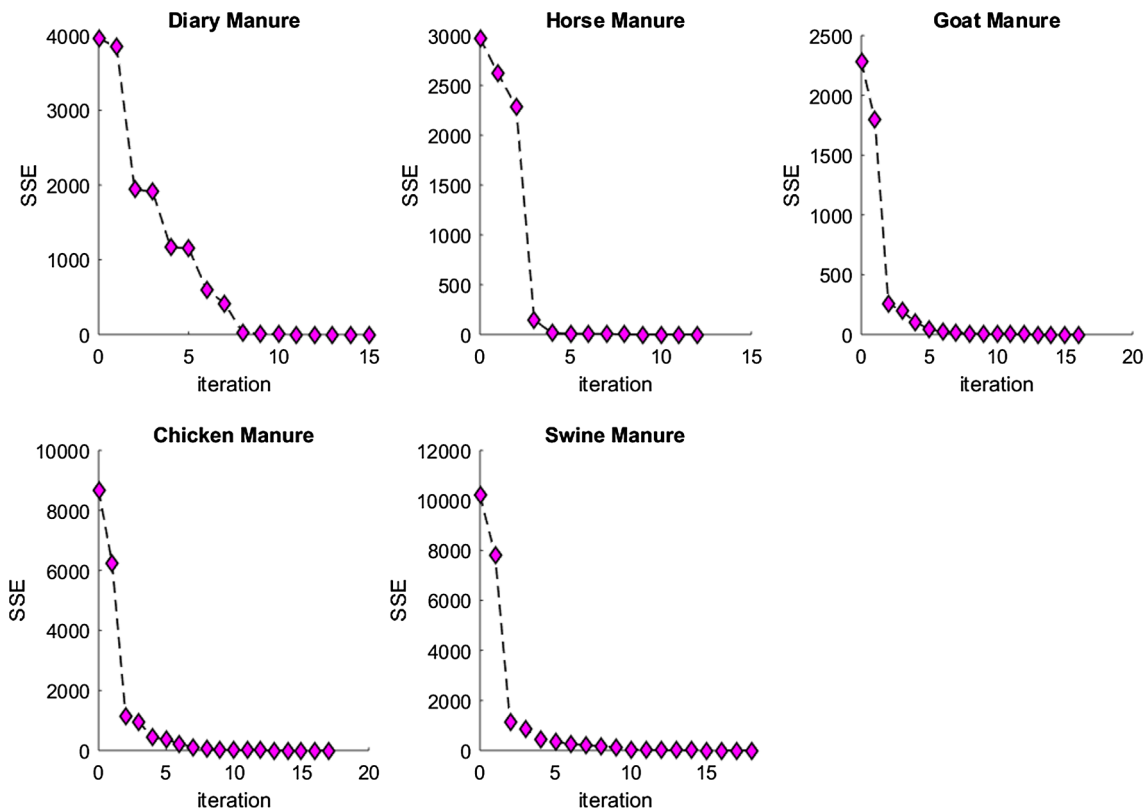


Fig. 9. Convergence history of parameter estimation process for different organic substrates.

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}	
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.

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

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

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

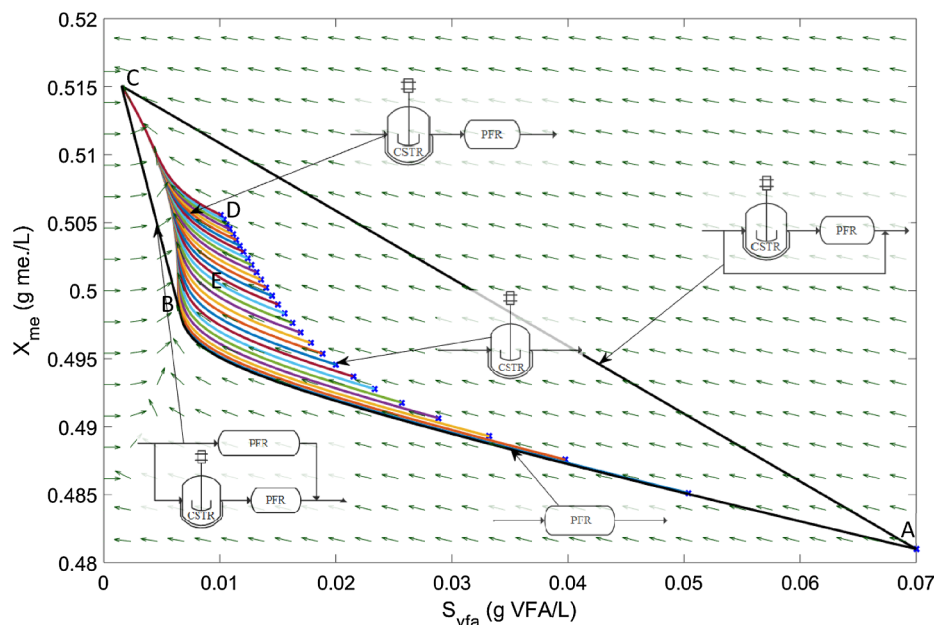


Fig. 10. Attainable region for anaerobic treatment of dairy manure in 2D space.

