

Short communication

A novel hybrid digestion-gasification process integrated with membranes for efficient conversion of biomass to bio-alcohols

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Abstract

There is an urgent need to develop technologies which enable the conversion of biomass into liquid biofuels to fill the gap between limited fossil fuel supplies and increasing worldwide demand. In order to achieve the EU 2030 vision of at least 15% of the fuels used in the road transportation sector will be biofuels derived from non-food biomass feedstocks, the R&D of clean, inexpensive, highly end-user compatible biofuels from a virtually inexhaustible source of biomass should be pursued to make breakthroughs in cost-effective biomass to liquid biofuels (BTL) technologies. Thus, an innovative, consolidated, and sustainable technology using a hybrid digestion-gasification process integrated with membranes to produce next generation bio-alcohols from different biomass feedstocks was designed. The proposed concept was theoretically estimated to achieve an overall BTL efficiency of 44% and a cost reduction for bioalcohol production of 18.6%. Moreover, this technology can potentially achieve an overall CO₂ emission reduction of > 75% for road transport based on the preliminary analysis.

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Keywords: Biofuels; Biomass; Chemical looping gasification; Membrane; Anaerobic digestion

1. Introduction

The transport sector consumes 23% of global primary energy [1] and emits large amount of greenhouse gas due to the major use of the petroleum derived fuels. With the inevitable depletion of petroleum-based resources, there has been an increasing worldwide interest in finding alternative resources. Biomass is one of the most important primary and renewable energy resources for sustainable future [2]. There is an urgent need to develop technologies which enable the conversion of lignocellulosic and waste biomass resources into useful energy, from thermodynamic efficiency and an environmental impact standpoint [3]. Substantial attention has been received towards the development of technically feasible methods to convert biomass into biofuels [4], which will reduce not only the dependence on fossil fuels, but also environmental

pollution [5]. Biofuels are the promising way to provide adequate fuels at a time when yields from existing oil fields are declining and new fields are not yet up and running. Biofuels can help fill the gap between limited fuel supplies and increasing worldwide demand that is almost sure to widen in the coming years. Directive 2009/28/EC on promotion of renewable energy sources has set a 10% of renewables in the transport sector as target by 2020, and due to the lack of large-scale alternatives (especially heavy duty transport, shipping and aviation will require significant fuel demands, and their electrification potential regarding e-mobility is quite low), biofuels shall be the major contributors. However, the current biomass-to-biofuel technologies are still uncompetitive due to the challenges listed in Table 1. The anaerobic digestion research should be continued on searching for efficient and cost-effective integrated systems to produce biomethane and utilize the digestates. The biomass gasification should focus on bringing down energy consumption in biomass pre-treatment and the cost of the required large amounts of pure oxygen.

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Table 1
Technology gap between the status and the need.

Challenge	The status	The need
Feedstock potential	Limited feedstock supply chain	Next generation non-food feedstock, high resource potential
BTL efficiency	Conventional gasification technology with low carbon conversion efficiency; Syngas fermentation to ethanol with low yield; Energy intensive separation process	Advanced solid biomass conversion technology; Syngas catalytic conversion to advanced biofuels; Energy efficient, environmentally friendly separation technology
End-user compatibility	Compatible with low content bioethanol (10%) for most car engines	Better end-user compatibility (mixed bio-alcohols containing mainly butanol and less methanol)

Environmentally friendly separation technologies such as membranes (e.g., biogas upgrading [6]) should be integrated into bioenergy plants to improve energy efficiency of a whole biofuel production process. Novel thermal conversion of biomass to syngas such as catalytic and chemical looping gasification were reported in the literature to improve the conversion efficiency [7,8]. Moreover, the bioalcohol productivity/yield in a syngas fermentation process needs to be further improved. Therefore, the R&D of clean, inexpensive, highly end-user compatible biofuels from a virtually inexhaustible source of biomass (e.g., a variety of waste and forestry residue feedstocks) should be pursued to make breakthroughs in cost-effective biomass to liquid biofuels (BTL) conversion technologies.

