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Syngas biomethanation: Current state and future perspectives

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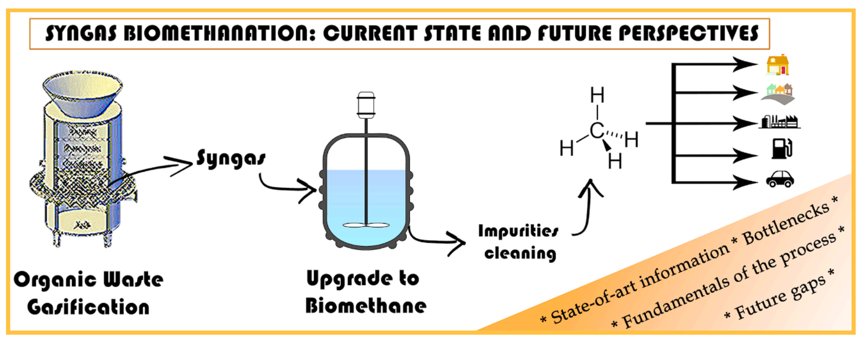
HIGHLIGHTS

- Biomethane from syngas is an alternative to natural gas in a bio-circular economy.
- Organic waste gasification from several sources can produce quality syngas.
- Biotrickling filters can replace catalytic reactors for bioCH_4 synthesis from syngas.
- Syngas bioconversion to biomethane needs optimization of CO and H_2 mass transfer.
- Methanogenic microbiology must be boosted to optimize syngas bioconversion.

GRAPHICAL ABSTRACT



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ABSTRACT

In regions highly dependent on fossil fuels imports, biomethane represents a promising biofuel for the transition to a bio-based circular economy. While biomethane is typically produced via anaerobic digestion and upgrading, biomethanation of the synthesis gas (syngas) derived from the gasification of recalcitrant solid waste has emerged as a promising alternative. This work presents a comprehensive and in-depth analysis of the state-of-the-art and most recent advances in the field, compiling the potential of this technology along with the bottlenecks requiring further research. The key design and operational parameters governing syngas production and biomethanation (e.g. organic feedstock, gasifier design, microbiology, bioreactor configuration, etc.) are critically analysed.

1. Introduction

Anthropogenic activities have significantly changed the dynamics of the planet and caused many environmental problems in recent decades (Seo et al., 2022). The human population is expected to grow from 6.8 billion to >9 billion by 2050, while energy demand is expected to nearly double. Furthermore, the production of organic waste will continue to rise, posing a global problem. The large volume of organic waste, if not properly handled, may degrade air, water, and soil quality, causing detrimental consequences for the environment (Lin et al., 2018). An attractive alternative to the use of these organic wastes is their energetic valorization. This fact, together with the decreasing production costs

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and enforcement of greener environmental energy regulations, may support a steady increase in renewable energy consumption (e.g. 3% in 2020) along with a slowdown in the demand of fossil fuels (Duarah et al., 2022). Solar, biomass, wind, geothermal and hydropower are the main renewable energy sources in the energy pool of most countries (Singh et al., 2022). Bioenergy currently makes up about 10% of the global primary energy supply, and holds the potential to offer >60% of the world's energy supply. The production of bioenergy is expected to triple by 2060 (Scarlat and Dallemand, 2019), (Paniagua et al., 2019).

The current geopolitical scenario and international environmental agreements require in the European Union novel energy production systems non-based on fossil fuels, capable of simultaneously reducing greenhouse gas emissions and environmental impacts and aligned with the promotion of circular economy (Correa et al., 2019). In this context, renewable energy technologies based on biomass utilization can play a key role. The aforementioned organic wastes are one of the most common types of biomass fuels receiving special attention as a potential source of renewable energy (Safarian and Unnthorsson, 2018). Recent estimations determine a world annual production of 2.01 billion tonnes of these residues, the third part still being mismanaged and seriously threatening the environment (Szulc et al., 2021). Within the existing biomass-to-energy conversion technologies, thermochemical processes exhibit a high untapped potential (Ayub et al., 2022). These technologies include direct combustion, liquefaction, pyrolysis and gasification processes. In comparison to commercially available technologies like incineration, gasification is a novel but promising technology. Waste gasification can be used as a more reliable energy supply technology for places that are remote from central energy networks and require a district heating and power system. In addition, gasification shows considerably lower environmental impacts due to the reduced water requirements and lower emissions of gaseous pollutants into the atmosphere (Safarian et al., 2020).

The gas stream resulting from waste gasification process, typically referred to as syngas, constitutes an energy vector that can be upgraded, stored and distributed (exported/imported) globally using the already existing infrastructures (Mărculescu et al., 2022). This syngas can be upgraded into biomethane, which exhibits comparable applications than natural gas in power and heat generation, transportation, and chemical sector. Moreover, biomethane holds a critical advantage over liquid biofuels since it is totally miscible with natural gas (Grimalt-Alemany et al., 2018). While natural gas consumption will remain constant for at least a few decades, biomethane is expected to cover the new gas demand due to their renewable nature and low CO_2 footprint (Skorek-Osikowska, 2022). Thus, the biomethanation process has been extensively studied in the past years as a promising energy alternative, although several limitations associated with the different variables involved might still be overcome (Figueras et al., 2021).

Thus, this review compiles and critically discusses most recent data published on the topic of syngas biomethanation, not only from a microscopical perspective but also paying especial attention to the gas biofiltration process. To this aim, the review includes first a detailed description of the conversion of organic waste into syngas and the impact of different gasification variables on the syngas composition. The syngas biomethanation process is then tackled from both a microscopical and macroscopical perspectives, with special emphasis on bioreactors configuration and process limitations. Finally, future prospects for this sector are discussed.

2. Organic waste conversion into syngas via gasification

Gasification provides an efficient and robust route to thermochemically converting a broad portfolio of wastes into an energy vector through an indirect combustion (Di Giuliano et al., 2022). Thus, the gasified organic residue is converted into a valuable synthesis gas (called syngas) via partial oxidation at high temperatures. This partial oxidation can be undertaken with air, oxygen or steam (Saleem et al., 2020). In the co-production of bio-fertilizer (bio-char and ash) and syngas, organic waste gasification is a single-step thermo-chemical process widely accepted as energy-efficient and cost-competitive (Ansari et al., 2020). Drying, pyrolysis, partial oxidation, and gasification are all examples of complex thermochemical reactions that result in the simultaneous interconversion of solid and gaseous species. This partial oxidation generates the heat that powers the other reactions while also lowering the organic waste feedstock's initial moisture content. The heat from the oxidation zone and the limited oxidising agent also cause waste pyrolysis at 200 to 700 °C (O_2 or air), with the concomitant formation of a gas stream consisting of a mixture of hydrocarbons, N_2, H_2, CO, CO_2, H_2O and other minor compounds (Chen et al., 2019). Char, which further takes part in gasification, is predominantly formed during the pyrolysis process (Narnaware and Panwar, 2022). The gasification process, on the other hand, occurs allothermally at a specific temperature and pressure, which are referred to as gasification temperature and gasification pressure.