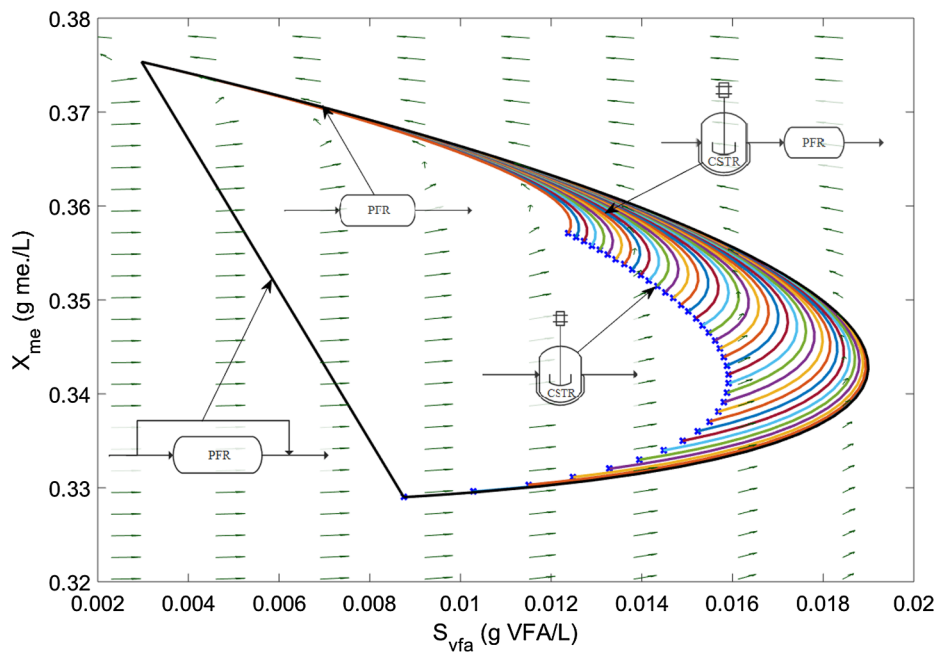


Fig. 11. Attainable region for anaerobic treatment of horse manure in 2D space.

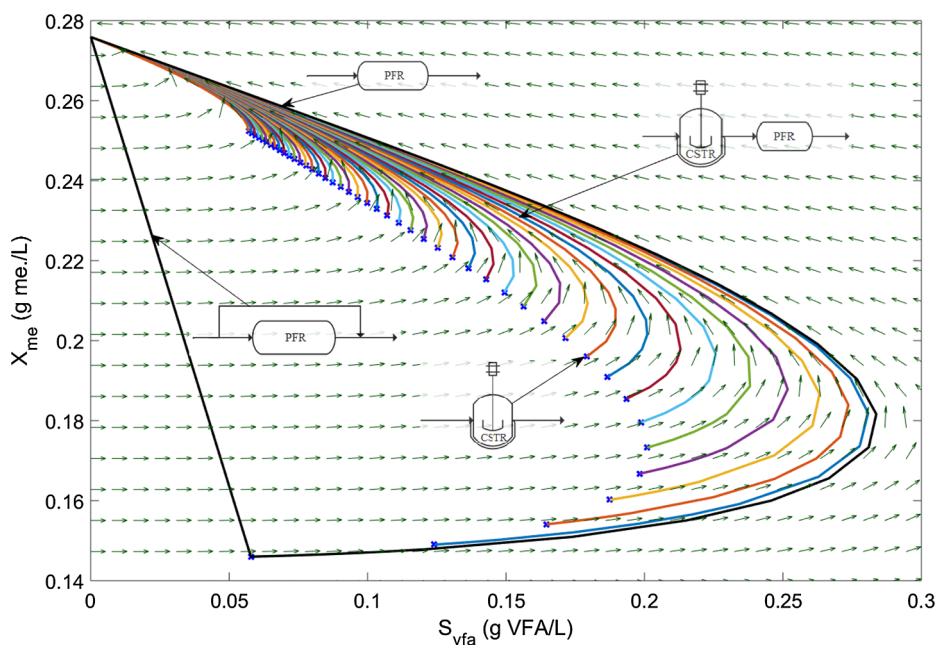


Fig. 12. Attainable region for anaerobic treatment of goat manure in 2D space.