This work is aiming at developing competitive biofuel production process from biomass that can significantly reduce biofuel production cost compared to the current technology. Thus, a hybrid digestion-gasification process to convert different type of biomass feedstocks to syngas was designed, and the bio-syngas is further converted bio-alcohols through thermo-catalytic reaction. Moreover, the energy efficiency of the whole process will be improved by process integration with membrane technology. The energy efficiency, the production cost, the CO₂ emissions and the sustainability of the whole process were estimated and discussed. The novelty of this concept is to enhance flexibility in the adopted technologies so that they can function within a wider range of compositions and supplies and provide process footprints that are particularly attractive related to their greenhouse gas (GHG) performance.

2. Methods

2.1. Conceptual design

Based on the technology gap described in Table 1, enlarging the feedstock supply chains of non-food biomass from both forestry residues and wastes using different conversion technology can potentially address the challenge of feedstock potential. Forestry residue is one of the most abundant materials from the forestry sector at a lower cost than starch- and sucrose-based materials of fixed renewable carbon on earth. The production of biofuels from forestry residue feedstocks opens large opportunities in terms of enhancing sustainability, avoiding land conflicts and expanding resource potential. Moreover, biogas produced from

wastes (e.g., animal manure, sewage sludge) through anaerobic digestion (AD) [9,10] provides great potential for bio-syngas production through biomethane reforming. The added fuel synthesis through bio-syngas catalytic conversion can replace a substantial amount of the petroleum-derived fuels used in the transport sector, and provide an important solution to mitigate CO₂ emissions [11]. Moreover, production of next generation biofuels of mixed (C₁–C₄) bio-alcohols (bio-methanol, bioethanol, and mainly biobutanol) is more cost-effective compared to the Fischer–Tropsch (FT) diesels. Analysis of the life cycle biomass-to-fuel energy utilization efficiency showed that biomethanol is better than FT bio-diesels [12]. Moreover, biobutanol is another type of biofuel that has several advantages such as low-volatility, higher energy content, lower water absorption, and better blending ability, which can be used in conventional combustion engines without modification. As bioethanol has already been commercially used as fuels in the road transport sector, in principle, there are no technical hurdles either in terms of vehicle application or of distribution infrastructure of the mixed (C₁–C₄) bio-alcohols. Thus, the mixed bio-alcohols will take the advantages of both biomethanol and mainly biobutanol, and their gasoline blend can provide better fuel performance (i.e., power and thermal efficiency) and lower CO₂ emissions compared to single alcohol-gasoline blend and pure gasoline as reported in the literature (power and thermal efficiency improve 8.2% and 7.8%, respectively) [13,14]. Therefore, the mixed (biobutanol enriched) bio-alcohols provide a better fuel compatibility with the petroleum-derived fuels for road transport.

A unique concept by strategically combining biological and thermochemical processes integrated with membranes to convert various biomass feedstocks to biofuels was designed (see the illustration in Fig. 1). Two parallel technical routes of anaerobic digestion (A) and gasification (B) are implemented to convert different biomass to syngas. This concept for biofuel production, utilizing forestry residues and wastes, is flexible in its feedstock requirements, and uses process synergy, integration and feedstock fractionation to ensure affordable renewable energy access that does not compromise, but rather compliment, the food production. Biogas is produced in an anaerobic digester coupled with an environmentally friendly membrane separation system for purification to produce high purity biomethane (> 98%) at a low methane loss, which improves the energy efficiency of the conversion of liquid biomass to biomethane. The upgraded biomethane is

