Bioprocesses for resource recovery from waste gases: current trends and industrial applications

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Abstract

Air pollution is a topic of important global concern because it has contributed significantly to an increase in the earth's global warming potential and contributed to severe health and environmental impacts. In this review, the different bioreactor configurations commonly used for waste gas treatment, namely the biofilters, the biotrickling filters and the bioscrubbers, and their industrial applications were compared in terms of the type of inoculum, the packing material/media, removal efficiency and elimination capacity. Typically, biofilters are operated under the following range of operating conditions: gas residence time = 15–60 s; gas flow rate = 50–300,000 m^3 h^{-1}; temperature = 15–30 °C; pH = 6.0–7.5; filter area = 100–3000 m^2;

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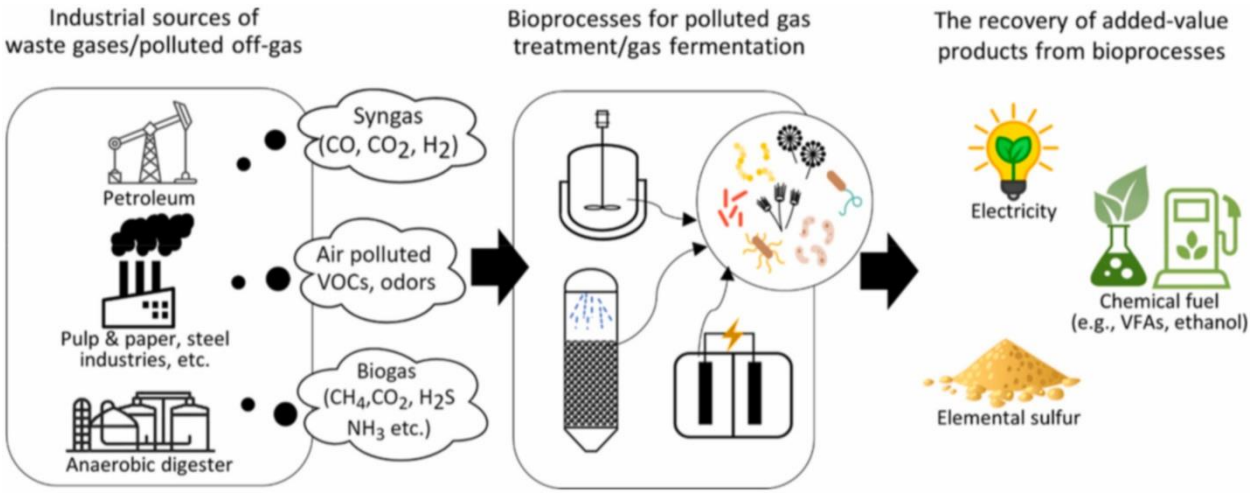
relative humidity >95.0%; and removal efficiencies >75.0% depending on the waste gas composition and concentration. The biotechnological approaches for resource recovery, i.e., the conversion of C1 gaseous compounds (CO, CO_2 and CH_4) to liquified value-added products or biofuels have been discussed. From this review, it was evident that the performances of different aerobic, anoxic and/or anaerobic lab, pilot and full-scale bioreactors for waste gas treatment and resource recovery depend on the composition, the individual concentration of pollutants present in the waste gas and the gas flow rate. Although most of the research on product recovery from waste gas is rather limited to lab/pilot-scale studies, there are some key commercialized technologies that have proven to be economical at the full-scale. Thus, this review, comprehensively presents a complete overview of the current trends and limitations of conventional waste gas treatment systems, the benefits of novel bioreactor configurations and their potential to be applied for resource recovery from waste gases.

Graphical abstract



Keywords

Waste gas, Bioreactors, Volatile pollutants, Syngas fermentation, Elimination capacity, Resource recovery

1. Introduction

Air pollution has been a great threat alleviating the problem of global warming and according to the WHO [1], it is the single largest contributor to negative health and environmental impacts in the 21st century. Due to stringent environmental regulations, although the air quality in many parts of the world has improved over the last few decades, the levels of atmospheric air pollutants in many developed and developing nations still exceed the guidelines prescribed by

the WHO. The most commonly monitored air pollutants are particulate matter (PM) and gaseous pollutants such as ozone, volatile organic and inorganic compounds and oxides of nitrogen, carbon and sulfur [2]. The European Environmental Agency (EEA) has classified six important sources of air pollution, as follows: (i) intense agricultural activities, (ii) waste burning and mining activities, (iii) energy production facilities, (iv) transportation sector, (v) natural phenomena, and (vi) fuel combustion contributing to indoor activities [3]. However, among the different air pollutants, the volatile organic compounds (VOCs) are an important class of pollutants and they are considered as hazardous air pollutants (HAPs) by the United States Environment Protection Agency (USEPA) [4]; IARC, 2012) because they are released from wastewater treatment plants, composting facilities, vehicle exhaust, process industries (e.g., petrochemical, pharmaceutical, paint and varnish, pulp and paper) and the use of solvents [5].

On the other hand, there has been significant biotechnology-based research at the pilot scale for utilizing syngas (CO and H_2 as the major fraction, CO_2, CH_4, H_2S, and NO_x as the minor fraction), i.e., through the process of syngas fermentation, and converting them to liquid fuels and platform chemicals or other value-added products [[6], [7], [8]]. Thus, in order to promote the concept of circular bio-economy in biorefineries, it is important to convert waste gas into resourceful products that can be used as a fuel or raw material in other industrial processes. The conventional waste gas treatment process includes bioreactor configurations such as the biofilters, biotrickling filters (BTF) and the bioscrubbers. They have proven to be effective for the treatment of a wide variety of VOCs (e.g., mercaptans, benzene, toluene, methanol, methyl ethyl ketone and formaldehyde) and volatile inorganic pollutants (VICs) (e.g., ammonia, H_2S) [9].

However, it is noteworthy to mention that the removal efficiency (RE) of the pollutants in a multicomponent gas mixture depends on the source and composition of the waste gas, its physico-chemical characteristics (hydrophobic/hydrophilic) and the concentration of individual pollutants present in the complex mixture [10]. Besides, in industrial situations, when the concentration of the waste gas exceeds a threshold limit under transient loading conditions, it is advantageous to combine a physico-chemical treatment step (e.g., adsorption and UV photoreactor) with the biological step [11]. Thus, in the last 10 years, with advancements in scientific and analytical technologies, there have been innovations in waste gas treatment. Rather than focusing on the conventional end-of-pipe treatment for waste gas, the focus has shifted towards resource recovery from waste gas using innovative bioreactor configurations, yielding promising results at the pilot and industrial scales. The main aim of this review is to compare and discuss the performance of lab, pilot and industrial scale bioreactors for waste gas treatment, their mechanisms, and recent innovations in the field of resource recovery from waste gas.

2. Industrial bioreactor configurations: pilot and full-scale case studies

Table 1 provides detailed information about the construction and operational characteristics and performance of various bioreactor configurations such as biofilters, trickle-bed biofilters, and bioscrubbers. Evidently, both pilot and full-scale experiments have been reported, ranging from 35 to 540 days of operation, with inlet VOCs such as benzene, toluene and xylene, and VICs such as H_2S, mercaptans, among others. The highest inlet loading rate reported was using Ecobase™ packing media (524 g m^{-3} h^{-1}) while treating H_2S [33]. In that study, at residence times varying between 2 and 10 s, the BTF removed more than 99.0% of the pollutant at an ILR of 247 g m^{-3} h^{-1}. Lackey et al. [13] reported longer empty bed residence time (EBRT) (15–54 min) than the conventional biofiltration or waste gas treatment systems (i.e. <2 min) during the operation of a trailer-mounted biofiltration system that includes an air stripper, biofilter unit, process control systems, gas chromatograph, online data logging/acquisition system, and a ~1900 L propane tank to treat trichloroethylene (TCE)-contaminated air stream from the Anniston Army Depot located in Anniston, USA. Different bioreactor types have individual operational requirements (Fig. 1) and their application and selection can be based on different purposes, e.g. pollutant types and concentrations, cost and location. There are few specialized biofilters such as rotating drum biofilters (RDB), tubular-type biofilters, biowindows, biocovers etc. that have been discussed with regard to their pilot- and full-scale applications. Though relatively lesser number of investigations has been reported on the pilot and full-scale bioreactors, countries such as China, USA, Taiwan, The Netherlands, Spain, and Poland have installed such bioreactors in various industries. Hence, understanding the dynamics of microbial population in the bioreactors, design characteristics of bioreactor systems and their operational parameters including the inlet concentration or pollutant load, pH, temperature, flow rate, and residence time is important, which would provide exhaustive information in developing robust bioreactor systems for their optimum performance.

2.1. Biofilter

Biofiltration is widely used for the treatment of waste gases such as volatile organic compounds (VOCs) and VICs (particularly, H_2S and NH_3) emitted from various process industries like textile dyeing industry, composting manure and dead pig pits, and storage tanks [[12], [13], [14], [15]]. To improve the REs and elimination capacities (ECs) of the biofiltration systems, optimization of EBRT and inlet concentration is very much important. This can be achieved by modifying the biofiltration technologies, improving the inlet feeding strategies and designing advanced/hybrid reactor configurations.

Table 1. Pilot and full-scale bioreactor configurations employed for waste gas treatment.