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

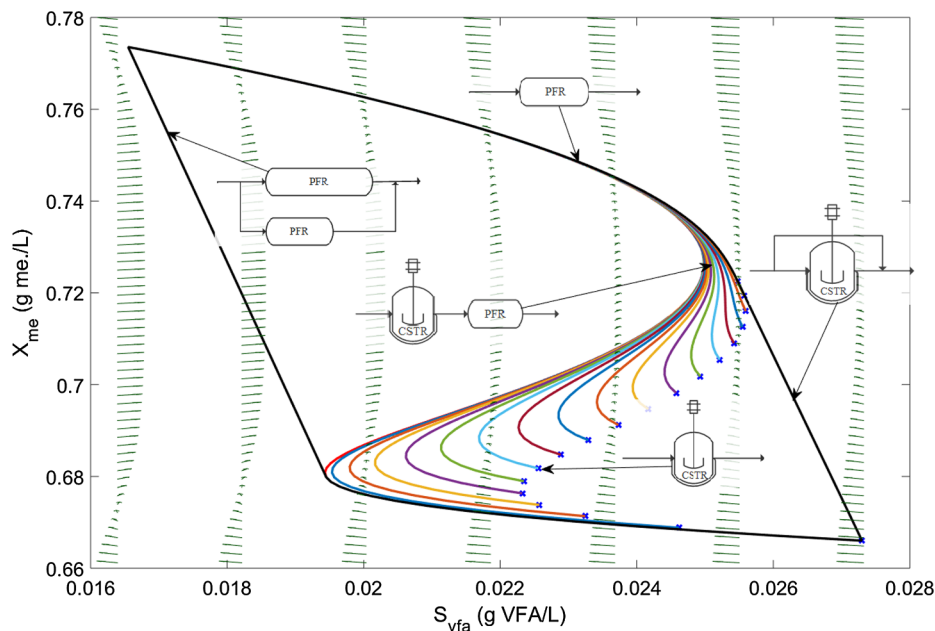


Fig. 13. Attainable region for anaerobic treatment of chicken manure in 2D space.

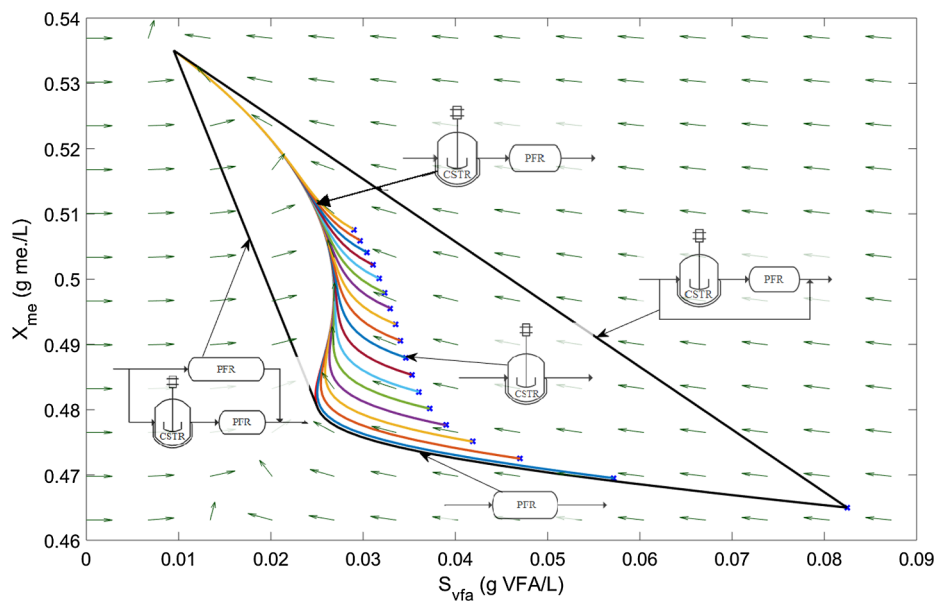


Fig. 14. Attainable region for anaerobic treatment of swine manure in 2D space.