The thermal gasification process can be carried out using different gasifying agents. This parameter greatly influences the composition of the final syngas. The global gasification reactions for 1 mol of organic waste using air, steam and oxygen can be described by **Eqs. (1), (2) and (3),** respectively (Khalilarya et al., 2021):

$$CH_\alpha O_\beta + \omega H_2O + \gamma(O_2 + 3.76N_2) \rightarrow n_{H_2}H_2 + n_{CO}CO + n_{CO_2}CO_2 + n_{H_2O}H_2O + n_{CH_4}CH_4 + n_{N_2}N_2 \quad (1)$$

$$CH_\alpha O_\beta + (\omega + \varepsilon)H_2O \rightarrow n_{H_2}H_2 + n_{CO}CO + n_{CO_2}CO_2 + n_{H_2O}H_2O + n_{CH_4}CH_4 \quad (2)$$

$$CH_\alpha O_\beta + \omega H_2O + \delta O_2 \rightarrow n_{H_2}H_2 + n_{CO}CO + n_{CO_2}CO_2 + n_{H_2O}H_2O + n_{CH_4}CH_4 \quad (3)$$

Where $CH_\alpha O_\beta$ is the organic waste chemical formula. α and β, are, respectively, the hydrogen and oxygen molar ratios. n_{H_2}, n_{CO}, n_{CO_2}, n_{H_2O}, n_{CH_4} and n_{N_2} is the molar number of hydrogen (H_2), carbon monoxide (CO), carbon dioxide (CO_2), steam (H_2O), methane (CH_4) and nitrogen (N_2), respectively. γ, ε, and δ are the input air, steam and oxygen, respectively, referred to 1 mol of dry ash-free organic waste. Finally, w is the moisture/dry ash-free organic waste ratio described according to Eq. (4):

$$\omega = \frac{M_{waste} \cdot MC}{M_{H_2O} \cdot (1 - MC)} \quad (4)$$

MC is the moisture content of the organic waste, and M_{waste} and M_{H_2O} stand for the molecular weight of the organic waste and water, respectively.

Gasification is one of the classical methods of H_2 production via thermal decomposition of coal and biomass, and it is considered more energy efficient than combustion processes to produce energy. An overall explanation of the different types of gasifiers employed with their operating conditions is shown in Fig. 1.

In the context of the circular economy needed to guarantee the sustainability of anthropogenic activities, gasification can help reducing the volume of organic wastes while generating energy in the form of syngas. Promising studies have recently shown that heterogeneous wastes can be converted to syngas via gasification (Ayub et al., 2022; Di Giuliano et al., 2022; Lee, 2022). However, syngas has a relatively low calorific value (in particular when produced via air gasification) and a high content of tar, which hinders its direct chemical or biotechnological conversion.

3. Syngas composition

The syngas composition varies according to both the composition of the organic waste and the experimental conditions of the gasification process. Operational parameters such as the gasification time, the type

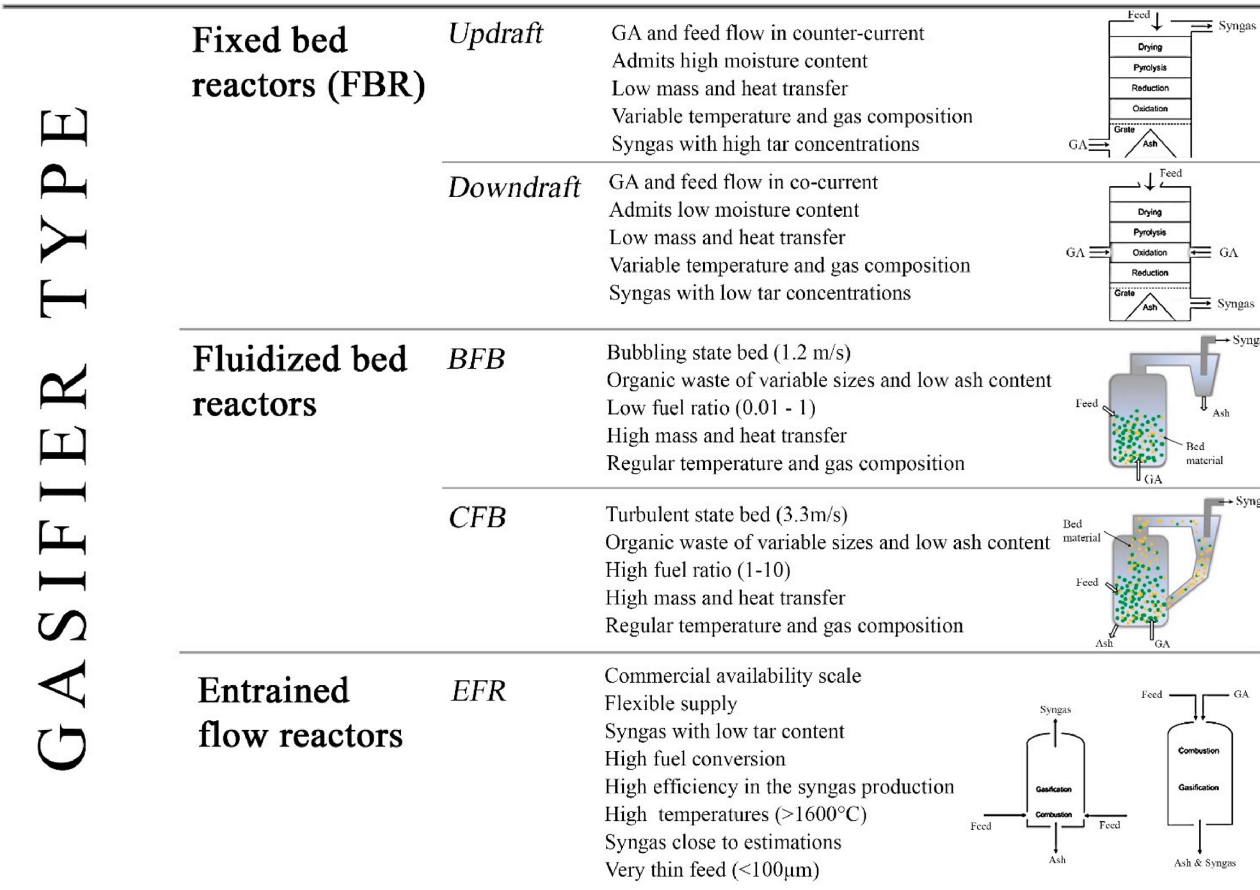

Fig. 1. Main characteristics and schemcatic representation of the different types of gasifiers. Adapted from (Basu, 2018; Bermudez and Fidalgo, 2016; Pio and Tarelho, 2021). Gasifying agent (GA), Fuel ratio = Fixed carbon/ Volatile matter, Bubbling fluidized bed (BFB), Circulating fluidized bed (CFB), Entrained flow reactors (EFR).

and flow rate of gasifying agent (GA), the temperature, or the moisture content of the feedstock influence the syngas composition (Aryal et al., 2021). Similarly, syngas composition can vary depending on the size, shape and density of the organic waste (Cerone et al., 2020). The most important parameters influencing syngas composition are described below:

3.1. Waste composition

The composition of the organic waste gasified clearly influences syngas composition. Traditionally, coal was used as a feedstock for syngas production, which entailed multiple technical and environmental issues such as particle agglomeration, fused-ash slagging and emission of SO_x, NO_x, and H_2S (Gupta and De, 2022). Hence, research has been devoted to explore the potential of alternative feedstock such as biomass, municipal waste, biosolids (stabilized residues derived from the treatment of biological sewage sludge) or plastic waste. The syngas composition of the most common organic wastes is summarized in Table 1**.** The composition of syngas from lignocellulosic biomass differs depending on the GA employed, especially in terms of H_2 content. For instance, gasification of pine wood with steam supports a H_2 content of 60.3% (Kartal and Özveren, 2021), while gasification of wood chips with air cannot provide H_2 contents higher than 20% using BFB reactors (Bandara et al., 2021). (Rasmussen and Aryal, 2020)

Table 1
Syngas composition for several feedstocks depending on the gasification agent employed.