Pollutants treated	Scale of operation	Time of operation (d)	Inoculum used	C_{in} or IL	RE (%)	EC (g m^{-3} h^{-1})	Process parameters			Bioreactor size	Packing material	Packing height (m)	Region and references
							pH	EBRT (s)	Q (m^3 h^{-1})				
Biolfilters													
VOCs[1], H_2S	Pilot-scale	420	Mainly *Thiobacillus*	4.5 g m^{-3} h^{-1}	>90.0	0.86	4.0	12	1020	2.3 m dia.	Black lava rock	1.2	USA [16]
Toluene, H_2S, NH_3	Pilot-scale	>400	Mainly Firmicutes and Proteobacteria	3.8 m^3 m^{-2} min^{-1}	>80.0	NA	NA	90	60	1 m × 1 m	PUF[15] cubes	2	China [17]
TCE[2]	Pilot-scale	351	Propane oxidizing consortium	0.3–3.8 g m^{-3} d^{-1}	32.3–68.4	0.32	NA	2040–3240	NA	2.4 m × 4.88 m	Compost poultry litter, kenaf, dolomitic limestone	2.74	USA [13]
H_2S, NH_3	Pilot-scale	>150	Mainly *Bacillus*	5.37, 0.14 g m^{-3} h^{-1}	91.9, 100	NA	4.6, 1.5	27.98	3	0.011 m × 0.007 m	Polyethylene	1.41	Australia [18]
VOCs (NAOCCs[3], AlHs[4], AHs[5], HHs[6])	Pilot-scale	90	Mainly Proteobacteria	NA	11.7–67.9	NA	7.0–7.5	15.2	22000	8 m × 8 m	Ceramic particles, wood chips, bamboo charcoal	2.6	China [14]
VOCs	Pilot-scale	~120	Hydrocarbon degrading bacteria	4.2 g m^{-3} h^{-1}	86.0	5.3	5.6	60	76.46	2.43 m × 1.83 m	Open-pore reticulated PUF cubes	NA	USA [19]
NH_3, DMS[7], DMDS[8], DMTS[9], TMA[10]	Pilot-scale	42	Nitrogen oxidizing bacteria	53.2 g m^{-3} h^{-1}	88.7, 95.4, 94.0, 99.1, 100	47.2, 0.22, 1.3, 0.29, 0.6	6.8–7.5	60	2.543–8.478	0.3 m × 1 m	Mature compost on sieve	1.2	China [15]
VOCs, H_2S, NH_3	Pilot-scale	NA	Compost bacteria	5–500 g m^{-3} h^{-1}	93.0–99.0	2.33, 0.97, 2.25	6.8–8.4	43–45	NA	1.3 m × 3 m	Stumpwood chips-bark-compost bed	NA	Poland [20]
Toluene and xylene	Pilot-scale	240	ATCC[12] 31483, 39213, 21499, 15590	143 g m^{-3} h^{-1}	70.0, 50.0	80 and 35	NA	78 and 102	5	0.3 m dia.	Compost and conditioned peat	2	Canada [21]
H_2S	Full-scale	~210	Primarily *Bacillus*	55.15 g m^{-3} d^{-1}	90.0	0.41–2.21	6.5–7.0	60	60	1 m^3 (volume)	PUF	1.2	China [22]

Pollutants treated	Scale of operation	Time of operation (d)	Inoculum used	C_{in} or IL	RE (%)	EC (g m^{-3} h^{-1})	Process parameters			Bioreactor size	Packing material	Packing height (m)	Region and references
							pH	EBRT (s)	Q (m^3 h^{-1})				
NH_3, VOCs	Full-scale	125	Activated sludge from WWTP[13]	0.16–0.55 g m^{-3}	97.0–99.0	149–113	7.0	15	3000	1.8 m dia.	PUF	1	Korea [23]
p-Xylene	Full-scale	~540	Aerobic xylene degrading consortium	20–500 g m^{-3} h^{-1}	95.4	19.8–46.5	NA	55–77	NA	1.9 m × 1.7 m	Compost	3.6	Taiwan [12]
Toluene	Full-scale	391	Activated sludge from WWTP	18.7–149.3 g m^{-3} h^{-1}	99.0	18.3–83.0	NA	30–5	0.25–1	0.16 m dia.	PUF	0.3	China [24]
Odors, VOCs, bioaerosols	Full-scale	307	Activated sludge from WWTP	0.95–41.26 mg m^{-3}	86.0–98.0	NA	4.0–5.0	24–48	5760	52.8 m^3 (volume)	PUF and volcanic rock	5.6	China [25]
Biotrickling Filters													
Butyl acetate, n-butyl alcohol, phenylacetic acid	Pilot-scale	>90	Activated sludge from WWTP	373.4, 317.2, 209.5 g m^{-3} h^{-1}	92.0–100	NA	NA	28.42	3.5–9.5	0.075 m^3 (volume)	ZX02 carrier	2.8	China [26]
H_2S	Pilot-scale	100	SOB[14] from Activated sludge-WWTP	0.59–5.0 g m^{-3} h^{-1}	>99.0	NA	3.3–7.4	25	80	0.6 m dia.	Bamboo charcoal and ceramsite	3	China [27]
VOCs	Pilot-scale	365	Activated sludge from WWTP	5–72 g m^{-3} h^{-1}	>60.0	NA	6.5–7.5	10–40	65–300	0.75 m^3 (volume)	Flexiring™ propylene rings	NA	Spain [28]
VOCs	Pilot-scale	>35	Proteobacteria and Firmicutes dominated	NA	>90.0	NA	NA	32–59	1008–550	2 m dia.	Ceramsite	6	China [29]
VOCs	Pilot-scale	365	Activated sludge from WWTP	500–600 mg C m^{-3}	80.0–95.0	NA	6.5–7.5	10–40	65–300	0.75 m^3 (volume)	Polypropylene rings	NA	Spain [30]
Styrene	Pilot-scale	>120	*Pseudomonas* (E−93486)	NA	78.0–94.2	NA	7.0	41–62	100–150	1.084 m × 3.5 m	Polypropylene Ralu rings	1.8	Poland [31]
Styrene	Pilot-scale	365	Activated sludge from WWTP	24.9 g m^{-3} h^{-1}	75.6	18.8	>6.7	31–66	33–71	0.6 m^3 (volume)	PAS™ Winded Media	NA	Spain [32]
H_2S	Full-scale	180	EcoFilter EF51 installed bacteria	247–524 g m^{-3} h^{-1}	99.0–95.0	~400	NA	2.8	2803.5	1.52 m dia.	EcoBase™ synthetic media	2.74	USA [33]

Pollutants treated	Scale of operation	Time of operation (d)	Inoculum used	C_{in} or IL	RE (%)	EC (g m^{-3} h^{-1})	Process parameters			Bioreactor size	Packing material	Packing height (m)	Region and references
							pH	EBRT (s)	Q (m^3 h^{-1})				
MeSH[11], H_2S, VOCs	Full-scale	>365	Activated sludge from WWTP	0–15 ppm_v	90.0–100, 98.9, 93.0	(0.43–1.16, 5.0) g m^{-3} h^{-1}, 82.7 × 10^5 OU_E m^{-3} h^{-1}	2.6–8.0	10–50	2432 ± 121	400 m^2 m^{-3} (specific SA[16])	OdourPack™ structured plastic	5	UK [34]
Trickle-bed biofilters													
p-Xylene	Pilot-scale	118	Aerobic xylene degrading consortium	200 ppm_v	>80.0	NA	NA	96–32	3.6–10.8	1.5 m × 0.4 m × 0.4 m	Compost	0.2	Taiwan [35]
VOCs, H_2S	Pilot-scale	>60	*Thiobacillus* sp.	20 g m^{-3} h^{-1}	>95.0	18	5.5–7.5	NA	2–30	2.2 m × 0.8 m	Polyethylene rings	1.2	Poland [36]
Bioscrubbers													
VOCs	Pilot-scale	484	Mainly *Methanosaeta*	1126 ± 470 mg N m^{-3}	97.0	NA	6.8–8.8	NA	184–1253	3.06 m × 0.5 m	Cross-fluted flow fills, KFP319/619	2	The Netherlands [37]

Note: C_{in} – Inlet concentration, IL – Inlet loading, RE – Removal efficiency, EC – Elimination capacity, EBRT – Empty bed residence time, RE – Removal efficiency, NA – Not available, OU_E m^{-3} h^{-1} – European odor unit. **Superscripts:** 1 = Volatile organic compounds, 2 = Trichloroethylene, 3 = Nitrogen- and oxygen-containing compounds, 4 = Aliphatic hydrocarbons, 5 = Aromatic hydrocarbons, 6 = Halogenated hydrocarbons, 7 = Dimethyl sulfide, 8 = Dimethyl disulfide, 9 = Dimethyl trisulfide, 10 = Trimethylamine, 11 = Methanethiol, 12 = *Pseudomonas putida* (ATCC 31483), *Pseudomonas putida* (ATCC 39213), *Rhodococcus* sp. (ATCC 21499) and *Arthrobacter paraffineus* (ATCC 15590), 13 = Wastewater treatment plant, 14 = Sulfur oxidizing bacteria, 15 = Polyurethane foam, 16 = Surface area