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

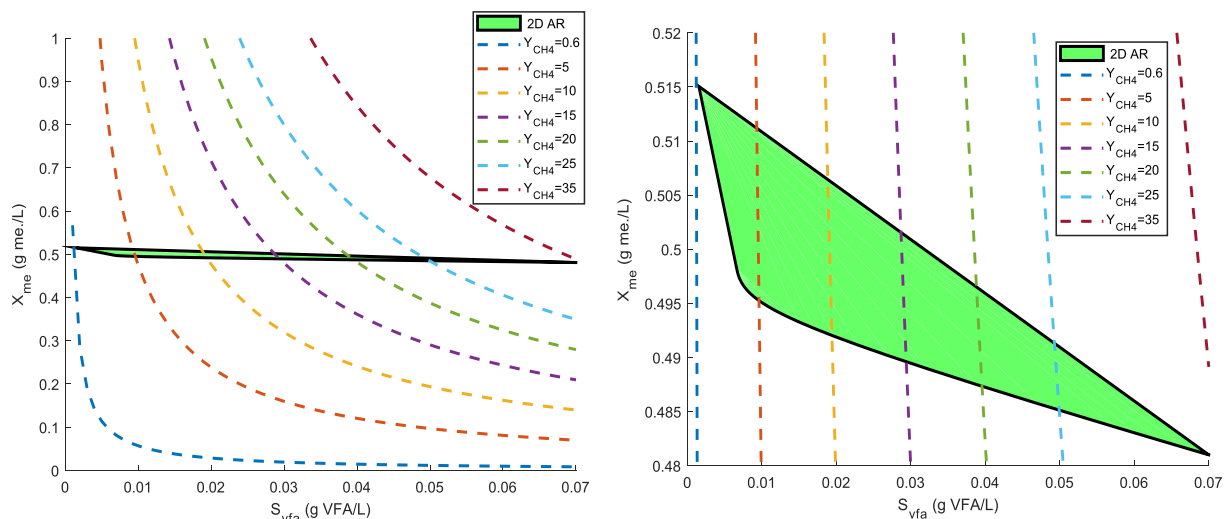


Fig. 15. Contours of volumetric methane productivity overlaid onto AR for dairy manure.

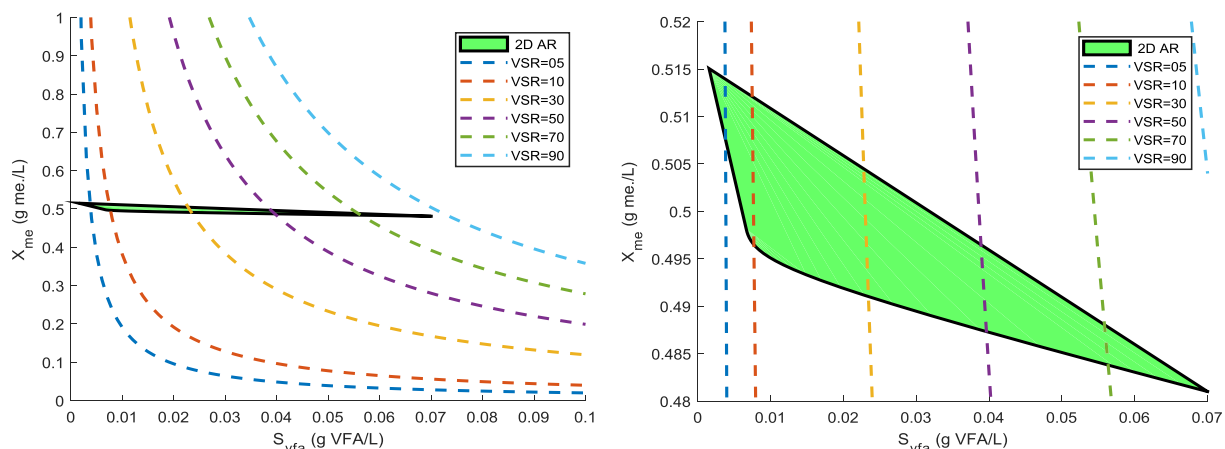


Fig. 16. Contours of volatile solids reduction overlaid onto AR for dairy manure.