Raw material	H_2 (%)	CH_4 (%)	CO (%)	CO_2 (%)	N_2 (%)	Reactor type	Reference
Gasification agent: Steam							
Pine Wood	60.3	1.6	15.3	22.4	–	BFB	(Kartal and Özveren, 2021)
Corn straw	26–29	13–16	33–35	22–25	–	FBR	(Hu et al., 2019)
Sewage sludge	58–63	1–3	13–18	14–17	–	FB	(Hu et al., 2020)
Municipal solid waste	42–45	14–17	15–18	23–26	–	FBR	(Fu et al., 2022)
Gasification agent: Air							
Almond shells	14.3	2.3	30.8	8.4	43.7	FBR	(Cerone et al., 2020)
Plastic waste	18–22	0–4	12–18	8–12	60–63	FBR	(Mojaver et al., 2022)
Food waste	1–5	1–6	3–8	12–18	57–68	FBR	(Mărculescu et al., 2022)
Wood chips	10–20	1–5	13–18	12–16	36–60	BFB	(Bandara et al., 2021)
Wood Pellet	16–20	2–4	12–16	12–16	42–50	BFB	(Bandara et al., 2021

compared the syngas obtained for straw and wood pellet with similar results in terms of CO, H_2 and CH_4. However, the gasification of straw resulted in operating problems due to the agglomeration derived from the higher alkali content. The gasification of plastic materials produces combustible gasses like H_2, CH_4, C_2H_6 and C_3H_8 (Shadangi, 2022). A reduction of H_2 production by a factor of ten was however observed when plastic was combined with biomass at 800 °C (Mărculescu et al., 2022). (Al-asadi et al., 2020) also demonstrated that the addition of Me/Ni/ZSM-5 catalysts or the use of more oxygen in the N_2/O_2 mixture can improve syngas production (H_2 and CO). Municipal solid waste (MSW) has been successfully gasified under several operating conditions with acceptable results in terms of high contents of CO, H_2 and CH_4 in the syngas (Khalilarya et al., 2021) as well as higher LHV (lower heating value) (16 MJ/Nm^3) (Veses et al., 2020). As a result, MSW gasification is a viable and cost-effective option for the final disposal of these wastes (Lee, 2022). Sewage sludge, both wet and dried, has been already studied for bio-syngas production. (Yang et al., 2021) proposed a two-stage sorption-enhanced steam gasification of sewage sludge for syngas production with a H_2 production 3 times higher when compared with no steam addition and also a higher purity of H_2 and CO gases. Other studies have mixed sewage sludge with pine sawdust, obtaining a maximum dry gas yield (1.23 Nm^3/kg), H_2 yield (14.44 mol/kg) and a carbon conversion efficiency (84.56%) using 60% of sewage sludge (Hu et al., 2016). If the gasification process is focused on the production of methane, the gasification of agricultural waste supports a CH_4 content of 45–75% (Gao et al., 2018), that of urban sewage sludge, 60–65% CH_4, and that of landfill wastes, 35–65% CH_4 (Guerrero et al., 2020).

3.2. *Temperature*

Temperature also influences syngas quality (Fuchs et al., 2020). According to the literature, as the temperature of gasification rises, the concentration of the resulting H_2 and carbon conversion efficiency rise, while the concentration of tar in the syngas falls (Müller et al., 2017). Recent investigations have demonstrated that temperature is a key parameter when PET (polyethylene terephthalate) was gasified. Thus, the yields of H_2 (+87.7%), the dominant gas product CO_2 (+40.3%), and biphenyl (+123%) all improved when the temperature was raised from 750 to 800 °C. The gasification products of MSW were also a function of temperature (Kardani et al., 2021). As a result, raising the gasification temperature increased syngas production and H_2 content (Lee, 2022). In addition, (Wu et al., 2019) reported that the optimal temperature for H_2 production from lignocellulosic biomass was 850 °C, which minimized the activation energy of H_2 formation. Recent studies have attempted to model the influence of temperature on the composition of the final syngas. Thus, (Mikulandrić et al., 2020) accurately modelled the composition of the syngas with a 90% match with short time variations (up to 5 min).

3.3. *Gasifying agent*

The gasifying agent is a key operational parameter that remarkably influences syngas composition. Table 1 shows how the presence of N_2 in the syngas is linked to the use of air and steam as GA for several feedstocks. When lignocellulosic biomass is gasified, the GA and the *equivalence ratio* (ER) play a critical role on syngas composition. The ER is defined as the ratio of the actual air-to-fuel ratio and the stoichiometric air-to-fuel ratio. Thus, ER accounts for the net effect of airflow rate, feed supply rate and the residence time (Martínez et al., 2011). When using air as gasifying agent, the composition and LHV of the syngas linearly changes with ER. Thus, the LHV increases with the decrease in ER. Interestingly, when using steam as gasifying agent, the LHV remains nearly constant when varying the steam to fuel ratio (Karatas and Akgun, 2018). The yield in both fluidized bed and moving bed reactors is directly proportional to the ER variation and the gases residence time in the reduction zone, according to a series of studies (Sheth and Babu, 2009). On the other hand, the syngas yield for dried sewage sludge is typically greater when using air as GA compared to steam/O_2 during the gasification. This is attributed to both the high nitrogen content in the syngas and the high char gasification rate mediated by air (Jeong et al., 2022). Despite the low heating value of syngas produced by air gasification processes, it has the lowest production cost (Pio et al., 2018), which is key to the commercialization of MSW gasification. For PET gasification, the increase in the steam to fuel ratio does not support a significant increase in H_2 yield (Li et al., 2022). However, the use of steam instead of air increased H_2 levels by a factor of 3 during sewage sludge gasification (Nipattummakul et al., 2010).

3.4. *Moisture*

Moisture content of the feedstock also influences the composition of the syngas. The gasification process can employ fuel with a moisture level ≥40%, although moisture levels >30% hinder ignition and reduce the syngas heating value (McKendry, 2002). Overall, the decrease in the moisture of the feedstock biomass implies a positive effect on biomass gasification (Jahromi et al., 2021) and enhances the cold gas efficiency, CGE (chemical energy in the product gas versus the energy in the initial solid fuel) (Niu et al., 2013). However, the supercritical water gasification process and the gas shift reaction in the gasifier benefit from increased moisture content. Syngas composition is also affected by this parameter. In this sense, based on accepted models (Kirsanovs and Zandeckis, 2015) working with lignocellulosic biomass, it can be stated that the increase in the moisture content can exert a positive impact on syngas CH_4 content. Thus, the amount of CH_4 obtained in this type of fuel increased from 1.72% to 40% when increasing the moisture content from 0% to 40%. However, a higher fuel moisture content mediated a detrimental impact on CO content during the gasification of the same waste. Indeed, the CO concentration dropped significantly from 30.5% to 6.20% when the moisture content increased from 0 to 40% likely due to the drop in temperature in the gasifier reactor. On the other hand, CO_2 content in the syngas raised from 5.63% to 19.23% with increasing moisture levels.