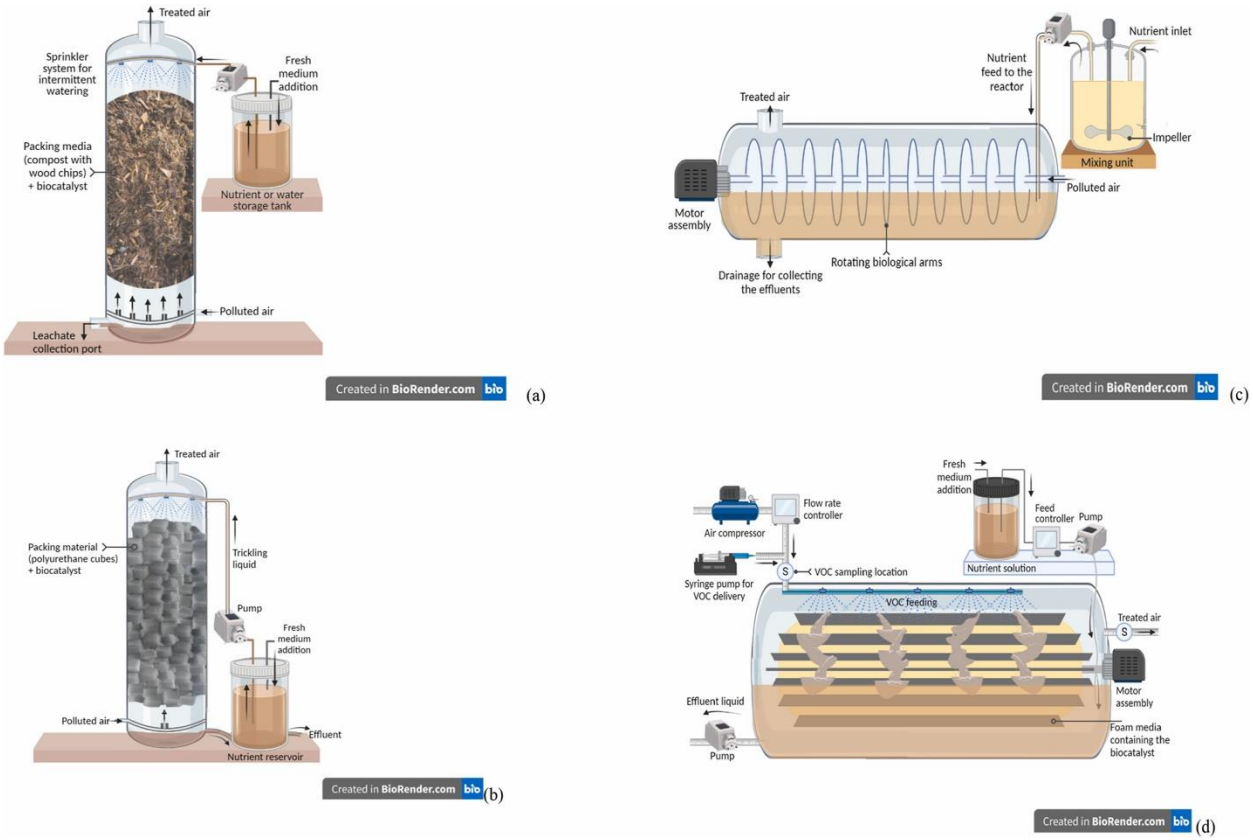


Fig. 1. Schematic of: (a) biofilter, (b) biotrickling filter, (c) rotating disc biofilter and (d) rotating drum biofilter.

2.1.1. Biofilter configuration

To cater the requirements of the process industry, biofilters are arranged in several patterns (single-staged or two-staged) in order to increase the overall performance of compost biofilter [12,15], biocovers [38,39], biowindows [40], tubular [24], trickle-bed biofilter [35], RDB [19] and combining the existing biofilter with other reactors by including adsorption zones [25] and chemical scrubbers [14] (Table 1). A novel pilot-scale RDB was found to treat VOCs released from a paint drying unit with EC ranging from 0.125 to 5.25 g COD m^{-3} h^{-1}. The RDB offered several benefits including the provision of a medium for the uniform distribution of pollutants, nutrients and moisture on the biofilm, resulting in a stable configuration amenable for high VOC loadings (~4 g COD m^{-3} h^{-1}). During the shutdown periods when the pollutants were not fed to the bioreactor, the biofilm was supplied with sucrose as an auxiliary substrate to eliminate the degradation of microbial activity and a RE of ~86.0% was achieved during normal operation [19]. Various types of RDBs have been developed, including single- and multi-layer RDBs and hybrid RDB. Yang et al. [41] have tested those three RDB types for the removal of VOCs (i.e., diethyl ether, toluene, and hexane) at a loading rate of 2.0 kg COD m^{-3} d^{-1} and the hybrid RDB showed the highest RE compared to other reactor types. Many practical difficulties are

encountered during the simultaneous removal of more than one pollutant including reduced REs, which is attributed to the competitive inhibition by pollutant degrading bacteria feeding both the primary substrate and target compound at once [13].

For the simultaneous removal of VIC pollutants like H_2S and NH_3, treatment technologies like scrubbers and adsorption zones were installed along with the biofilter to form a fully integrated reactor. One such reactor system installed at a wastewater treatment plant (WWTP) in Australia emitting H_2S and NH_3 resulted in an average RE of 92.0% and 100% with an average mass inlet loading of 5.37 g S m^{-3} h^{-1} and 0.14 mg N m^{-3} h^{-1}, respectively [18]. The sulfuric acid formed by the presence of sulfur oxidizing bacteria (SOB) was scrubbed with the incoming NH_3 in another reactor to generate ammonium sulfate that was extracted and recovered through the leachate. Since the growth of nitrogen degrading bacteria is difficult at such low pH conditions (pH <2.0), chemical treatment of ammonia is usually preferred [18]. A multi-layered RDB designed to remove VOCs (toluene and diethyl ether) was able to demonstrate RE >99.0% because of maintaining optimal conditions like moisture content, depth of submergence, pH, inlet concentration and gas flow rate [42]. Therefore, designing biofilters depends on the inlet gas streams (composition and concentration), biomass accumulation patterns and biomass control strategies.

Chemical neutralization has been reported in other studies where H_2S is bio-oxidized into sulfur and sulfate whereas ammonia is converted to nitrate or dissolved into the liquid phase, which indicates that the previously mentioned technique can be potentially used for closed-loop removal of ammonia (RE >90.0% for H_2S and NH_3) [43]. The adsorption zones also help in regulating the flow rates of pollutants that are fed to the microorganism, as the flow rate of a biofilter plays an important role to determine the RE of a biofilter (decreases with increased flow rate) [25].

2.1.1.1. BIOCOVER

Biocover is a type of huge biofilter system that functions as a landfill top cover that have been used in the recent times for simultaneous CH_4 and odor mitigation by biological conversion (mainly using methanotrophic bacteria) in an apt environment curated under the polyethylene sheets covering a landfill or dewatered sludge zone of a WWTP. The biocover land is filled with waste compost, sewage sludge or biologically treated waste. The REs achieved by a pilot-scale biocover with compost, soil and perlite was more than 85.0% in all seasons for CH_4 and odor removal [39]. This type of biofilter was found to be very effective for the removal of odor in a landfill site in Canada with the concentration of odors ranging from 640,000 to 4,000,000 OU m^{-3} (RE ~100%) [38].

2.1.1.2. BIOWINDOW

Biowindows are biofilters having similar features of biocovers like the amount of spread onto the landfill and bacterial composition (methanotrophic bacteria). A full-scale study was conducted at a landfill site in Italy with comparisons demonstrated between traditional biofilter and biowindow (filled with compost). The REs for CH_4, odor and non-methane VOCs in the biowindow were 88.0%, 93.8% and >80.0%, respectively, with the inlet odor activity values of 19096 $OU_E\ m^{-3}$ [40].

2.1.2. Biofilm

The biofilm in a biofilter comprises of microorganisms needed for the degradation of gaseous pollutants like VOCs, amines, mercaptans and sulfur-containing compounds. The biofilm, developed on the packing material, requires suitable environmental conditions such as moisture, temperature, and pH for the survival and growth of microbial communities. Activated sludge (mostly from municipal WWTP) and inoculum from previously operated biofilters have been extensively used for inoculation in biofilters and biofilm development [22,44]. Han et al. [17] studied the co-treatment of VOCs and odor at a domestic waste landfill site in Beijing wherein biofilters were installed at two locations on the same site, sealing zone and leachate treatment zone (LTZ). The bacterial diversity and pollutant intake capacity were different (~1.5 times higher in LTZ) in these two zones where the dominant bacteria were *Mycobacterium* in the sealing zone and *Bacillus* in the LTZ, indicating that the differences in the composition of inlet gaseous pollutants control the evolution of bacterial community in the pilot-scale biofilter. Considering the practical difficulty to maintain the original inoculum in a full-scale waste gas treatment system, the inoculated biocatalyst does not need to remain dominant as long as the performance remains high (RE >85.0%). The time required for the biofilm development influences the pollutant removal process and the start-up time usually ranges from 5 days to few months, depending on the type of biofilter configuration. In several studies, external inoculation of microorganisms in the laboratory or activated sludge from an old biofilter has shown to significantly reduce or even eliminate the lag phase resulting in higher REs (>90.0% for H_2S and VOCs) within the first week of operation [14,17,22]. The formation of biofilm should be monitored periodically because excess biomass accumulation can cause pressure drop, clogging, channeling and a decline in the reactors long-term performance. Hence, understanding the structure of biofilm formation in the biofilter helps the plant operators to choose the right biomass control strategies that can be implemented in their industry [43].

2.1.3. Packing material

Packing material is yet another important component of a biofilter to provide a platform for microbial support and growth. Several types of packing materials like polyurethane, polyethylene, lava rock, and compost have been used in different biofilter systems and each of

them has its own merits. For a large scale biofilter to be set-up, a packing material must be (i) cost-effective with least compaction over time, (ii) should have a high specific surface area for biofilm growth and gas-biofilm mass transfer, (iii) should possess high porosity for the homogeneous aeration of gases and (iv) should have mineral and high-water retention capacity to maintain the desired moisture level and minimize bed drying [16,19,22,25,40].

Polyurethane foam (PUF) cubes are the most commonly used packing material in biofilters for the removal of VOCs, ammonia, bioaerosols and odorous gaseous pollutants [16,18,21,24]. They possess characteristics such as high porosity (97.0%), low density (19 kg m^{-3}), high water adsorption capacity and adequate pore size which provide the best conditions for inoculation and bacterial growth (similar properties are found for rock lava and polyethylene). Composts are yet another widely used packing material in biofilters wherein instead of providing an inorganic packing, waste matter or dewatered sludge in combination with pebbles and sand is used and therefore, inoculum preparation is not needed [12,13,21]. The RE in a compost biofilter is highly affected by factors such as the source of compost (domestic waste manure, WWTP sludge, etc.), pH of the compost, oxygen concentration and nutrients, among others. Temperature is a very important factor that must be considered for compost systems as certain disease-causing bacteria and viruses can be eliminated only above temperatures of 35 °C (e.g., 40–50 °C for pathogenic avian influenza viruses and Newcastle disease virus). By maintaining an overall composting temperature >55 °C for simultaneous removal of NH_3 and VOCs, an average RE of 93.1%, average EC = 603 mg m^{-3} h^{-1} (VOCs) and 47 g m^{-3} h^{-1} (NH_3) was observed in a compost biofilter with inlet NH_3 concentration of 12.7–446.8 mg m^{-3} [15].