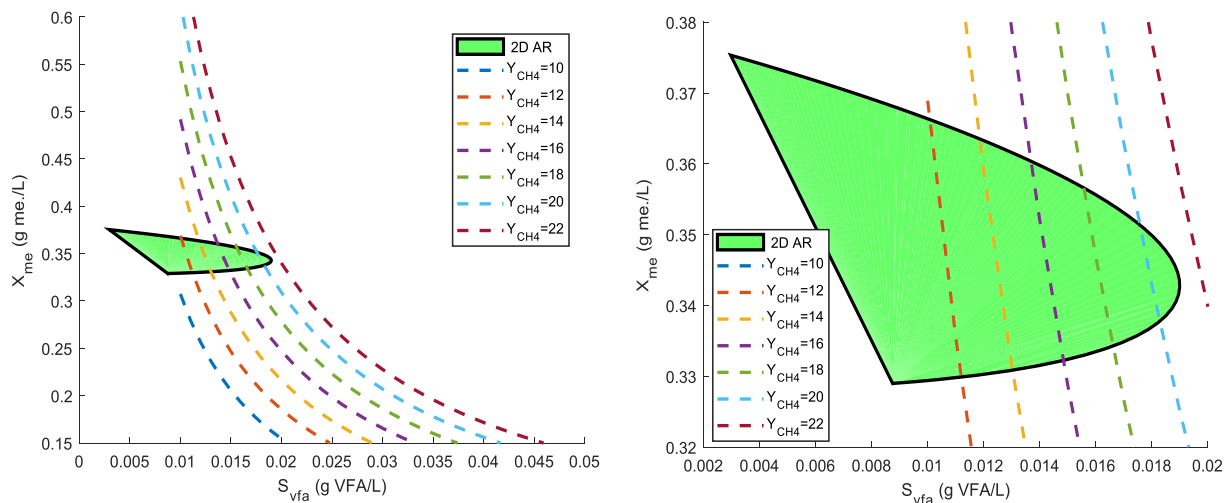


Fig. 17. Contours of volumetric methane productivity overlaid onto AR for horse manure.

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

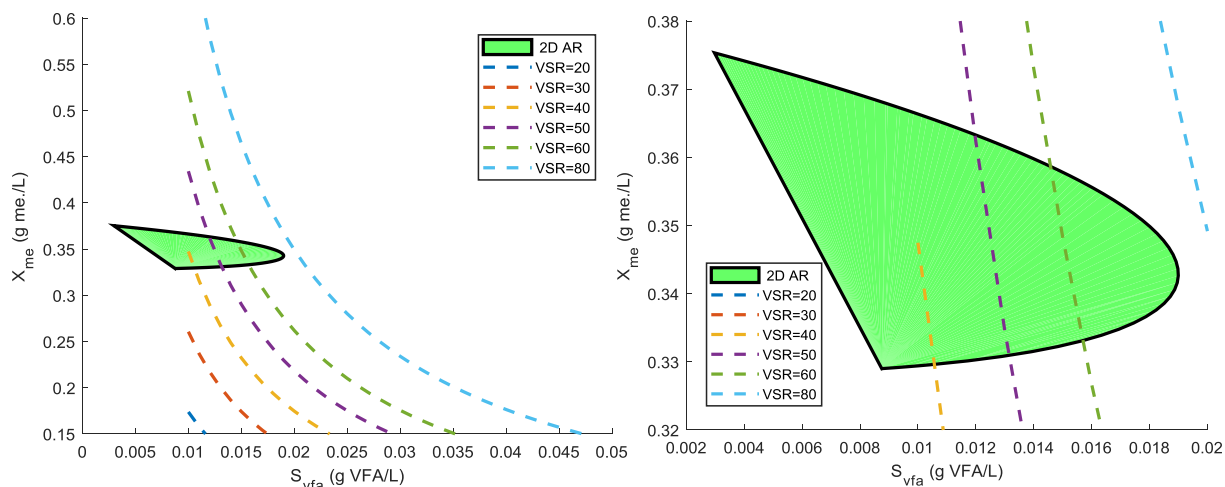


Fig. 18. Contours of volatile solids reduction overlaid onto AR for horse manure.

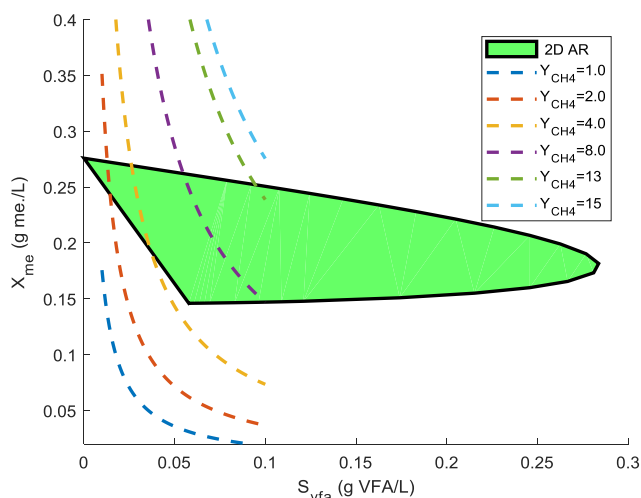


Fig. 19. Contours of volumetric methane productivity overlaid onto AR for goat manure.