Waste biomass moisture content also influences de calorific values of syngas obtained from gasification. The higher the moisture content, the lower the energy for the syngas. For example, a 7% reduction in the moisture content (from 29 to 27%) of an herbaceous biomass caused a nearly double calorific value of syngas (2.63 MJ/Nm^3 vs 4.95 MJ/Nm^3) (Atnaw et al., 2014).

3.5. *Gas partial pressure*

This parameter affects the gas composition depending on the waste gasified. Hence, H_2 production from the gasification of plastic materials decreased when increasing the operational pressure in the gasifier. This decrease was more prominent than for polypropylene (~5% decrease in H_2 production with an increase in the pressure of 900 kPa). Similarly, CO content decreased by ~3% during polypropylene gasification at a similar pressure increase. However, the effects of operational pressure in the gasifier on CO_2 production were negligible (Mojaver et al., 2021). Increases in CO partial pressure (P_{CO}) in syngas biomethanation processes typically result in partial inhibition, which affects CH_4 yield and productivity. Although changes in P_{H2} have been reported to affect microbial activity (as higher P_{H2} reduces microbial diversity), the concentration of H_2 exerts a milder effect on the consortium's performance (Grimalt-Alemany et al., 2018). Particularly, for coal gasification using steam as GA, the composition of the gas changed as the steam partial pressure increased. With lower steam partial pressures, H_2 and CO_2 content decreased, while CO content increased. Indeed, changing the partial pressure of the steam can control the H_2/CO ratio of the synthesis gas (Sharma et al., 2009). (Hantoko et al., 2019) studied the composition of the synthesis gas obtained from the gasification of sewage sludge and reported that, despite the constant composition of CO and CO_2, H_2

content decreased slightly and CH_4 increased (in both cases with variations of less than 5%) when the pressure was increased by 10 points (from 25 to 35 MPa).

3.6. Syngas feed impurities

Apart from CO, CO_2, H_2, H_2O, and CH_4, raw syngas commonly contains solid particles (mostly ash), condensable volatiles, and gases produced after gasification, such as acetylene (C_2H_2), ethylene (C_2H_4), ethane (C_2H_6), benzene (C_6H_6), hydrogen sulfide (H_2S), sulfur dioxide (SO_2), ammonia (NH_3), nitrogen (N_2), hydrogen cyanide (HCN) or carbonyl sulfide (COS) among others. The type and concentration of syngas impurities, and their impact on microbial processes, can be influenced by a number of factors, including gasifier design and performance and/or gas clean-up methods. Impurities can cause cell toxicity or enzyme inhibition, varying redox potential, osmolality, and pH (Xu et al., 2011). Prior to the syngas biomethanation, syngas pollutants must be eliminated to avoid detrimental effects on bioconversion performance. For instance, several enzymes in acetogenic bacteria are inhibited by tars, NOx and NH_3. Despite the fact that low levels of impurities have been shown to have no effect on biomethanation performance, more research is needed to determine raw syngas minimum clean-up requirements. In this context, most existing investigation at laboratory scale use synthetic syngas commonly composed of carbon monoxide (CO), carbon dioxide (CO_2), and hydrogen (H_2), the composition of this "clean" syngas clearly differing from industrially produced syngas.

To improve the economic viability of waste-to-biomethane via gasification-biomethanation, the cost of syngas cleaning must be reduced (Santos and Alencar, 2020). A typical syngas purification scheme consists of a quench tower followed by a washing step with water solution and then an alkaline solution. Particulates, metals, and HCl are all removed during this treatment. A second upgrading step is required to complete purification and sulphur removal following a wet electrostatic precipitator to remove fly ashes together with a homogenization tank to buffer fluctuations in syngas flow and composition (due to heterogeneity in the gasifier feed). The residual contaminants in the syngas are at ppm or ppt levels after this treatment sequence, but they are still able to deactivate conventional downstream chemical catalysts. Advanced gas cleaning technologies, such as those based on catalytic dust filters or analogous, could improve syngas cleaning and decrease costs (Centi and Perathoner, 2020).

4. Syngas biomethanation

A great variety of wastes with high organic carbon content can be gasified and the resulting syngas be upgraded (Demey et al., 2019). This process is limited by the low productivity, CO inhibition (Li et al., 2022) and the extreme operational conditions. Several catalytic systems, commonly based on nickel, have been reported in the literature (Stangeland et al., 2017). Among the main problems to be tackled, lowering the process temperature while boosting conversion yield, manufacturing reliable catalytic systems, and regulating reaction heat are pointed out as the most important challenges in this mature technology. In this sense, the engineering of the reactor and the optimization of the catalyst composition and formulation are crucial in order to control the above mentioned difficulties. As previously stated, Ni is by far the most commonly used catalyst due to, among others, its high activity, strong CH_4 selectivity and low cost (Al-Timimi and Yaakob, 2022). However, other works have tested Ru and Fe catalysts with good results (Kirchner et al., 2018). The usage of nanoscale catalysts may aid in improving their activity and stability. Dispersion of nanoparticles on different supports has been explored, with hydrotalcites and lanthanum oxide receiving current interest (Aresta et al., 2018).

On the other hand, the biological method converts syngas to methane through the metabolism of methanogenic microorganisms (biomethanation) at milder temperatures (35–75 °C) and atmospheric pressure. The biomethanation of syngas by microorganisms can take place in two ways. As a methane precursor, the first uses an acetate pathway. Microbial cells that perform this reaction include *Acetobacterium woodii* and *Enbacterium limosum*. Following that, methanogenic bacteria like *Methanosarcina barkeri* convert acetate to methane. The H_2/CO_2 pathway is used in the other pathway. Microorganisms such as *Methanothermobacter thermoautotrophicus* and *Clostridium thermoaceticum* can convert CO into CO_2. Some microorganisms, such as *Methanosarcina formicicum*, convert the H_2 and CO_2 produced and initially present in the syngas into methane. When compared to catalyst agents, the use of microorganism makes the process more resilient to impurities in the feed gas and is more environmentally friendly (Ba et al., 2020). In addition, biological syngas methanation can convert CO/CO_2 and H_2 into CH_4 using different biological routes harboured by bacteria and archaea (Fig. 2), which supports syngas biomethanation independently from the $CO/CO_2/H_2$ ratio (Figueras et al., 2021). Thus, the exploitation of the biological routes for syngas conversion into methane has recently attracted a great interest. In order to upgrade the resulting syngas molecules like CO and CO_2 into biomethane using H_2 as electron donor (typically present in syngas), multiple strategies can be implemented. CH_4 can be produced from non-converted CO_2 using traditional biogas scrubbing or by methanizing CO_2 with additional H_2 (Angelidaki et al., 2018). During gasification, a CO_2 sorbent can be used to reduce the concentration of CO_2, resulting in a syngas with a higher concentration of H_2 (Salaudeen et al., 2020). H_2 can also be produced using renewable electricity and water electrolysis (Aryal et al., 2021).