The height of the packing material affects the operational efficiency of a biofilter, as demonstrated in a landfill site in China for H_2S removal [22]. At an inlet load of 55 g m^{-3} d^{-1}, the EC at the lower part and upper part of the biofilter was 2.21 g m^{-3} h^{-1} and 0.41 g m^{-3} h^{-1}, respectively indicating that the lower part of biofilter was encountered with the most loads, thereby resulting in higher EC [22]. The configuration of packing media is also an important factor that impacts the performance of a biofilter. For example, a RDB was assembled in two configurations, namely single-layered and multi-layered (four concentric polyurethane foam layers) that showed better stability, even distribution of the biomass and higher RE (~99.0%) of diethyl ether, even at higher organic loading rates (8.0 kg COD m^{-3} d^{-1}) [45]. This study showed the potential of applying RDB to be used as full-scale systems because of its ability to avoid channeling and by-pass of the gas-stream during long term operation.

2.1.4. Operating parameters

The parameters that are needed to be considered while operating a full-scale biofilter for the survival of microorganisms and optimum removal of pollutants includes pH, temperature, contact time or the EBRT, water affinity (or hydrophilicity), moisture content and gas flow rate of the pollutants. These parameters are difficult to maintain in reactors of different sizes and also with changing environmental conditions. For example, a study conducted in China, where the

daily outside temperatures were fluctuating between −5.6 °C and 14.3 °C, it was reported that the microbial activity increased the overall temperature of the compost in biofilter which was maintained by continual monitoring and mechanically turning the compost (temperatures were maintained at >55 °C) [15].

The moisture content is monitored periodically to ensure smooth functioning and is decided from previous lab-scale investigations to pre-humidify the inlet gas-streams. The moisture content of the packed bed or humidity of the incoming stream should be maintained in the range of 40%–60% (*d/w*) for compost biofilters treating p-Xylene, toluene and other VOCs [12,15,21,25]. Jorio et al. [21] studied the removal of toluene from air stream by using biofilters and found higher affinity of microbial population growth for aromatics i.e., for toluene removal. Hydrophilic pollutants (*viz.* ethyl acetate and butyl acetate having higher Henry's constant) are usually degraded faster than hydrophobic ones due to the high gas-liquid mass transfer rates that makes them more bioavailable to the microbial community [14,22,25,46]. The water holding capacity of the media is an important parameter since water content is usually difficult to control [47]. The water holding capacity of compost is usually very high i.e., 138 ± 11% *dw* that ensures the continual presence of moisture required for microbial growth [40].

The gas flow rate and contact time or the EBRT is closely related. In general, greater the flow rate, lesser is the contact time and thus there is a reduced reaction time for the microbes to assimilate the pollutants. Therefore, to achieve higher REs, a greater EBRT is required [16]. The pH of the biofilm is yet another important parameter that has been observed in most biofilters to be maintained at a near neutral pH of 6.0–7.5 which ensures the survival and maintenance of most pollutant degrading bacteria. The lower pH conditions (<3.0, created by SOB) are observed to create an unsustainable environment for the growth of NH_3 and VOC degrading bacteria in biofilters that simultaneously remove several pollutants (H_2S, NH_3, VOCs) (Table 1). It is also harmful to the packing material as it gets degraded, which eventually increases the head loss resulting in reduced efficiency and increased operation cost [21]. To encounter this, traditionally biofilters use NaOH for pH control; however, many new biofilter configurations have started dividing the packed bed into acidic zones and neutral zones for the simultaneous removal of VOCs, H_2S, NH_3 and other pollutants by acidophilic and heterotrophic bacteria (which can survive between a pH of 1.5 and 6.0) [16,48]. One of the other techniques to avoid the acid production and accumulation is periodic back washing of the packing bed by water or using a sprinkler system [18]. For instance, a pilot study conducted for scrubbing odorous gases from a WWTP employed a biofilter system that produced leachate at the bottom of biofilter, after washing down H_2S, in the form of concentrated hydrogen sulfate that was collected and used for pH control of the biofilter. The collected hydrogen sulfate at the bottom of the reactor was used for the chemical conversion of pollutants like NH_3 by passing over the sulfate ions to further produce ammonium sulfate pellets which was later recovered [18].

2.2. Biotrickling filter

In the last 30 years, BTFs have been widely implemented in industries for the treatment and removal of waste gases like VOCs, H_2S, styrene and others including acidic vapors. The advantages that BTF offers to overcome the restrictions of the conventional biofilters include: (i) greater stability of operation, yielding higher performance, (ii) no requirement for pre-humidification of the waste gas, and (iii) capability of performing with higher H_2S concentration and higher flow rates [28,49]. One of the major advantages of using BTF is that lower EBRT can be achieved (up to 2.8 s) i.e., lesser contact between the biofilm and the pollutant, which is an important factor for operating bioreactors at an industrial scale that further holds a potential for attaining comparable RE with those of conventional physicochemical techniques for pollutant removal [33]. Like biofilter, BTF also uses a wide range of packing materials including ceramsite, propylene rings, and other synthetic materials (Table 1). The synthetic packing media in BTFs provide various practical advantages such as: (i) high surface area and high void fraction resulting in minimal gas residence time, better contact and distribution of gas and water; (ii) compact size due to higher surface area of bed; (iii) easier control of pH, temperature, moisture content and biofilm due to the existence of the free liquid phase and; (iv) long life of the packing media implying reduced replacement or reconditioning cycles [30]. The BTF using synthetic packing materials are inoculated usually with activated sludge from WWTPs (Table 1). The overall RE of the BTF system decreases with increased complexity in construction and operation due to changes between pure culture conditions in the pilot-scale/full-scale environment, competitive exclusion, nutrient availability and toxic effect of contaminants on the microbes present in the biofilm [26].

A model was developed for the simulation of an industrial BTF for styrene removal and was validated by a pilot-scale BTF in a fiber-reinforced facility in Spain [50]. The model was calibrated with laboratory data and accounted for key features required for the industrial set-up i.e., variable inlet loading conditions (5–23 g m^{-3} h^{-1}), EBRT of 31 s and intermittent irrigation of the packing material. Intermittent recycling of trickling water has been demonstrated to be beneficial for saving energy costs and raw material requirements [14,32]. Activated carbon (AC) filter is widely used in BTFs to regulate inlet loadings of industrial reactors and increase the REs [34]. VOC removal by BTF in a paint-spraying booth of a furniture industry was conducted with various irregular inlet loadings, i.e. under transient state conditions (5–72 g m^{-3} h^{-1}) and several shut-down periods [30]. A buffering system containing AC for VOC adsorption was installed before the BTF to regulate the strength of loadings provided to the reactor (EBRT = 10–40 s), which resulted in relatively better performance of the system [28,30].

Similar to a biofilter, the bacterial configuration of biofilm in a BTF is dependent on the type of pollutant treated by the system [29,30]. Yang et al. [29] reported that the bacterial community is co-dependent when H_2S is removed effectively (RE >90.0%) because *Proteobacteria* survived in the lower and middle layers of the packing bed, while *Firmicutes* favored the upper layer with higher humidity. The temperature during BTF operation plays an important role in improving the

EC. The application of thermophilic bioreactors (i.e. the bioreactors that operate at high temperatures, >55 °C) could likely eliminate clogging problem as the biomass growth is not very high (slow growth of thermophilic bacteria) [51]. Since biomass growth is controlled, the amount of organic acids produced on the packing media is minimal, thus reducing its wear and tear. During winters, the temperature has to be controlled and therefore increased inlet temperatures are provided to ensure continual microbial growth [28,29]. Thermophilic BTFs can be employed for high temperature pollutant emitting industries like tanneries and food processing industries [51]. Sempere et al. [34] observed a nine-fold increase in odor elimination capacity in a BTF at peak higher ambient temperatures. Increasing the inlet temperature in a BTF is achievable since there is continual supply of trickling water unlike biofilters that can dry up and subsequently reduces the RE [52].

2.3. Bioscrubbers and other bioreactors

Bioscrubbers are not as widely used as BF or BTF due to the high energy consumption and high cost that precedes their installation. A pilot-scale anaerobic bioscrubber (with <10% renewal of water owing to recirculation) in a flexographic painting facility displayed its capability of handling high VOC load up to 2000 $m^3\ m^{-2}\ h^{-1}$ with RE >95.0% (for inlet loading of 1126 ± 470 mg N m^{-3}) [37], despite various interruptions in inlet loading or shutting down periods during the weekends. Other types of bioreactors used at an industrial scale include trickle-bed bioreactors and combined bioreactor systems that include biological aeration zones [48]. In a compact trickle-bed bioreactor, with inlet concentrations of VOCs and H_2S as 240 and 660 ppm, respectively, an EC of 18 g $m^{-3}\ h^{-1}$ and RE of ~99.0% was achieved for H_2S. As observed during its operation, the advantages include low water consumption (<50 L d^{-1}), low operational cost and relatively safer process [36].

The techniques and instruments used for analyzing the pollutants present in the inlet and outlet of a bioreactor are similar for every configuration (i.e., biofilter, BTF, or bioscrubber). Sulfur in the form of H_2S and its degraded products are analyzed using Inductively Coupled Plasma (ICP) and the ones on the surface of the packed bed with Scanning Electron Microscopy and Energy-dispersive X-ray apparatus (SEM/EDX) [22]. Gas chromatography is employed for the detection of a variety of VOCs as it is the most reliable and cost-effective way of analysis [14,15,23,25,32]. Flame ionization detector (FID) and Olfactometer are used extensively to analyze VOCs and classifying odors. FISH (Fluorescence *in-situ* hybridization) technology is effectively applied for the detection of bacterial dynamics on the packing bed of the BTF [53]. An average of 80% species of VOC degrading bacteria including *Alphaproteobacteria* and *Firmicutes* were detected at a furniture painting facility in Spain [53].