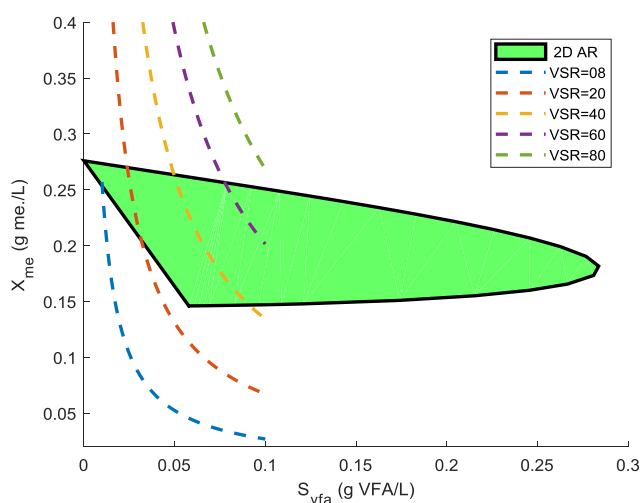


Fig. 20. Contours of volatile solids reduction overlaid onto AR for goat manure.

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:

- Identify 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 dairy, 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

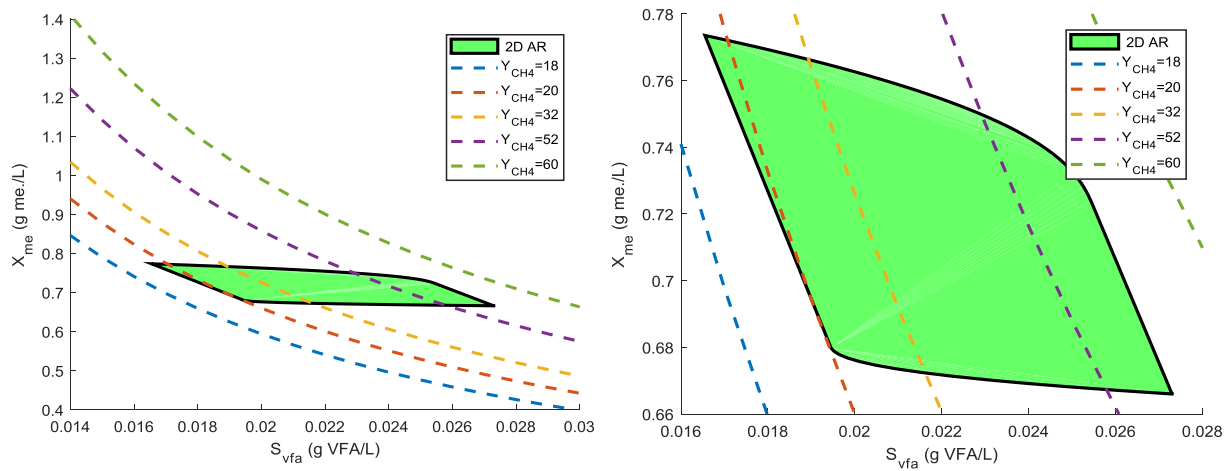


Fig. 21. Contours of volumetric methane productivity overlaid onto AR for chicken manure.