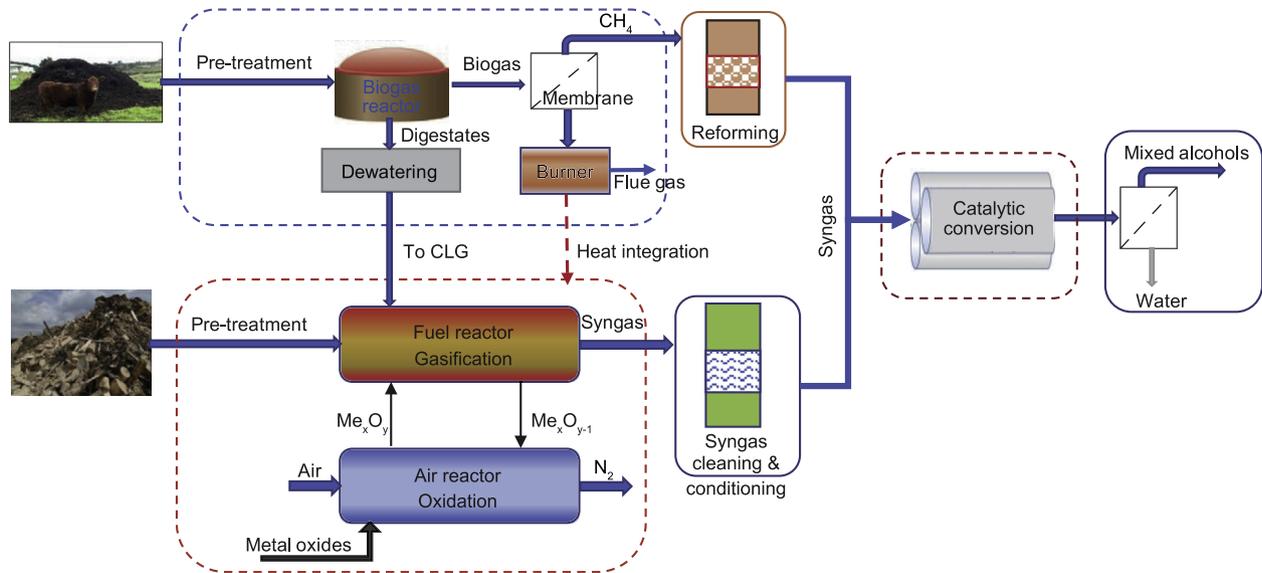


Fig. 1. Hybrid digestion-gasification concept for enhanced conversion of BTL.

converted to syngas using autothermal reforming (ATR) process which is highly efficient and mature technology for syngas production from methane compared to a steam methane reforming process (SMR). The advantage of ATR is that the H_2/CO ratio of syngas can be varied/adjusted—this is particularly important to produce selected bio-alcohols. Moreover, biomass chemical looping gasification (CLG) is employed to enhance the carbon conversion efficiency of solid biomass to syngas. The oxygen carriers (OC) such as metal ferrite [8,15,16] can be used in CLG, and act as catalysts for reforming the tar to produce cleaner syngas. The digestates produced in the biomass digestion reactor and the extra heat generated from the burning of lean biogas can be integrated to the gasification process. Overall, the combination of biomass digestion and gasification processes provides a high-efficiency technology for the conversion of biomass into bio-syngas based on the mass and heat integration. The bio-syngas produced from these two processes will be cleaned by the syngas cleaning unit and delivered to the syngas platform. The targeted bio-syngas will be catalytically converted to mixed bio-alcohols (biomethanol, bioethanol and mainly biobutanol) with better end-user compatibility (i.e., the fuels can be directly blended into petroleum-derived fuels at various contents). It should be noted that the produced mixed bio-alcohols can also be separated to produce different pure alcohols if necessary. However, this is not included in the current work.

2.2. Technology advance evaluation

This concept provides a competitive process for biofuel production from various biomass that can significantly reduce biofuel production cost compared to the current technologies. For the technical route A (see Fig. 1), the integration of carbon membrane technology for biogas upgrading to produce high

purity biomethane (> 98%) for autothermal reforming can potentially reduce the energy consumption for gas purification. The flexible, high performance cellulose-based hollow fiber carbon membranes with CO_2/CH_4 selectivity >100 [6,17] will clearly be beyond the current state-of-the-art in area of biogas upgrading with respect to the energy efficiency, the selectivity of CO_2 over CH_4 , and the cost.