3. Conversion of C1 gaseous compounds to liquid value-added products/fuels

The microbial conversion of gaseous substrates to several end-products dates back to early 1900s [54]. A gas mixture containing different proportions of CO, H_2 and CO_2, and other gases, such as nitrogen, CH_4, sulfur compounds and light hydrocarbons is called syngas. These are mainly produced from fossil sources such as coal, natural gas, petroleum residuals and biomass (non-food lignocellulosic materials, algae, residues from 2nd generation ethanol production, municipal solid waste etc.) through thermochemical processes [55]. Effluent gases from coal-fired plants, cement industry, etc. are another promising feedstock for syngas production. These gas mixtures are principally composed of CO_2 and CO and can be utilized directly as a carbon source to obtain value added chemicals via thermal, photochemical, biochemical and electrochemical processes or converted to syngas [56]. In contrast, off-gas from the steel industry contains a considerable amount of CO and H_2, which can be directly utilized as syngas [54]. Coal and petroleum gasification is commonly used for syngas production where natural gas is converted to syngas via steam reforming process. Among them, gasification is one of the oldest technologies on a commercial basis and majority of the syngas demand is met by coal gasification [57]. However, the utilization of coal as a feedstock has several drawbacks regarding its high ash content, resulting in the formation of huge amounts of solid residues [58]. Compared to coal, petroleum residues have high content of carbon; however, its gasification reactivity is lower, which requires high gasification temperatures to obtain syngas [59]. Moreover, syngas produced from petroleum residues contain significant amount of sulfur compounds that need to be removed during the cleanup process. As a renewable alternative to coal and petroleum residues, biomass can also be converted to syngas via gasification process. Over the last decades, there have been many attempts to develop and implement the biomass gasification processes to industrial scale [60,61]. Recent studies reported that biochar, which is a product of biomass pyrolysis, can reduce operation difficulties arisen from undesired properties of biomass such as heterogeneity, high moisture content, low fixed carbon content and low energy density etc. [62,63]. Besides, the utilization of biochar instead of biomass can reduce the formation of tar that leads to blockages and fouling in the equipment and low conversion efficiency [64]. Syngas composition and gasification efficiency varies depending on the feedstock type, gasification agent, gasifier design and process conditions such as temperature and the type of catalyst.

There are different promising carbon capture and utilization (CCU) technologies such as syngas fermentation, microbial electrosynthesis and methanotrophic fermentation that have gained considerable attention recently, a way to mitigate the C1 gas and convert them into value added chemical commodities. Pretreatment of gas for impurities and toxic chemicals such as SO_x, NO_x may be required if their limits exceed the tolerance level of the microbial community [65]. In the presence of residual oxygen, a system using anaerobic microorganisms particularly working with pure cultures, the exposure to oxygen could hamper the growth and overall productivity of the process. Techniques such as adsorption and membrane separation can be used to separate CO_2, CO and H_2 from the gas mixtures by maintaining the appropriate operating conditions [66,67].

3.1. Syngas fermentation technology

The conversion of syngas to ethanol [68], butanol [69], hexanol [69] and valuable acids; such as acetate and butyrate, by bioprocess technology using acetogenic carboxydotrophs is referred to as syngas fermentation [70] (Fig. 2). The acetogenic bacteria such as *Clostridium ljungdahlii* [71], *Clostridium ragsdalei* [72] and *Clostridium autoethanogenum* [73] can convert syngas into ethanol and acetic acid using the Wood-Ljungdahl pathway [70]. Syngas fermentation has been gaining attention due to the superiorities such as low operation cost, less sensitivity of biocatalysts to syngas impurities than chemical catalysts, high selectivity and production of specific high value-added chemicals. However, there are disadvantages such as mass transfer limitations of gaseous substrates and high production costs [74]. Besides, syngas can also be converted to valuable products such as 2,3-butanediol [75] and poly-3-hydroxybytuyrate [76].

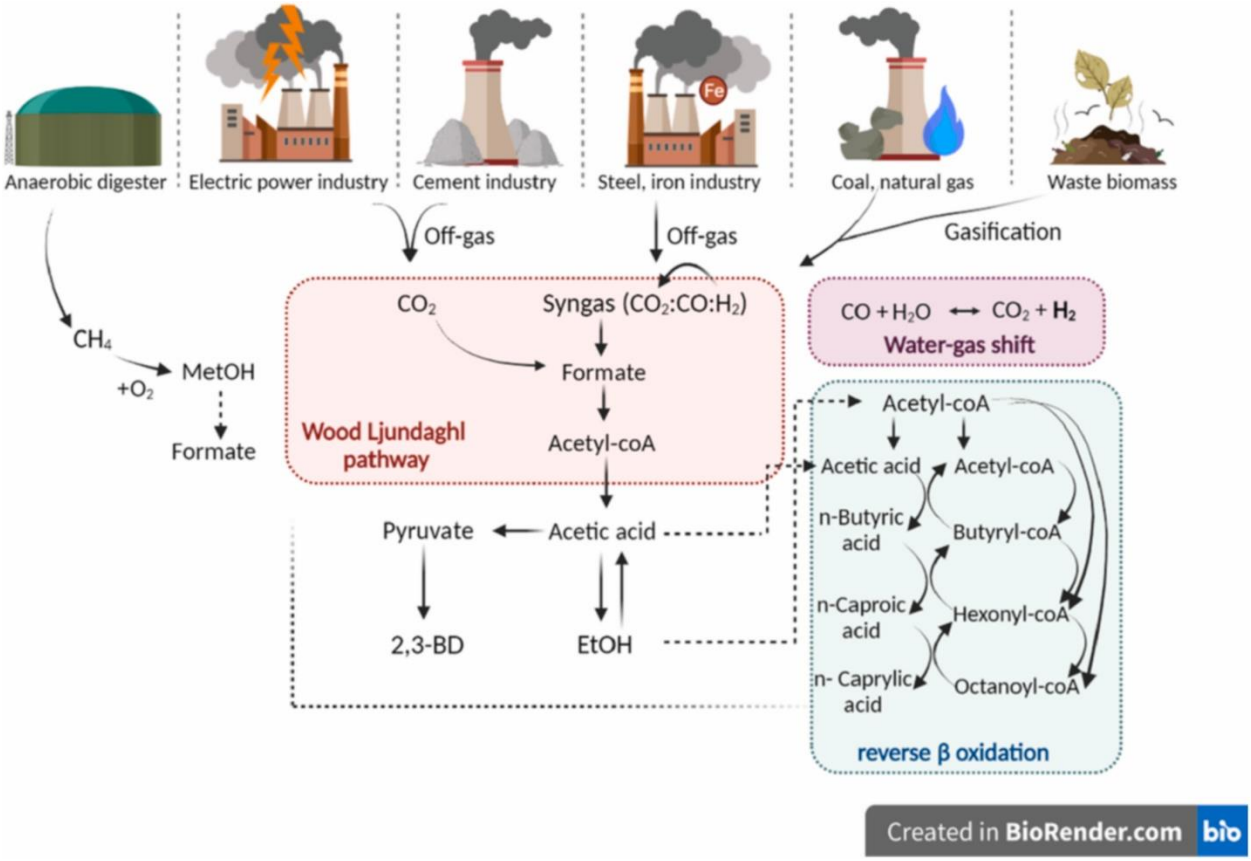


Fig. 2. A simplified pathway of C1 gas fermentation to value added products. Reverse β-oxidation pathway was adopted from [95].

The most critical parameter affecting the growth of acetogen and its metabolites production is the pH of the fermentation medium. Acetogens have two metabolic phases: acetogenic phase and solventogenic phase. During the acetogenic phase (at optimal pH), growth and acetic acid production occur simultaneously, and the conversion of produced acids to its corresponding alcohols happens during the solventogenic phase (at pH lower than the optimal growth pH) [77]. Therefore, by properly adjusting the fermentation pH, the product spectrum from syngas fermentation can be significantly narrowed to achieve the desired value-added products.

Another important approach to improve the process yields is by media optimization and using additives. Nutrients such as vitamins, minerals and trace metals in fermentation media improve the cell growth and fermentation productivity, but it causes an increase in the production cost [78,79]. This prompted the development of low-cost media such synthetic and defined medium without the morpholinoethane sulfonic acid buffer [80], corn steep liquor as a substitute for expensive yeast extract [81], cotton seed extract as a replacement for vitamins and minerals [82], malt and vegetable extract [83], and incorporation of biochar as a source of minerals, metals and as a pH buffering agent [84,85]. Moreover, incorporation of different types of nanoparticles (palladium on carbon and alumina, silica, carbon nanotubes, alumina, and iron oxides) to the fermentation medium has also been tested to reduce the limitation of gas-liquid mass transfer and increase the dissolution of CO, CO_2 and H_2 [86].

Since acetate and ethanol are the dominant end products of syngas fermentation, it is reasonable to combine syngas fermentation with chain elongation to produce more valuable biochemicals such as medium chain fatty acids (MCFAs) from C1 gases. Chain elongation is an anaerobic bioprocess to produce (MCFAs) by converting volatile fatty acids (VFAs) and an electron donor such as ethanol or lactate [87]. Ethanol is one of the most preferred electron donors utilized by mostly studied chain elongating bacteria, *Clostridium kluyveri*, by using reverse β-Oxidation pathway [88] (Eq. (1)).

$$5C_xH_{2x-1}O_2^- + 6C_2H_6O \rightarrow 5C_{x+2}H_{2x+3}O_2^- + C_2H_3O_2^- + 4H_2O + H^+ + 2H_2 \quad (1)$$

In comparison to ethanol, the recovery of MCFAs (caproic, heptanoic, caprylic acid) is easier because of its low solubility in aqueous phase. The market value is higher than ethanol because of its widespread application in areas such as pharmaceuticals, chemical additives, and precursors of fuels like jet fuels and biodiesels [89,90]. Table 2 summarizes the chain elongation studies combined with syngas fermentation. Furthermore, valuable metabolites are produced through the integration of syngas fermentation with other bioprocesses including malic acid, C16 and C18 lipids [91,92]. As seen from Table 2, for C1 substrates, acetogens (*C. autoethanogenum, C. ljungdahlii),* chain elongators (*C. kluyveri)* or a co-culture containing acetogens and chain elongators have been used in bioreactors that aimed at carrying out syngas fermentation and/or direct chain elongation studies (Table 2).

Table 2. MCFAs production through combined C1 gas fermentation and chain elongation process.