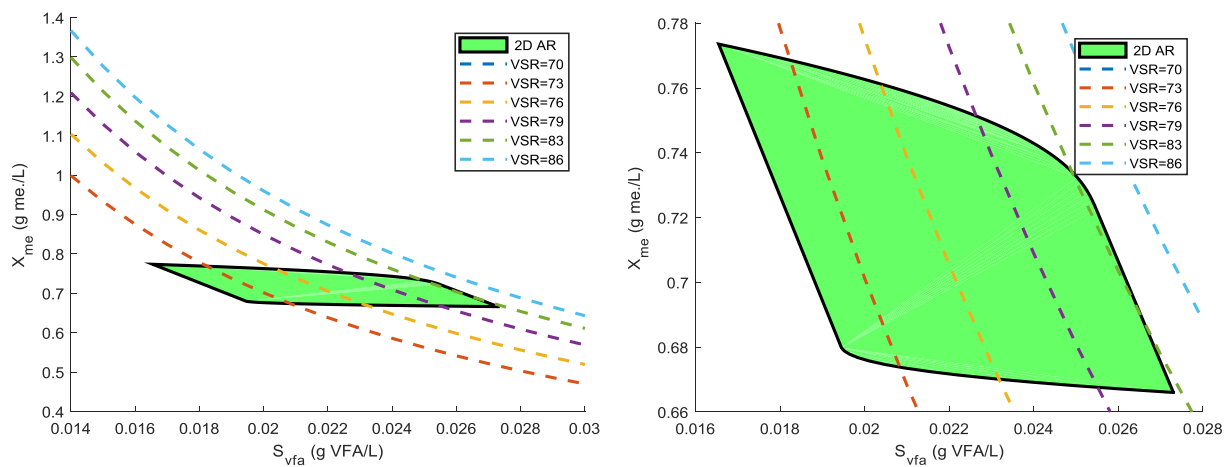


Fig. 22. Contours of volatile solids reduction overlaid onto AR for chicken manure.

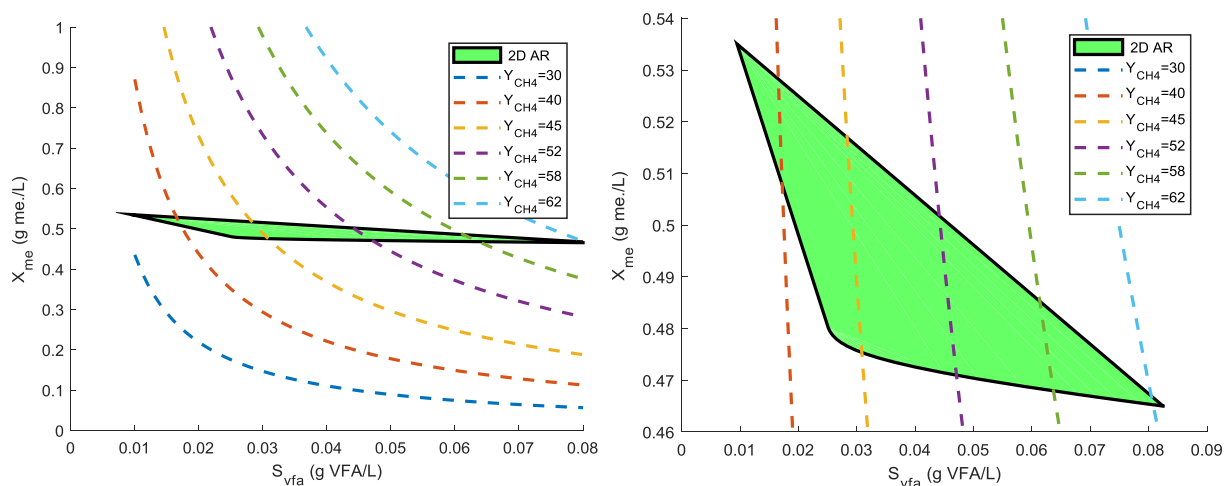


Fig. 23. Contours of volumetric methane productivity overlaid onto AR for swine manure.

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 dairy, 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

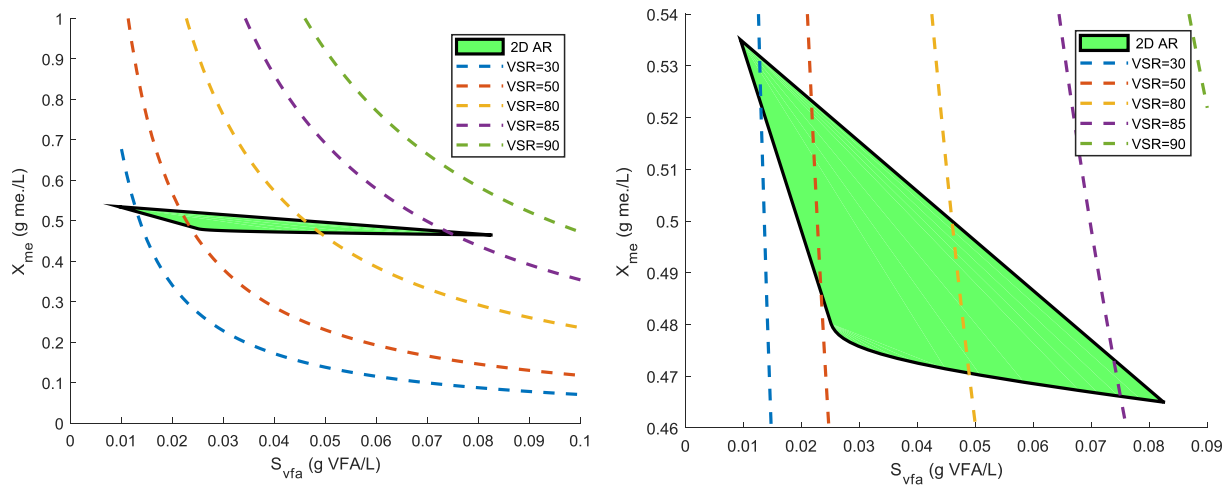


Fig. 24. Contours of volatile solids reduction overlaid onto AR for swine manure.