For the technical route B of CLG, the fuel reactor (FR) is operated at high concentrations of H_2 and CO , which decreases the gasification rate, being necessary the use of longer residence time and higher solids inventory. Oxygen carrier used in biomass CLG process will transfer oxygen from air to fuel reactor and convert biomass into syngas, which can avoid the use of traditional gasification agents such as oxygen or enriched-air as it is usually high energy-intensive for purification of O_2 from air. In addition, the required heat for endothermic reactions in the fuel reactor is provided by the hot solids coming from the air reactor. Therefore, no additional carbon combustion is needed for providing heat to the process. Moreover, the high resistance to sintering and agglomeration of OC will be used to avoid the risk of agglomeration in the fuel reactor. It should be noted that solid materials with high selectivity for H_2 production are preferred to avoid the stand-alone water-gas shift (WGS) reactor. Thus, a multifunctional oxygen carrier capable of promoting solid fuel conversion and syngas reforming at a low cost for solid biomass CLG technology is preferred.

The produced syngas will be cleaned and upgraded by thermocatalytic processes to achieve a proper H_2/CO ratio and very low contents of impurities such as nitrogen, sulfur, halogens, alkali metals, tar and particles (soot, ash). As syngas requirements on these impurities depend on the catalyst used, it is crucial to develop novel catalysts that are more resistant to such contaminants. Moreover, the use of multi-function

catalysts, for instance suitable for tar, NH_3 and H_2S abatement, could also reduce the number of operations usually needed for hot cleaning processes to ensure the production of syngas with the suitable composition for chemical synthesis [18,19]. The clean syngas will be mixed in the syngas platform to provide a flexible bioalcohol production process (adjustable H_2/CO ratio), and further converted into bio-alcohols by a thermo-catalytic process. High performance catalysts are required to achieve high CO conversion rate (55%) to bio-alcohols at lower temperature and pressure conditions, which can beyond the state-of-the-art technology with alcohols selectivity of 46.7% and CO conversion rate of 30–52% reported so far that are usually operated at high temperature of 300–350 °C and high pressure of 3–8 MPa [18,19]. The BTL efficiency, biofuel production cost and CO_2 emissions were evaluated to document the technology advance.

The concept introduces an integrated approach to enhance the total carbon conversion efficiency in processing digestates through anaerobic digestion and solid waste through gasification. There is significant scope to upgrade intermediate streams, to deploy process and process integration innovations, also to improve heat recovery from lean biogas burners so that to meet process heat requirements. Model-assisted work will be possible to extrapolate into a rather wide range of feedstocks whereas synthesis technology will make possible to adjust designs and solutions to different locations and regions. Exergy and energy integration set targets explaining the room to improve the solutions; coupled with mathematical optimization and supply chain analysis, which are the tools to analyze different scenarios and business models. The process integration modelling and development of control systems will improve the BTL efficiency and allow further commercialisation into multiple markets for liquid biofuels. Based on the designed concept, the BTL efficiency, biofuel production cost and CO_2 emissions were evaluated based on modelling to document the technology advance, and the overall BTL efficiency of the process is expected to beyond the state of the art technology of 36% estimated by Dietrich et al. [20].

3. Results and discussion

3.1. BTL efficiency

The proposed concept provides a unique platform to process different type of biomass in different channels to maximize the BTL efficiency, and the efficiency of each element is estimated as shown in Fig. 2. The biomass input of 150 kWh (lower heating value, LHV) was assumed in these two processes, and a 60% digestion efficiency of biomass to biogas was reported by using an optimized codigestion process [21], and a biomethane recovery of > 95% in the upgrading process using a two-stage carbon membrane technology can be achieved [17]. Moreover, the commercial autothermal reformer can reach a thermal efficiency of > 85% [22]. A gasification efficiency of 70% is expected by using multifunctional oxygen carriers and the optimized operating condition (current oxygen gasification has efficiency of < 65%). In addition, the efficiency of syngas conversion to mixed bio-alcohols of 60% is expected if advanced catalysts can be developed in the future. Thus, the overall BTL efficiency is calculated based on the method reported by Dietrich et al. [20], and estimated as,

$$\eta_{BTL} = 131 \text{ kWh} / (150 \text{ kWh} + 150 \text{ kWh}) \times 100 = 44\% \quad (1)$$

which is higher than the current BTL efficiency of 36% reported by Dietrich et al. [20] It should be noted that the estimation uncertainty is mainly determined by the efficiencies of the gasification and syngas conversion processes since all the other elements have already been demonstrated at higher technology readiness level (TRL) of > 5.