4.1. Microorganisms involved in syngas biomethanation

The two main carbon sources of syngas are CO and CO_2, which are used by methanogenic microorganisms to build-up new biomass and CH_4. The biological conversion of CO_2 to CH_4 with H_2 addition is a well-known process implemented for biogas upgrading (Kougias et al., 2017). CO conversion to CH_4 is, on the other hand, much less well-studied, and CO has been shown to be toxic to microorganisms (Wang et al., 2021). One of the routes for converting CO into methane consists of an initial conversion to acetate catalyzed by acetogenic bacteria. This CO metabolization can be carried out by some species from genera *Clostridium*, *Acetobacterium* and *Sporomusa*, which are capable of producing acetate and alcohols (Novak et al., 2021; Renaudie et al., 2022; Song et al., 2021). The reaction is followed by acetoclastic methonagenesis. The second route for syngas biomethanation involves metabolizing CO to H_2/CO_2 via carboxydotrophic hydrogenogenesis, also known as water shift reaction. Certain species of the genres *Rhodospirillum, Thermincola, Desulfotomaculum, Carboxydothermus, Caboxydocella* and *Moorella* convert CO to H_2/CO_2 (Kato et al., 2021; Liu et al., 2020). This reaction is followed by hydrogenothrophic methanogenesis. In methanogenic environments, acetate is either consumed by acetoclastic methanogens that directly used for methanogenesis or it is oxidized by syntrophic acetate-oxidizing bacteria. Syntrophic acetate oxidation (SAO) is dependent on the interspecies transfer of hydrogen and/or formate, where the syntrophic partner (e.g., a hydrogenotrophic methanogen) consumes the fermentation products (Dyksma et al., 2020; Sun et al., 2014). SAO has been identified as a significant anaerobic pathway when combined with hydrogenotrophic methanogenesis under thermophilic conditions (55 °C) (Dolfing, 2014).

Although pure microbial cultures have supported a good biomethanation performance, recent studies have revealed the key role of microbial consortia within the overall biomethanation process (Logroño et al., 2022; Szuhaj et al., 2021). Thus, mixed culture-based biomethanation has multiple advantages over monoculture fermentation in terms of resilience and sensitivity to inhibition. However, mixed culture fermentation often require a greater level of control and a thorough understanding of how microbial composition governs syngas biomethanation, and particularly CO conversion. Furthermore, H_2 has a

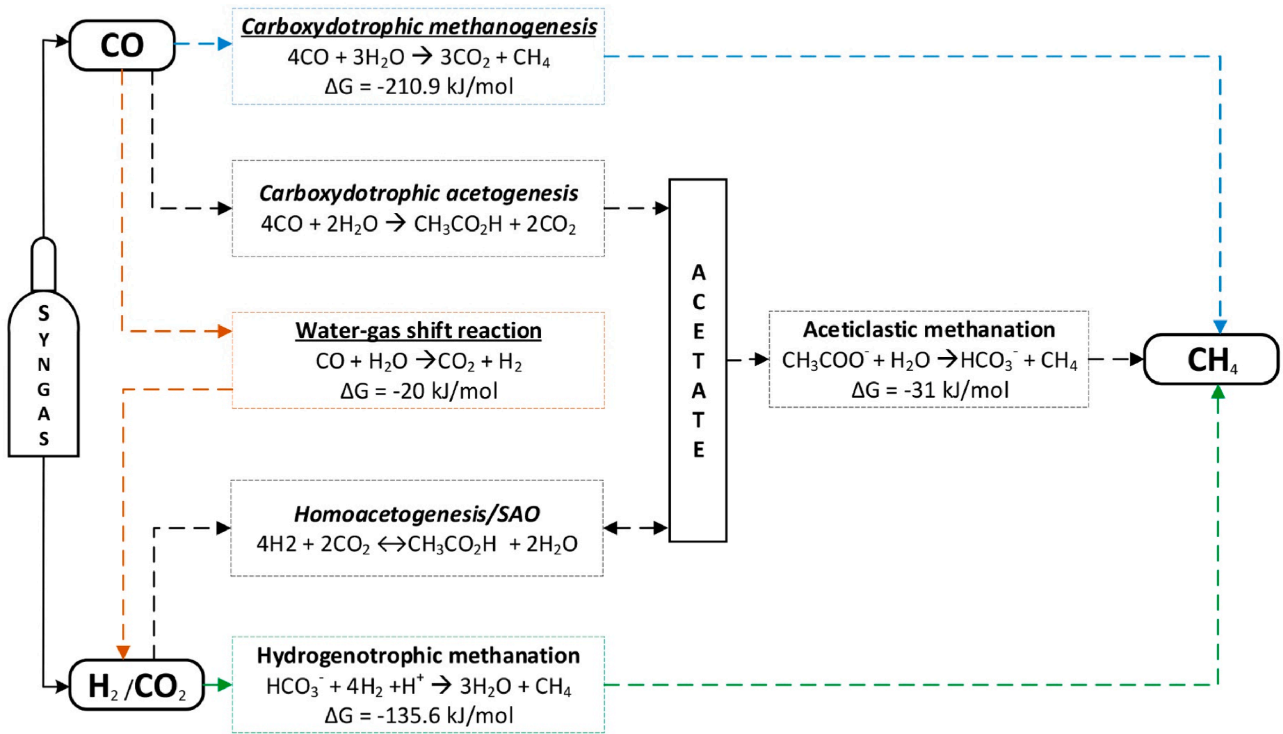

Fig. 2. Routes for syngas bioconversion into CH_4. Adapted from (Grimalt-Alemany et al., 2018; Rafrafi et al., 2021). SAO: syntrophic acetate oxidation.

significant influence on syngas biomethanation, since it ultimately determines the maximum CH_4 level in biomethane. Indeed, H_2 is typically required to completely convert CO and CO_2 present in syngas to virtually pure methane, a process known as syngas upgrading (Li et al., 2020). Similarly, the use of archaeal biofilms cultivated on membrane surfaces in a custom-made membrane biofilm reactor for hydrogenotrophic methanation has already been positively tested (Pratofiorito et al., 2021) reached a maximum methane production per reactor up to 1.17 $Nm^3/(m^3 \cdot d)$, exhibiting that the concept of membrane bound biofilms improves mass transfer by directly delivering substrate gases to the biofilm. When designing the biomethanation process, modelling the behaviour and performance of mixed cultures under conventional operational scenarios might be quite valuable. Process modelling recently revealed that differences in biomethane productivity were due to the prevailing catabolic routes, rather than to the kinetic parameters of the microbial consortium (Grimalt-Alemany et al., 2020a). As a result of this finding, the study of microbial kinetics, which was previously of critical importance, has been pushed to the background.