Substrate	Microorganism		Reactor operation mode	n-butyrate (mmol L−1)	MCFAs Concentration (mmol L−1)			Reference
					n-caproate	n-heptylate	n-caprylate	
Acetate + CO	Syngas fermentation (*C. autoethanogenum*)	Chain elongation (*C. kluvyeri*)	Continuous	5.3−14	1.8−5.5	–	–	[93]
Acetate + syngas	Syngas fermentation (*C. ljungdahlii*)	Chain elongation (*C. kluvyeri*)	Continuous	30.7	11	–	<0.04	[94]
Acetate + ethanol	Syngas fermentation effluent	Chain elongation (*C. kluvyeri*)	Continuous	9.4 ± 0.7[a]	39.9 ± 0.9[a]	–	1.4 ± 0.2[a]	[95]
Acetate + ethanol	Syngas fermentation (*C. ljungdahlii*)	Chain elongation (mixed culture)	Continuous (anaerobic filter)	220.18	8.6	–	–	[96]
CO_2 + H_2	Direct chain elongation (mixed culture)		Hollow fiber membrane	20.4	8.4	–	2.9	[97]
CO	Direct chain elongation (mixed culture)		Semi-continuous	8.1	1.9	1.6	1.0	[87]
CO + ethanol	Direct chain elongation (mixed culture)		Batch	8 ± 0.4	4 ± 0.8	≤1.2	–	[98]
Ethanol + acetate	Chain elongation (*C. kluyveri*)		Batch	<11.3	72.49	–	–	[99]
Acetate/butyrate/ethanol	Chain elongation (*C. kluyveri*)		Batch	–	167	–	–	[90]
CO + H_2 + succinate	Co-culture multispecies GEMs of *C. autoethanogenum* and *C. kluyveri*		Continuous		0.59[b]	–	–	[100]
CO/butyrate	Mixed culture (mainly *Eubacterium limosum*)		Batch	–	7.7	–	–	[101]

Note: GEM – Genome-scale, constraint-based metabolic models; [a] – mmol l^{-1} d^{-1}; [b] – mmol l^{-1} h^{-1}

Bioreactor configuration is one of the most important parameters for the improvement of the syngas fermentation process yields. The most commonly used reactor types for lab scale and industrial scale applications are the continuous stirred tank reactor (CSTR) and batch reactor [102]. The mass transfer limitations of the CSTR can be improved using different types of impellers such as rushton impeller, fluid foil impeller, and concave turbines [103] or by using different gas delivery systems [104]. To overcome the gas transfer limitations during ethanol and other added value compounds production, several reactor configurations such as gas to atomized liquid contactor [105], horizontal rotating packed bed reactor [106], trickle-bed reactor [107], and hollow fiber membrane (HFM) reactors [108] were used. Anggraini et al. [108] demonstrated a threefold increase in ethanol production when a CSTR was combined with a HMF. The use of the HMF has resulted in a significant decrease in the mass transfer barrier of syngas and a substantial increase in ethanol production was observed. Two-stage systems that separate the acetogenic and solventogenic phases by using different reactors is another effective approach to enhance the process yields [104].

Another promising way for the mitigation of CO, one of the syngas components, is by the water gas shift reaction [109]. Carboxydotrophic hydrogenogenic microorganisms can convert CO as the source of electrons and carbon into molecular hydrogen (H_2) using carbon monoxide dehydrogenase and hydrogenase [110]. The involvement of water in this reaction results in 1 mol of CO_2 and 1 mol of H_2 [111]. The H_2 produced can be used as an electron source for chemicals and fuel productions through chain elongation and bioelectrochemical processes. Table 3 summarizes the types of reactors recently used for liquid biofuel and hydrogen production. The following types of bioreactors have been reported in the literature: Batch, CSTR, bulk-gas-to-atomized-liquid reactor, horizontal rotating packed bed biofilm reactor, trickle-bed reactor (TBR), combination of CSTR and bubble column reactor, hollow fiber membrane biofilm reactor (HfMBR), monolithic biofilm reactor, bubble free membrane reactor, and gas-lift reactor. Concerning the microorganisms, *Alkalibaculum bacchi*, *Carboxydothermus hydrogenoformans*, *Clostridium aceticum*, *Clostridium butyricum*, *Clostridium autoethanogenum*, *Clostridium carboxidivorans*, *Clostridium ljungdahlii*, *Clostridium propionicum*, *Clostridium ragsdalei*, *Methylosinus trichosporium* and *Rhodopseudomonas palustris* have been frequently reported in studies involving C1 gases (Table 3).

Table 3. Various reactor studies performance using C1 gases under varying process conditions and the observed limitations.

Reactor type	Feedstock	Gasification conditions	Syngas composition	Operating conditions	Microorganism	Product	Limitations	References
Batch	Straw	Company: Bioliq® process - High-pressure entrained flow - Gasifying agent: O_2 - Temperature: >1200 °C - Pressure: up to 80 bar	29% CO 3% CO_2 28% H_2	Mesophilic, 250 mL	*Clostridium ljungdahlii*	Acetate:11.2 g L^{-1} Ethanol: 2.8 g L^{-1}	Possible impurities HCN (0.91 ppm), NH_3 (150 ppm), H_2S (54.1 ppb), COS (12.3 ppb)	[112]
TFB (tar free bioreactor) followed by fed batch system	Forest residue-charcoal mixture	- Downdraft fixed bed type air - Gasifying agent: air - Temperature: 800–1050 °C	23% CO 8% CO_2 1% CH_4 46% N_2 13% H_2	Mesophilic, 500 mL	*Clostridium butyricum*	Ethanol: 0.07–1.38 g L^{-1}	Possible impurities and changing composition of syngas	[113]
Batch	Commercial syngas	Company: White Martins Praxair Inc	25% CO 44% H_2 10% CO_2 10% N_2 11% CH_4	Mesophilic, 50 mL	*Clostridium carboxidivorans*	Ethanol: 2.28 g L^{-1} Butanol: 0.74 g L^{-1}	Possible impurities and changing composition of syngas	[114]
Continuous and batch	Switchgrass	- Fluidized bed - Gasifying agent: air - Temperature: 750–800 °C	16.5% CO 15.5% CO_2 5% H_2 56% N_2 4.5% CH_4	Mesophilic, 3 L CSTR, 250 mL batch	*Clostridium carboxidivorans* P7	Ethanol: 0.2–0.7 g L^{-1}	Possible impurities C_2H_2 (0.1%) C_2H_6 (0.35%) C_2H_4 (1.4%) N_2O (150 ppm)	[115]
CSTR	Artificial syngas	Company: Stillwater Steel	38% CO 28.5% H_2 28.5% CO_2 5% N_2	Mesophilic, 3 L fermenter	*Alkalibaculum bacchi* CP15 and *Clostridium propionicum*	Butanol: 0.4 g L^{-1} Propanol: 0.98 g L^{-1} Ethanol: 0.09 g L^{-1}	Possible impurities and changing composition of syngas	[116]
CSTR and batch	Commercial syngas		20% CO 15% CO_2 5% H_2 60% N_2	Mesophilic, 7.5 L CSTR, 250 mL batch	*Clostridium* P11	Ethanol: 6.1 g L^{-1}	Possible impurities and changing composition of syngas	[81]
Bulk-gas-to-atomized-liquid reactor (BGAL)	Artificial syngas		12.5% H_2 37.5% CO_2 50% CO	Mesophilic, 10 L CSTR and BGAL reactor	*Clostridium carboxidivorans* P7	Ethanol: 4.38 g L^{-1}	High equipment costs	[105]

Reactor type	Feedstock	Gasification conditions	Syngas composition	Operating conditions	Microorganism	Product	Limitations	References
Horizontal rotating packed bed biofilm reactor	Artificial syngas		20% CO 5% H_2 15% CO_2 60% N^2	Mesophilic, 3.3 L	*Clostridium carboxidivorans* P7	Ethanol: 7.0 g L^{-1}	Mass transfer limitations into the medium from headspace	[106]
A trickle-bed reactor (TBR)	Artificial syngas		38% CO 5% N_2 28.5% CO_2 28.5% H_2	Mesophilic, 500 mL	*Clostridium ragsdalei*	Ethanol: 13.2 g L^{-1} Acetate: 4.3 g L^{-1}	Working with mimicked coal gasification gas. The real gas results will be lower.	[117]
Two-stage system (CSTR + CSTR)	Artificial syngas		30% CO_2 40% CO 30% H_2	Mesophilic, 3 L fermenters	*Clostridium ragsdalei*	Ethanol: 14.74 g g^{-1} cells Acetate: 17.51 g g^{-1} cells	CSTR systems need extra HFM systems for cell recycling	[72]
Two-stage system (CSTR + bubble column reactor)	Artificial syngas		60% CO 35% H_2, 5% CO_2	Mesophilic, 1 L CSTR, 4 L BCR	*Clostridium ljungdahlii*	Ethanol: 19.73 g L^{-1} Acetate: 8.55 g L^{-1}	Working with mimicked pyrolysis gas. The real gas results will be lower.	[118]
Batch system	Artificial syngas		70% CO 20% H_2 10% CO_2	Mesophilic, 50 mL	*Clostridium carboxidivorans*	Butanol: >1.0 g L^{-1} Hexanol: up to 1.0 g L^{-1} Ethanol: >3.0 g L^{-1}	System needs to be improved for commercialization	[79]
Hollow fiber membrane biofilm reactor (HfMBR)	Artificial syngas		40% CO 60% H_2	Mesophilic and thermophilic 390 mL	*Mixed Culture*	Acetate: 4.22 g L^{-1} Butyrate: 1.35 g L^{-1} Caproate : 0.88 g L^{-1} Caprylate: 0.52 g L^{-1}	System needs to be improved for commercialization	[119]
Stirred tank retrofitted gas-lift bioreactor	Artificial syngas		3% CO_2 12% H_2 10% N_2	Mesophilic, 2.0 L stirred tank and 2.5 L retrofitted gas-lift bioreactor	*Clostridium aceticum* or *Clostridium carboxidivorans*	Acetate: 180 mM Ethanol: 6 mM	High equipment costs	[120]