3.2. Energy efficiency of separation process

Separation process usually accounts more than 60% of the total energy consumption in most chemical production processes, this will be similar in a biological process. Thus, seeking an advanced separation technology is crucial to design an energy-efficient process, and the same applies to a BTL

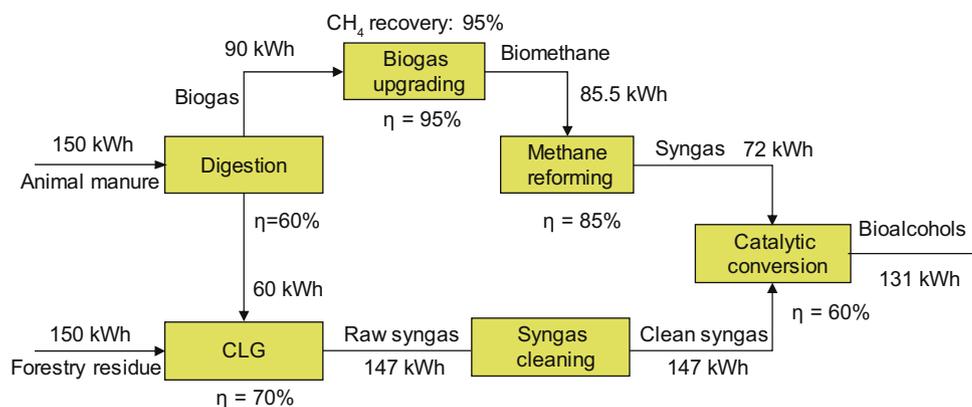


Fig. 2. Illustration of LHV flow from biomass to biofuels.

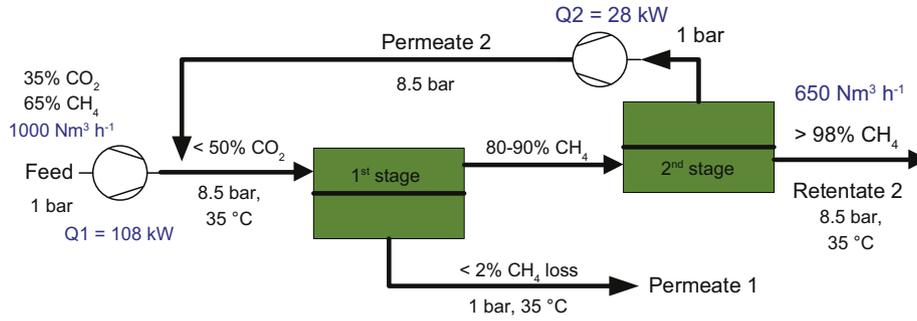


Fig. 3. Illustration of energy efficiency of membrane technology.

process. In this concept, membrane separation technology was proposed for both gas and liquid separations to reduce the total energy consumption.

For a $1000 \text{ Nm}^3 \text{ h}^{-1}$ feed biogas, a two-stage carbon membrane system (illustrated in Fig. 3 [6,17]) can produce high purity biomethane of 98% (with the membrane selectivity of CO_2/CH_4 of 100) at a low methane loss ($< 2\%$) with a specific energy consumption of 753 kJ Nm^{-3} upgraded biogas based on HYSYS simulation, which is much lower compared to a state-of-the-art amine absorption process of 1807 kJ m^{-3} upgraded biogas [23]. Thus, energy efficiency is improved more than 58% by using membrane technology in a biogas upgrading process (see the illustration in Fig. 3). However, it should be noted that the impurities (e.g., H_2O , H_2S , etc.) in raw biogas may have significant influences on membrane separation performance, which should be considered to identify suitable membrane materials in the future work.