In this context, the most recent works in syngas biomethanation focused on the use of artificial hyperthermophilic archaeal co-cultures capable of transforming synthetic carbon monoxide (CO) from flue gases to methane. Synthetic co-cultures represent a novel approach for the synthesis of bio-based products where interspecies interactions occur without the complexity of open mixed cultures, thus minimizing side reactions and increasing product selectivity. When grown as co-cultures, microorganisms can act more efficiently than when cultivated independently. Indeed, an effective transfer of metabolites and macromolecules, such as proteins and RNA, occurs in co-cultures, causing the strains to potentially influence each other's metabolism directly (Diender et al., 2021). The co-culture of *Carboxydothermus hydrogenoformans* (a carboxydotrophic hydrogenogen) with *Methanothermobacter thermoautotrophicus* (a hydrogenotrophic methanogen) can effectively bioconvert syngas into biomethane. Thus, *C. hydrogenoformans* biotransforms the toxic CO into H_2 and CO_2 (waster-gas shift reaction) supporting the growth of *M. thermoautotrophicus*, which is capable of rapidly assimilating H_2 and CO_2 from the environment, creating thermodynamically more favourable conditions for *C. hydrogenoformans* growth. This process should be conducted under thermophilic conditions, where the Standard Gibbs free-energy change ($\Delta G_0'$) of the water–gas shift reaction becomes more negative. The superior performance of the symbiotic *C. hydrogenoformans* and *M. thermoautotrophicus* co-culture has been recently validated by the research group of Dr. Souza at Wageningen University & Research (The Netherlands) under suspended growth in stirred tank fermenters (Diender et al., 2018). Likewise, co-cultures of *Thermococcus onnuriqqqqneus* and *Methanocaldococcus jannaschii*, *Methanocaldococcus vulcanius*, or *Methanocaldococcus villosus* have been successfully tested by (Zipperle et al., 2021). In this study, up to 10 mol% CH_4 was produced by converting pure CO or synthetic CO-containing industrial waste gases employing the aforementioned co-culture in closed batch bioreactor.

5. Process limitations

Syngas bio-conversion to methane is governed by environmental, design and operational parameters. The most relevant limitations encountered during syngas biomethanation are described below.

pH. The syngas biomethanation route and process efficiency are also affected by this parameter (Li et al., 2022). pH can influence the activity of microorganisms involved during biomethanation. While archaea have a relatively narrow pH range for growth, ranging from 6.0 to 8.0, with optimal growth activity at 7.0, bacteria exhibit a much wider pH range (Garcia et al., 2000). The pH of the syngas biomethanation process would therefore depend on the biocatalysts involved, performing best at neutral pH (Li et al., 2022). The accumulation of volatile fatty acids (VFA) could lower the pH of the reactor, causing inhibition and ultimately process failure (Yuan et al., 2019). If H_2 is added to syngas, it will preferentially react with CO_2 rather than CO, resulting in higher H_2 consumption and thus an increase in the pH, which inhibits the activity of CO consuming bacteria (Li et al., 2020). When there is a high conversion rate of VFAs to biogas (methane and CO_2), as well as an additional CO_2 content in the syngas, special attention must be paid to maintaining a neutral pH (Westman et al., 2016).

Operational temperature influences both gas–liquid mass transfer and microbial kinetics during syngas bioconversion. Despite the relatively low gas solubility under thermophilic conditions, a previous study demonstrated that thermophilic conditions outperformed mesophilic conditions in syngas biomethanation (Alves et al., 2013). Furthermore, the operational temperature has an impact on CO metabolism. For instance, (Sipma et al., 2003) investigated CO metabolism at 30 and 50 °C in seven different anaerobic sludge inocula. At 30 °C and 55 °C, the results showed that acetate and H_2/CO_2 were the main precursors for methanogenesis, respectively. However, because of the higher acetate yield and syngas conversion rate, mesophilic conditions were found to be more suitable for the conversion of syngas to acetate than thermophilic and ambient conditions at pH 5.5. Under thermophilic conditions, CO was efficiently converted, but it was mostly converted to H_2, which was then transformed to acetate. (Luo et al., 2018). Temperature determines the kinetics of mixed microbial consortia and represents one of the most important parameters during syngas biomethanation. The temperature of the culture can influence the microbial interactions among members of the microbial consortium and govern its major metabolic pathways. Thus, acetate is the principal precursor of methanogenesis under mesophilic conditions, according to several research on CO biomethanation. On the other hand, H_2 is the most relevant precursor under thermophilic conditions, as the higher diversity of carboxydotrophic hydrogenogenic bacteria in thermophilic bioreactors suggested. It has been also hypothesized that hydrogenogenic processes become more exergonic at higher temperatures, thus promoting a stronger hydrogenogenic conversion of CO under thermophilic conditions (Conrad and Wetter, 1990). In addition, it has been demonstrated that greater temperatures cause alterations in consortia microbial structure, which ultimately leads to higher conversion rates during syngas biomethanation (Grimalt-Alemany et al., 2018).

The type of culture also influences the biomethanation process. Due to its increased functionality, robustness and lower vulnerability to environmental inhibitors, mixed cultures enable a more successful syngas biomethanation performance than pure cultures (Esquivel-Elizondo et al., 2017; Hill et al., 2017; Weiss et al., 2017). Members of the microbial co-culture syntrophically cooperate and allow a “division of labour” during the overall bioproduction/bioconversion process (Lindemann et al., 2016). Recent studies (Grimalt-Alemany et al., 2020b) have investigated the performance of *Acetobacterium* sp., *Methanospirillum hungateii*, *Methanospirillum stamsii* and *Methanothrix* sp. at mesophilic conditions, and *Thermincola carboxydiphila* and *Methanothermobacter* sp. at thermophilic conditions. These studies concluded that the microbial selection was not driven only by kinetic competition, since thermodynamic limitations also played a key role defining the dominant catabolic routes.

Low gas–liquid mass transfer typically limit biomethanation process (Andreides et al., 2022). The poor mass transport of gaseous CO and H_2 to the anaerobic cells due to their low aqueous solubility (Henry’s law constants, $H_{CO} = 42$ and $H_{H2} = 52$ at 25 °C). The volumetric mass transfer rate of CO and H_2 from the syngas (G) to the microorganism-containing aqueous phase (A) can be expressed as a function of $Kla_{G/A}$ (overall volumetric gas–liquid mass transfer coefficient, s^{-1}), and the CO or H_2 concentration gradient (g m-3) in the liquid side ($[Pollutant]_{G/H}$ - $[Pollutant]_A$). Thus, these high H values typically result in a low driving force for the mass transport of CO and H_2 from the syngas to the aqueous phase surrounding/embedding the anaerobic microbial culture, and therefore in a limited CO and H_2 bioconversion. A low gas–liquid mass transport entails process operation in large gas-phase bioreactors, which significantly increases both investment and operating costs (Asimakopoulos et al., 2018). Therefore, the development of next generation biomethanation processes based on syngas bioconversion requires the engineering of compact high mass-transfer bioreactors capable of supporting an efficient mass transport of CO and H_2 (Figueras et al., 2021).

The performance of syngas biomethanation is mainly determined by mass transfer processes of syngas components, which are dependent on both the volumetric mass transfer coefficient (determined by the characteristics of the bioreactor) and the partial pressure of these gases as the driving force for their transport to the microbial community (Grimalt-Alemany et al., 2018). The mass transfer of gas substrates to the liquid medium is proportional to the substrate partial pressure in the head space, because difference concentration is the driving factor for mass transfer (Mohammadi et al., 2011). The pressure of CO, P_{CO}, influences cell proliferation and product synthesis. An increase in P_{CO} can result in an increase in cell concentration as a result of the enhanced mass transfer, but also microbial inhibition due to CO toxicity. Furthermore, when the P_{CO} was increased above 1.35 bar the pathway of acetic acid toward ethanol generation was boosted (Hurst and Lewis, 2010). Similarly, (Skidmore et al., 2013) reported an enhanced conversion of acetic acid to ethanol when increasing the amount of CO as a result in the higher availability of reducing equivalents.