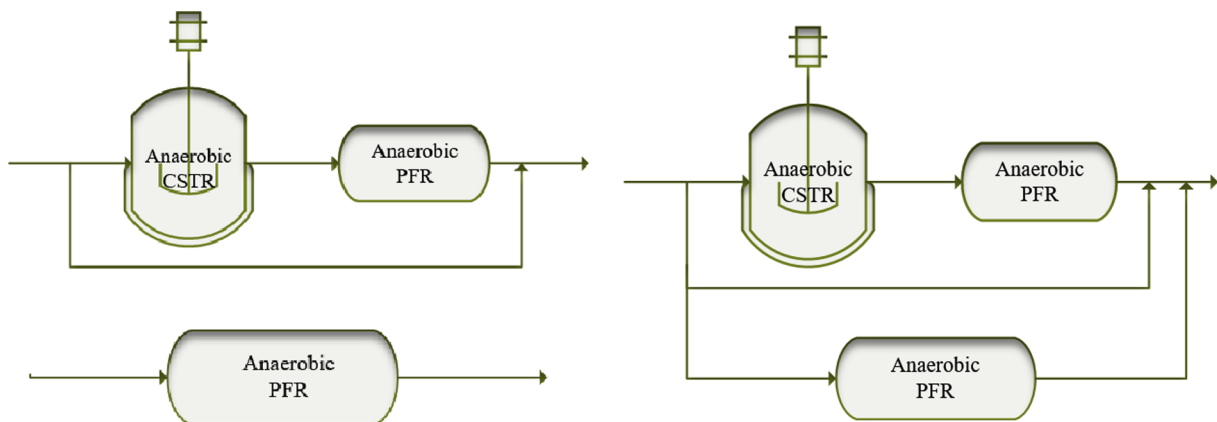


Fig. 25. Digester structures to attain methane productivity of 25 and 52 l/m³/d respectively diary manure and swine manure.

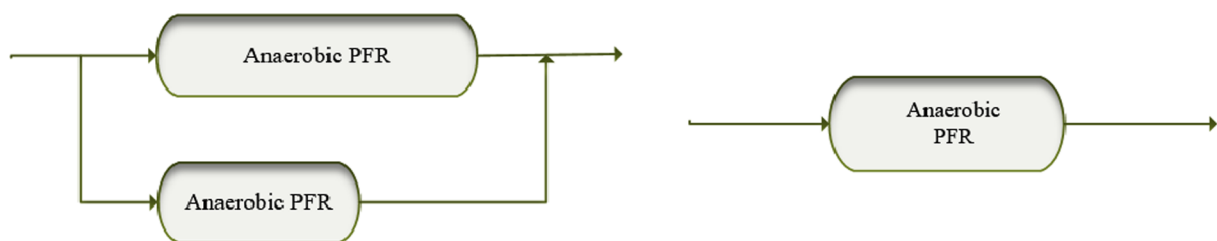


Fig. 26. Digester structures that can attain methane productivity of 18 and 20 l/m³/d respectively for horse manure, chicken manure.

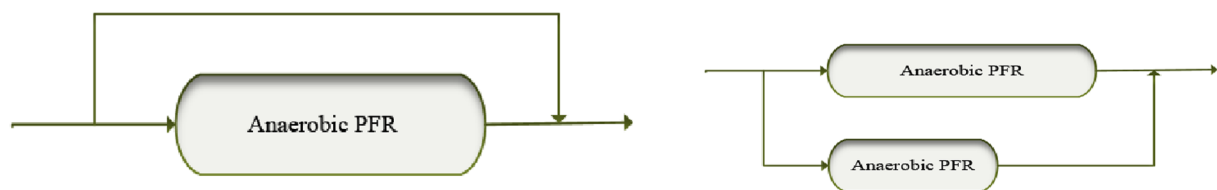


Fig. 27. Digester structures to attain methane productivity of 4.0 l/m³/d for goat manure.

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.

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