Reactor type	Feedstock	Gasification conditions	Syngas composition	Operating conditions	Microorganism	Product	Limitations	References
Continuous stirred reactor	Artificial gas		50% CH_4 50% O_2	Mesophilic, 5 L reactor	*Methylosinus trichosporium* NCIB 11131 (OB3b)	Methanol: 1.19 g L^{-1}	MDH inhibitor usage to prevent over oxidation of methanol	[121]
Continuous bubble free membrane reactor	Artificial gas		50% CH_4 50% O_2	Mesophilic, 300 mL	*Methylosinus trichosporium* OB3b	Methanol: 0.95 g L^{-1}	MDH inhibitor usage to prevent over oxidation of methanol	[122]
Gas-lift with recirculation	Artificial gas		100% CO	Continuous, thermophilic, 35 L	*Carboxydothermus hydrogenoformans* DSM6008	96.7% mol H_2 mol^{-1} CO	Liquid-gas mass transfer was limited under a specific gas recirculation and feed ratio Low cell density was observed	[123]
Pressurized hollow fiber membrane	Artificial gas		100% CO	Continuous, thermophilic, immobilized, 160 mL	*Carboxydothermus hydrogenoformans* DSM6008	92.0% mol H_2 mol^{-1} CO	CO transfer rate was limited because of the fouling and aging of the membrane Pressurized systems (over 2 atm) are needed for higher conversion rates	[124]
CSTR	Artificial gas		100% CO (and sucrose)	Mesophilic, two-stage: aerobic-chemoheterotrophic/anaerobic H_2 production, 5 L	*Rhodopseudomonas palustris* P4	41 mmol H_2 L^{-1} h^{-1}	Gas liquid mass transfer was limited by the CSTR system	[125]

Reactor type	Feedstock	Gasification conditions	Syngas composition	Operating conditions	Microorganism	Product	Limitations	References
Reverse membrane bioreactor	Artificial syngas		55% CO 20% H_2 10% CO_2	Thermophilic, immobilized two stage	Anaerobic mixed culture	1.92 mmol CH_4	Repeated batch productions required shorter retention times for enhanced co-digestion and production	[126]
Monolithic biofilm reactor	Artificial syngas		20% CO 5% H_2 15% CO_2 60% N_2	Mesophilic, immobilized, 8 L	*Clostridium carboxidivorans* P7 ATCC BAA-624	Ethanol: 4.89 g L^{-1} Acetate: 3.05 g L^{-1}	Productivities and cell density were decreased with the highest dilution rates	[127]

3.2. Bioelectrochemical technology

The bioelectrochemical system (BES) uses microbes or enzymes to perform oxidation, reduction, or both reactions, and has been widely researched for its potential to produce electricity by oxidizing the organic matter present in wastewater using electrochemically active bacteria in microbial fuel cells (MFC) [128]. Recently, Nevin et al. [129] proposed an alternative bioelectrochemical technique relying on the microbial ability to use electrochemically supplied electrons via a solid electrode in the cathode compartment to drive the conversion of CO_2 into acetic acid, which was found to be the prevalent end product from the process in most of the reported studies. However, a variety of high market value compounds, among others, C4 and C6 carboxylic acids and their corresponding alcohols can also be produced depending on the biocatalyst and operating conditions (Table 4) [130,131]. This microbe mediated electrochemical CO_2 or organic substrates conversion technique of generating extracellular multi-carbon organics is called microbial electrosynthesis (MES). One way of handling the external power supply needed to regulate the redox potential in the MES system is by coupling with renewable electricity supply sources such as solar and wind energy [132]. MES reactor consists of an anodic and cathodic compartment separated by a proton exchange membrane in which CO_2 reduction is catalyzed by the microbes present in the cathode side (Fig. 3). The anodic oxygen evolution reaction that generates protons and electrons through water splitting has been widely applied in MES, but this requires a considerable overpotential. Therefore, several other strategies have been proposed, for example utilizing a bioanode capable of oxidation of organics present in wastewater [133,134]. Anodes made from carbonaceous materials or modified by depositing noble metals such as Ti and Pt have been commonly used in MES [135].

One of the main factors influencing the MES performance is the selection of an appropriate cathode. Cathode material with higher surface area to volume ratio, excellent electrical conductivity, and biocompatibility are of paramount importance for biofilm development and electron transfer [133,147]. In this regard, research has been focused on developing new cathode materials, functional modification of the electrode surface, and binding of nanowires or nanoparticles to reduce the activation energy of electron transfer [132,144]. In most of the MES studied configurations, CO_2 was fed by sparging and electrodes as submerged [139]. However, low solubility of CO_2 causes low availability of substrate for electroautotrophs. For example, it has been reported that the mass transfer limitation is overcome by using a gas diffusion electrode (GDE) as a cathode, and it also helps to immediately adsorb CO_2 to its surface, thereby ensuring sufficient substrate availability [135]. An alternative way to improve the mass transfer is by reducing the bubble size of CO_2. This can be done by delivering CO_2 using a porous hollow fiber membrane electrode [148]. Examples of modification of cathode that have shown improvement in acetate production from CO_2 includes charging with chitosan [149], reduced graphene oxide (rGO) [150], and multi-walled carbon nanotube (MWCNT) on reticulated vitreous carbon (RVC) [144].

Table 4. An overview of different studies performed with MES using C1 gases.

Microbial inoculum	Gaseous substrate (%)	Cathode material	E cathode (V vs SHE)	Liquid end products	Reactor type	Max. acetate production	Acetate production rate	CE for acetate (%)	References
Anaerobic digester sludge	$CO:CO_2:N_2$ (40:10:50)	Graphite felt	−1.1 V (vs. Ag/AgCl)	Acetate, butyrate, propionate, isobutyrate and isovalerate	Two-chambered H-type	8470 ± 150 mg L^{-1}	344 mg L^{-1} d^{-1}	>200	[136]
Mixed sediment	$CO:CO_2:N_2$ (40:10:50)	Carbon felt	−1.1 V (vs. Ag/AgCl)	Acetate, butyrate, propionate, isobutyrate and isovalerate	Two-chambered H-type	6.89 g L^{-1}	0.71 g L^{-1} d^{-1}	184.4	[137]
Activated sludge	$CO:CO_2$ (50:50)	Carbon felt	−1.1 V (vs. Ag/AgCl)	C2–C6 carboxylates and alcohol	Two-chambered H-type	5.47 ± 0.10 g L^{-1}	ng	ng	[138]
Enriched mixed culture	$CO_2:N_2$ (80:20)	Graphite felts with a graphite stick sandwiched	−0.9 to −1 V (vs. Ag/AgCl)	Acetate, ethanol and butyrate	H-type glass reactor	10 g L^{-1}	~400 mg L^{-1} d^{-1}	48.0	[139]
Enriched mixed culture	$CO_2:N_2$ (10:90)	Carbon felt	−1.14 ± 0.04	Acetate, formate, propionate and ethanol	Three-compartment cell	13.5 g L^{-1}	0.64 g L^{-1} d^{-1}	61.0	[140]
Enriched mixed culture	$NaHCO_3$	MWCNT-RVC	−1.1	Acetate	Two-chamber system	1330 mg L^{-1}	ng	84.0 ± 2.0	[141]
Enriched acetogenic microbiome	only CO_2	RVC foam	−1.1 to −1.3	Acetate, formate, propionate and butyrate	Modular, continuous	3.6 g L^{-1}	0.78 g L^{-1} h^{-1}	35.0	[142]

Microbial inoculum	Gaseous substrate (%)	Cathode material	E cathode (V vs SHE)	Liquid end products	Reactor type	Max. acetate production	Acetate production rate	CE for acetate (%)	References
Enriched mixed culture	CO_2:N_2 (30:70)	Carbon felt	−1.02 ± 0.01	Acetate, butyrate and caproate	Flat-plate reactor, continuous	Acetate: 17.5 g L^{-1} Butyrate: 9.3 g L^{-1} Caproate: 3.1 g L^{-1}	Acetate: 9.8 g L^{-1} d^{-1} Butyrate: 5.7 g L^{-1} d^{-1} Caproate: 2 g L^{-1} d^{-1}	69.8 ± 2.8 (all organics)	[131]
Enriched acetogenic MES culture	CO_2:N_2 (30:70), or $NaHCO_3$, or only CO_2	Graphite felt	−0.85	Acetate, butyrate and caproate	Flat-plate reactor, continuous	Acetate: 8.2 ± 0.4 g L^{-1} Butyrate: 2.9 ± 0.3 g L^{-1} Caproate: 1.1 ± 0.3 g L^{-1}	Acetate: 11.55 ± 0.15 g L^{-1} d^{-1} Butyrate: 5.0 ± 0.9 g L^{-1} d^{-1} Caproate: 1.5 ± 0.5 g L^{-1} d^{-1}	91.0 ± 9.0 (all organics)	[143]
Enriched granular sludge	CO_2:N_2 (20:80)	VITO-CoRE® gas diffusion electrode	−1.1 to −1.3 V (vs. Ag/AgCl)	Acetate, butyrate and ethanol	Double-chamber MES	2.89 g L^{-1}	61 mg L^{-1} d^{-1}	35.0	[135]
Enriched carboxydotrophic mixed culture	only CO_2	Carbon cloth	−0.8	Acetate, butyrate, ethanol and butanol	Two-chambered H-type BES	1.2 g L^{-1}	63.16 mg L^{-1} d^{-1}	38.9	[130]
Pond sediments and wastewater treatment plant sludge	$NaHCO_3$	NanoWeb reticulated vitreous carbon	−0.85	Acetate	Two-chamber system	1.65 g L^{-1}	[a]1.3 mM cm^{-2} d^{-1}	70.0 ± 11.0	[144]
Sporomusa ovate	CO_2:N_2 (80:20)	Reduced graphene oxide paper	−0.69	Acetate	Dual chamber system	0.77 g L^{-1}	[a]168.5 ± 22.4 mmol m^{-2} d^{-1}	90.7 ± 9.3	[145]
Enriched culture (lab-scale anode and algae UASB sludge)	N_2:CO_2 (90:10)	Carbon felt	−1.28 ± 0.03	Acetate, formate, propionate and butyrate	Custom-made glass reactors	1.5 ± 0.2 g L^{-1}	0.05 ± 0.001 g L^{-1} d^{-1}	44.0 ± 5.0	[146]

Note: [a] – based on projected surface area; ng – not given; MWCNT – Multiwalled carbon nanotubes; RVC – Reticulated vitreous carbon; UASB – upflow anaerobic sludge blanket.