6. Process configurations

Cross draft gasifiers have been widely used for the production of syngas devoted to biological or catalytic methanation (Hauser et al., 2021). Recent investigations have employed this gasifier, obtaining a high methane productivity and a good CO and H_2 conversion with real syngas (Asimakopoulos et al., 2021). FBR gasifiers have also been used for the simultaneous biomethanation of exogenous and endogenous CO_2. In an unique two-stage method for biological conversion of syngas to biomethane, FBR reactors have also been successfully integrated with thermophilic anaerobic digestion of sewage sludge (Andreides et al., 2021). The authors concluded that the amount of hydrogen in syngas was the most important element in influencing the amount of CH_4 in biomethane. The also stated that H_2 concentration in syngas was the main factor determining the CH_4 content in biomethane. This section of the review will focus on the most common bioreactors for syngas biomethanation. Table 2 summarizes the main results from the most common bioreactor configurations devoted to the biomethanation of syngas.

6.1. Stirred-tank bioreactors

Stirred-tank bioreactors (STBRs) are the most common reactors for culturing biological agents like cells, enzymes, and antibodies. They are contactors that rely on internal mechanical agitation to keep the phases (gas, mineral medium and microorganisms) well mixed. The impeller must agitate at a fast-enough rate to disperse all phases and achieve a uniform concentration inside the bioreactor. The volume required for a bioprocess is determined by the technical design of an STBR, which is dependent on the production (Jafarinejad, 2017). STBRs are frequently used for the intensification of gas–liquid mass transfer in microbial fermentations and can be eventually used to support syngas biomethanation. The main parameter describing the intensity of CO and H_2 mass transfer in stirred tank reactors is the volumetric mass transfer coefficient k_La (Petříček et al., 2018). Several factors determine the k_La, including the geometry, impeller configuration, agitation speed, and gas flow rate of the reactor. Higher gas–liquid mass-transfer rates are often achieved at high agitation speeds and syngas flow rates, which increases the gas–liquid interfacial area due to bubble break-up. The main limitation of this bioreactor configuration is the high shear stress caused by mechanical agitation, which can damage cell integrity and ultimately deteriorate syngas bioconversion (Diender et al., 2018).

6.2. Trickling bed filters

Trickling bed filters (TBF) consist of a column packed with inert materials of high specific surface area, on which biofilm is developed. Syngas is pumped through the packed bed either downwards or upwards and a nutritious liquid media is trickled and recycled over the packing material to provide moisture and nutrients, forming a thin liquid layer

Table 2
Summary of the most recent syngas biomethanation studies.

Culture	Reactor	Operation mode	Feed syngas composition (%)	Vol. (L)	pH	T (°C)	GRT (h)	Final CH_4composition (%)	CH_4 Yield (mol CH_4/ mol syngas)	Reference
Co-culture *C.hydrogenoformans* and *M. thermoautotrophicus*	CSTR	Batch & Cont.	H_2/CO_2 (66.6/ 33.3)	1.50	7.2	65	2.08	70.0	Nd	(Diender et al., 2018)
ADS	CSTR	Batch.	$H_2/CO_2/CH_4/N_2$ (80/20/0/14.7)	9.50	8.1	55	1.19	49.0	0.16	(Voelklein et al., 2019)
ADS	CSTR	Cont.	$H_2/CO_2/CH_4/N_2$ (54/14/32/0)	9.50	8.1	55	1.14	61.0	0.22	(Voelklein et al., 2019)
Mixed microbial consortia (WWTP anaerobic sludges)	TBR	Cont.	$H_2/CO_2/CO/N_2$ (45/25/25/10)	0.18	7.0	37	3.00	67.0	Nd	(Asimakopoulos et al., 2020)
Mixed microbial consortia (WWTP anaerobic sludges)	TBR	Cont.	$H_2/CO_2/CO/N_2$ (45/25/25/10)	0.18	7.0	60	1.00	86.0	Nd	(Asimakopoulos et al., 2020)
Pseudomonas sp, *Methanobacterium formicicum, Peptococcaceae*	TBR	Cont.	$H_2/CO_2/CH_4/N_2$ (62/15/23/0)	0.80	7.1	54	4.00	96.0	Nd	(Ghofrani-Isfahani et al., 2022)
Digestate from biogas plant (mainly *M. thermautotrophicus* & *Clostridia* sp.)	TBR	Cont.	$N_2/CO_2/CH_4$ (65/15/23)	1.20	8.0	52	8.00	66.0	Nd	(Kougias et al., 2017)
Digestate from biogas plant (mainly *M. thermautotrophicus* & *Clostridia* sp.)	BC	Cont.	$N_2/CO_2/CH_4$ (65/15/23)	1.40	8.0	52	8.00	73.0	Nd	(Kougias et al., 2017)
MACE	BC	Cont.	$H_2/CO_2/CH_4/N_2$ (65/17.5/0/ 17.5)	0.29	7.5	52	1.32	67.1	0.18	(Sieborg et al., 2020)
MACE	BC	Cont.	$H_2/CO_2/CH_4/N_2$ (62/15/23/0)	1.00	8.3	54	2.1	95.1	0.25	(Porté et al., 2019)
ADS - *Coprothermobacter* & *Methanobacterium*	BC	Cont.	H_2/CO_2 (4.2 ratio)	22.00	7.5	55	Nd	94.0	Nd	(Léa et al., 2022)

ADS: anaerobic-digester sludge; BC: bubble column; Cont.: continuous; CSTR: Continuous stirred-tank reactor; GRT: gas residence time; MACE: mixed anaerobic culture enriched with hydrogenotrophic methanogens; Nd: not defined; TBR: tickling bed reactor; T: temperature; Vol.: volume. WWTP: Waste water treatment plant.

over the biofilm. The biofilm is composed of a specific arrangement of immobilised cells within a matrix of extracellular polymeric substances (Porté et al., 2019). These type of bioreactors are a suitable alternative to STBRs, being an efficient system to achieve high CH_4 quality and production capacities at lower energy demands (Strübing et al., 2019). In TBRs the microbes are immobilized on the packing material, which should have a high surface-area for gas–liquid mass transfer in order to favour a high density and activity of methanogenic archaea (Dupnock and Deshusses, 2017). The features have supported high H_2 conversion and CH_4 production rates in lab scale test (Sieborg et al., 2020).

TBRs under continuous mode inoculated with enriched mixed microbial consortia for syngas biomethanation have been poorly studied (Asimakopoulos et al., 2020). Due to the fact that hydrogenotrophic communities are capable of developing stable biofilms supporting a robust methanogenic activity, recent research activity has been focused on dedicated studies of trickling bed reactors for biological syngas methanation and biogas upgrading by mixed microbial cultures (Thema et al., 2019). However, operational limitations such as the high liquid recirculation costs or the high cost of synthetic packing media must be overcome to facilitate. (Thapa et al., 2022) have recently validated CO_2 biomethanation in these bioreactor configurations with a high CH_4 production rate (up to 2.65 $L \cdot L_{residue}^{-1} \cdot d^{-1}$ and maximum 98% CH_4 content) with a 100% H_2 utilization efficiency and *Methanoculleus bourgensis* as the dominant species in the liquid and biofilm phases.