Moreover, traditional distillation technology for alcohol/ H_2O separation is an energy-intensive process which makes the BTL technology uncompetitive as energy demands of this step accounts significant consumption of the whole process. Thus, bringing down the energy consumption in the bioalcohol purification step could significantly reduce the total energy consumption of a BTL process. Membrane pervaporation (PV) was considered as an energy-efficient process for alcohol/ H_2O separation compared to distillation [24]. At least 42% energy-saving can be achieved by using a hybrid membrane PV

process compared to the standard distillation producing azeotropic ethanol [25]. The energy consumption is expected to be further reduced by increasing the H_2O /alcohols selectivity of membrane materials. Overall, this concept provides a greener and more energy-efficient BTL process by integration of membrane separation technology.

3.3. Biofuel production cost

This concept converges two promising routes (CLG, AD) of waste streams (liquid, solid biomass) into next generation biofuels (biobutanol enriched mixed bio-alcohols) that can be integrated well and flexibly with distributed resources and available infrastructures - essentially integrating fragmented technologies using advances in science (catalysis, separation) and engineering (process integration). The bio-alcohol production cost is estimated as follows (see Fig. 4).

- 1) Liquid-biomass cost was estimated to be 0.014 € kWh^{-1} , solid-biomass 0.017 € kWh^{-1} .
- 2) After digestion and gas purification, the clean biomethane production cost of 0.25 € m^{-3} was estimated for a large-scale reactor (Biogas for road vehicles: Technology brief - IRENA), which is equal to 0.025 € kWh^{-1} (methane LHV 35.8 MJ m^{-3}). Considering a methane reforming efficiency of 85%, the syngas production cost was estimated to be 0.029 € kWh^{-1} .

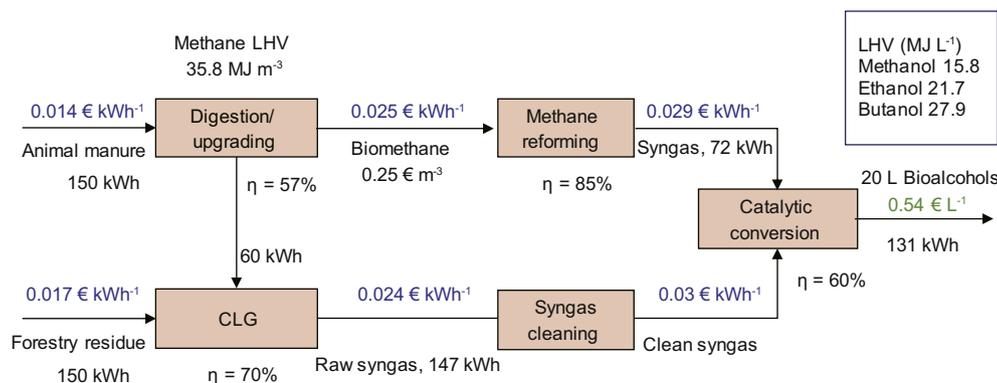


Fig. 4. Flow diagram on the estimation of bioalcohol production cost.

- 3) Based on the efficiency of CLG, the raw syngas production cost was estimated to be 0.024 € kWh^{-1} . By including syngas cleaning and conditioning cost, the production cost of clean syngas was estimated to be 0.03 € kWh^{-1} .
- 4) The total produced mixed bio-alcohols (assuming 30% C1, 10% C2, 60% C4) was estimated by,

$$\text{Productivity} = 131 \times 3.6 \text{ MJ} \\ ((15.8 \times 0.3 + 21.7 \times 0.1 + 27.9 \times 0.6) \text{ MJ L}^{-1}) = 20 \text{ L} \quad (2)$$

- 5) By assuming the syngas conversion efficiency of 60%, the specific bioalcohol production cost (€ L^{-1}) was calculated to be,

$$\text{Specific bioalcohol production cost} = (0.029 \\ \text{€ kWh}^{-1} \times 72 \text{ kWh} + 0.03 \text{ € kWh}^{-1} \times 147 \text{ kWh}) / 0.6 / 20 \text{ L} \\ = 0.54 \text{ € L}^{-1} \quad (3)$$

Compared to the current bioethanol production cost from wheat and sugar of 0.657 € L^{-1} (Road Transport: The Cost of Renewable Solutions), the overall cost reduction is 18.6%. The estimated biofuel production cost is very promising based on this concept. However, some barriers still need to be overcome, especially the efficiency of chemical looping gasification and syngas conversion efficiency. Thus, future development on cheap, advanced OC and catalysts are required to reach the estimated biofuel production cost.