6.3. Bubble column and gas-lift bioreactors

The use of bubble columns and gas-lift reactors in syngas biomethanation processes has also been investigated based on their large gas–liquid interfacial areas, high volumetric mass transfer coefficients, non-mechanical mixing, and low operating costs. The gas–liquid mass transfer coefficient in these bioreactor configurations is largely determined by the gas flow rate and the size of the bubbles (Grimalt-Alemany et al., 2018). The influence of these operational parameters on CO mass transfer was investigated in both a bubble column and a gas-lift reactor, with the K_La increasing as the gas flow rate and the pore size of the column diffuser decreased (Munasinghe and Khanal, 2010). (Léa et al., 2022) have recently evaluated the performance of a pilot scale bubble column reactor for ex situ biological methanation of syngas with mixed microbial culture, achieving a 94% syngas conversion (H_2/CO_2) into methane at 4 $N \cdot L^{-1} \cdot d^{-1}$. Similarly, (Kougias et al., 2017) reported the superior CH_4 production performance of bubble column reactors compared to TBR (73% vs 66% respectively) under the same working conditions. Gas-lift bioreactors use the power created by gas injection and the density difference between gas–liquid mixture and the liquid to circulate the gas–liquid mixture. This particular gas–liquid circulation pattern will enhance mass transfer, heat transfer and mixing (Li, 2017). Unfortunately, the number of studies assessing the performance of gas-lift reactors for syngas biomethanation is very limited and the works were carried out few decades ago. For instance (Guiot et al., 2010) bio-upgraded syngas into methane employing microbial granules derived from a wastewater treatment plant in a 30 L gas-lift reactor supplied with a gas mixture containing carbon monoxide at different gas feeding and recirculation rates. The yields achieved in this study (i.e. 60% CH_4) were lower than those reported in recent studies with alternative bioreactor configurations.

7. Research needs and future directions

The complex geopolitical situation and the high dependency of national economies on countries that export fossil fuels (especially natural gas and oil) is triggering research boosting biomethane production from biomass waste. In this context, technological advances that five years ago were a long-term project, start to become a reality today. Thus, novel technologies for syngas pre-treatment operating at low energy demands are nowadays needed. A variety of syngas cleaning techniques have been developed, including cyclones, electrostatic precipitators (Jeong et al., 2022), filters, rotating particle separators and water scrubbers (Tsai et al., 2021). Despite substantial advancements, these technologies must overcome the mass transfer limitation imposed by the hydrophobic nature of most tar components. In addition, a new generation of compact and high mass transfer bioreactors able of providing high syngas conversion efficiencies and methane productivities is needed. Gas-phase hollow fibre membrane reactors, bubble sprinklers, Taylor flow reactors or bioreactors based on bioactive coatings have been lately proposed to enhance the bioconversion of poorly water soluble substrates (Yasin et al., 2019). Bioactive coating-based biofilters represent an innovative biomethanation platform based on polymeric coatings (e.g. latex or hydrogels) engineered as nanoporous matrices with dense populations of anaerobes resulting in bioactive packing materials. The lab-scale production of H_2 using artificial photosynthetic leaves and indoor air treatment has been successfully demonstrated using this innovative biocatalytic approach (González-Martín et al., 2022). Bioactive polymeric coatings with a high affinity for CO and H_2 could be used in packed bed bioreactors to improve syngas uptake from the gas phase, avoiding the mass transfer limitations caused by the water layer that covers traditional biofilms. Most recent publications are based on the use of microbial consortia, whose symbiotic action is capable of increasing the yield of methane produced. However, a more detailed study of the different microorganisms involved as well as the metabolic routes associated would be of vital importance in order to optimize process performance. In addition, the optimization of the design and operation of bioreactors with immobilized co-cultures will also bring significant advances in the field of syngas biomethanation. The possibility of using mesophilic microbial cultures would eliminate all the disadvantages linked to thermophilic bacteria. In this context, (Mouftahi et al., 2020) recently reported that the biogas and bio-methane yield at 35 °C in terms of methane production per kg of volatile solids (~0.384 Nm^3 methane/kg) was enhanced during the co-digestion of three biowastes. The development of modelling tools capable of optimizing the variables involved in the syngas biomethanation process would entail a significant improvement of the waste to biomethane process. The integration of water electrolysis using renewable energies and biomethanation of the syngas produced from organic waste gasification will increase sectoral competitiveness and lower the footprint of bio-based industries.

8. Strategic importance, bottlenecks and potential solutions for biomethane

The EU has set more severe targets in terms of environmental protection, aiming at a near-zero emissions economy and 100% renewable energy production by 2050, which is critical to foster biogas technology (Cook, 2021). The relevance of biogas, especially biomethane, as a sustainable energy alternative is demonstrated in the increasing number of publications in the past 20 years (Calise et al., 2021). In this context, gasification represents a step forward towards a reduction in waste volume via conversion into energy in the form of syngas and other minor chemicals, which paves the way to the circularity of the process. Syngas must be upgraded to obtain a biomethane with an adequate quality for injection into the natural gas grid (Figueras et al., 2021). Thus, several biorefineries are planning or currently undertaken the upgrading of syngas to obtain a green substitute to natural gas while complying with current regulations encouraging a zero-waste concept–based bio-circular economy (Chakravarty and Mandavgane, 2022). Despite its potentiality, there are still important limitations in the scale up and commercialization of this technology, such as: (i) the presence of volatile siloxanes, (ii) the presence of organic compounds in the waste that are only partially degraded, (iii) the low reaction rates, which entails a large-capacity and more costly bioreactors, and (iv) the presence of excess CO_2, H_2S, and moisture together with methane, which makes the process less cost-effective (Jacob et al., 2020). Nevertheless, these inherent bottlenecks can be solved to a large extent by implementing different pre-treatment strategies (physical, chemical, biological and combined technologies) (Pascual et al., 2021). Another important limitation for the implementation of the biomethanation process is associated to the microbiology involved. In this sense, it is crucial to properly select the technology configuration and to carefully monitor and control critical operating parameters such as temperature, pH, mixing, retention time, or the presence of inhibitory substances (Adnan et al., 2019).

9. Conclusions

Organic waste gasification can produce a syngas with a composition governed by the nature of the waste, the type of gasifier, temperature, gasifying agent, etc. This syngas may be upgraded to biomethane using bioreactors operated at a low temperature and pressure. This process still exhibits several limitations in term of CO/H_2 mass transfer and microbiology, which are the basis for future studies in this area. The evolution of new bioreactor designs with a high mass transfer capacity at low operating costs, and based on synergistic co-cultures should pave the way of this novel waste-to-biomethane route.

CRediT authorship contribution statement

Sergio Paniagua: Writing – review & editing, Writing – original draft. **Raquel Lebrero:** Writing – review & editing. **Raúl Muñoz:** Writing – review & editing, Project administration, Supervision.

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.

Data availability

No data was used for the research described in the article.

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