3.4. Reduction of CO₂ emissions

Two advanced in-situ CO₂ capture technologies (i.e., CO₂ removal from biogas, and chemical looping CO₂ capture) are involved in this process, which can significantly reduce CO₂ emissions in a biofuel production process. The captured CO₂ from biogas (purity > 95%) can be utilized as raw materials in other downstream chemical processes or injected back to gas/oil wells for enhanced gas/oil recovery. Moreover, the end use of bio-alcohols has less CO₂ emissions compared to the petroleum-derived fuels, which can reduce CO₂ emissions in the road transport sector. The energy required for transport sector (including heavy road transport, shipping and aviation) in EU is estimated 18.37 million TJ by 2030 (considering a 10% increase from 2015 (16.7 million TJ))-this is equivalent to 5.74×10^{11} L gasoline. According to the EU 2030 vision of 15% biofuels will be blended to petroleum derived fuels for road transport (considering the specific CO₂ emission of 2.39 kg L^{-1} gasoline), the total CO₂ emission reduction is expected to be ($5.74 \times 10^{11} \times 15\% \times 2.39 \times 75\% \text{ kg} = 154 \text{ Mt}$) considering a targeted 75% savings on CO₂ footprints [26] for each liter of renewable fuel produced. This will significantly contribute to achieve the EU ambitious climate target for 2020 of a 20% reduction of CO₂ emissions (ca. 1265 Mt CO₂) compared with 1990's levels.

3.5. Sustainability

The production of first-generation biofuels from food industry, the second-generation bioethanol from lignocellulosic materials, and the biofuels from algae are today partly occupying land area, fertile soil and resources that otherwise could be used for food production, representing major environmental and societal drawbacks. The next-generation biofuels produced from this concept will be independent of land, water and chemical nutrients-fertilizers, and use sustainable forestry residues, waste biomass and effluents as feedstocks, which are abundant and versatile renewable resources on the earth that can enlarge the biofuel potential. Thus, this concept can overcome the drawbacks of other technologies and will not displace resources and areas suitable for food production.

Beside the main contributions on CO₂ emissions reduction, increasing share of renewable energy and improving energy efficiency of technology, this concept will additionally contribute to mitigation of soil/water pollution. Comparing to the conventional use of the digestates as fertilizer, the proposed concept provides a cost-effective (compared to the current bioethanol production process) and low environmental impact solution to utilize dewatered digestates as feedstock for chemical looping gasifier, which can mitigate the soil pollution of heavy metals in relation to the present legislation. Moreover, the wastewater produced in the digestion reactor can also be treated and reused in the process to reach the objective of maximisation of water reuse.

4. Conclusions

A novel BTL concept by strategically combining anaerobic digestion (biological process) with chemical looping gasification (thermochemical process) integrated with membrane separation technology was proposed to improve the carbon conversion efficiency from different type of feedstocks (liquid- and solid-biomass) to biofuels. The targeted bio-syngas can be catalytically converted to mixed bio-alcohols (biomethanol, bioethanol and mainly biobutanol) with better end-user compatibility by directly blending into petroleum-derived fuels at various contents. This concept bridges the gap between the status and the need described in Table 1, and thus provide a cost-effective technology for next-generation biofuel production from non-food biomass. The overall BTL efficiency was theoretically estimated to be 44%, which is higher compared to the current BTL technologies. Moreover, an overall cost reduction of 18.6% can be achieved if the expected efficiency of chemical looping gasification and syngas conversion can be reached. A significant reduction on CO₂ emissions is also expected based on the implementation of this technology to develop next-generation biofuels for road transport, which will significantly contribute on the sustainability of the development of renewable energy. However, the

major barriers related to the efficiencies of chemical looping gasification and syngas conversion should be overcome by developing cheap, advanced OC and catalysts to successfully implement the proposed concept.

Conflict of 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.

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