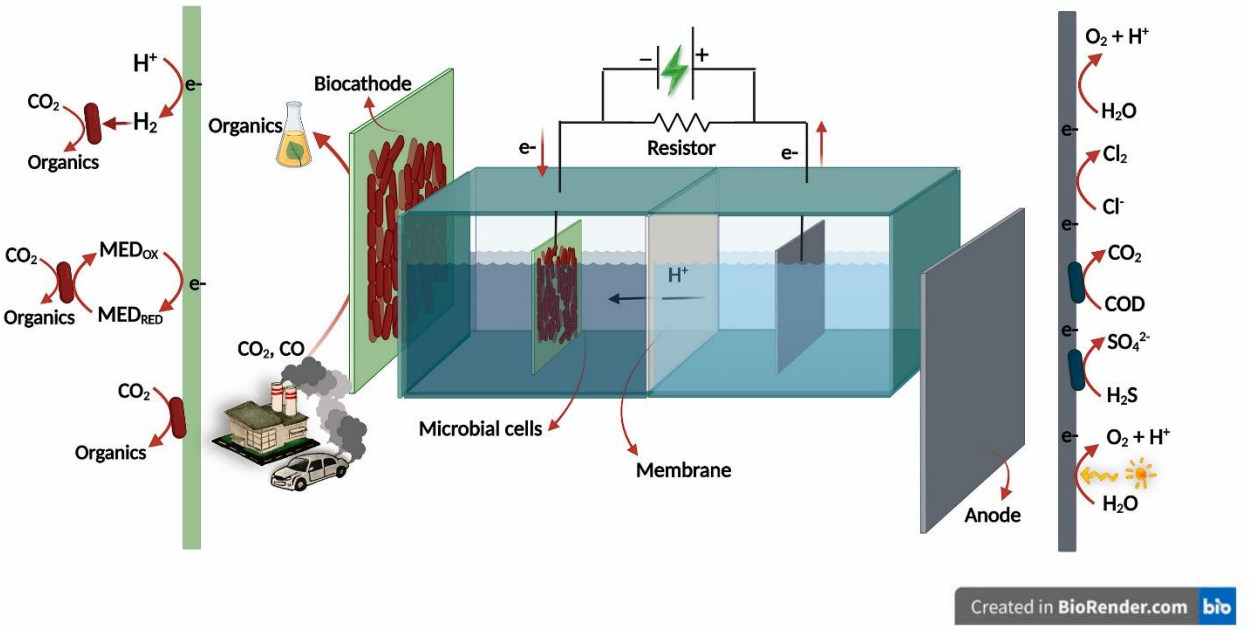


Fig. 3. Schematic of a microbial electrosynthesis (MES) system for treating C1 gases.

Acetogenic electroautotrophs responsible for CO_2 conversion either in pure and mixed consortium has been employed in MES either attached to the cathode as biofilm or present as planktonic cells; however, having a thick biofilm resulted in higher production rates of organics. Pure cultures of acetogenic bacteria representing this group belong to the genera *Sporomusa* and *Clostridium* [151]. In the enriched MES mixed culture, the presence of *Acetobacterium* spp, *Trichococcus* spp. and *Clostridium* spp were predominant [152,153]. Two potential extracellular electron transfer (EET) mechanisms have been proposed/discussed that facilitate the transfer of reducing power from the cathode to the microorganism, encompassing direct electron transfer (DET) and mediated electron transfer (MET) mechanisms. In direct transfer mechanism, the contact between the electrode surface and microbial cells/biofilm is associated with the cells producing conductive appendages, also called nanowires (pili), and/or outer membrane-bound c-type cytochrome proteins [154,155]. On the other hand, MET mechanism is mediated by self-produced electron shuttle molecules like formate, pyocyanin, flavin, etc., or by artificially supplemented electron shuttle molecules like quinone and viologens [137,156]. Electrochemically generated or biologically induced H_2 molecule has also been reported to mediate electron transfer in MES system [153]. Thus, thermodynamic barriers can be overcome by providing additional cathodic potential, which in turn plays a key role in the electron transfer mechanism.

In order to overcome the limitation of reducing equivalent while fermenting CO in conventional fermentation, BES has been proposed but not extensively examined [157]. In one such study, using an applied potential of −1.1 V vs. Ag/AgCl in a H type reactor using a mixed culture, acetic acid was electrosynthesized from syngas of 50% CO as the main product along with few

amounts of other VFAs [136]. Several parameters such as anodic pH, ion exchange membrane, applied potential and electron mediators have significant influence on the acetate and other VFA productions both in CO/CO_2 bioelectrosynthesis [137].

Reactor configuration that has been extensively reported in the literature is the H type MES system. However, stacked types either in tubular and flat form (each cell connected in parallel or in series) is easy to scale up and maintain since each module can work as separate cells [158]. The product spectrum from MES has been widening in recent years to butyric acid and caproic acid by properly adjusting the operational parameters such as pH, hydrogen partial pressure, fraction of CO present in the gas mixture [138,158]. Ethanol is considered as the electron donor for chain elongation; however, it is speculated that cathode can also provide electrons in MES [143]. Like in conventional syngas fermentation, the possibility of triggering solventogenesis occurs by lowering to more acidic pH, resulting in the conversion of acids to corresponding alcohols [68].

3.3. Methanotrophic fermentation technology

Methanol is an important liquid fuel additive, an important raw material for biodiesel production, and an important solvent in many chemical industries [159]. Methanol has greater energy density (15.6 MJ L^{-1} for methanol and 36.6×10^{-3} MJ L^{-1} for CH_4) which makes it a better energy carrier than CH_4.

Methanol production from CH_4 is a two-step process which requires high temperature (900 °C) and high pressure (3 MPa) [160]. Since this two-step process is not cost efficient and energy intensive, more specific processes such as microplasma technology or bioprocesses is gaining importance. This diverse group of methanotrophic bacteria converts CH_4 into methanol and are classified into three types I, II and X based on their characteristics [161]. Initial oxidation of CH_4 to methanol is catalyzed by methane monooxygenase (MMO) enzymes. Methanol can be oxidized to formaldehyde by methanol dehydrogenase (MDH) and formaldehyde can be oxidized to formate by formaldehyde dehydrogenase (FADH). Two different pathways include the routes for assimilation of formaldehyde into biomass, ribulose monophosphate (RuMP) pathway, or the serine pathway (Fig. 4).

Research on the production of methanol from CH_4 has focused on the use of MDH inhibitors or the isolation of new strains. The inhibition of MDH enzyme using inhibitors such as salts [162], phosphate, EDTA [163], CO_2 [164] and cyclopropane derived inhibitors [165,166] result in increased methanol production. Other studies reported in literature are generally based on batch cultivation and the use of *Methylosinus trichosporium* OB3b for methanol production. Sheets et al. [167] isolated CH_4 oxidizing bacteria from a solid-state anaerobic digester from genus *Methylocaldum* and reported 0.43 g L^{-1} methanol production from biogas. Methanol production from 0.5 g L^{-1} to 1.1 g L^{-1} was reported in different studies using *Methylosinus trichosporium* OB3b with CSTR [121] or membrane reactors [122] (Table 3).

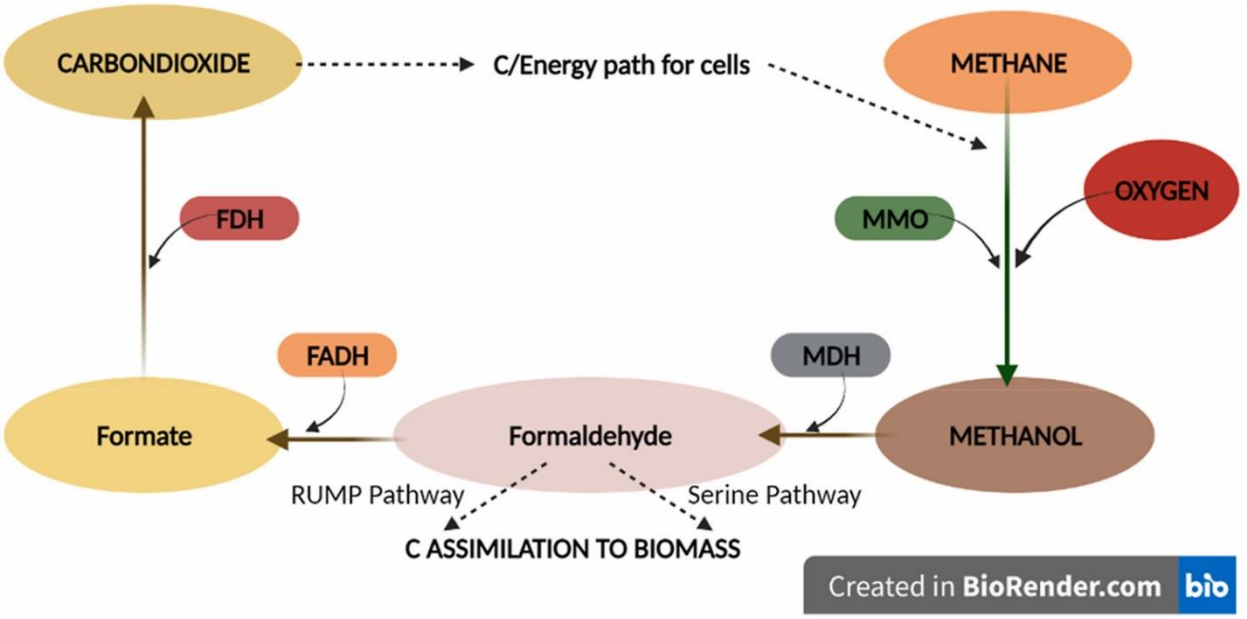


Fig. 4. Simplified pathway for methanol production from CH_4. MMO: methane monooxygenase; MDH: methanol dehydrogenase; FADH: formaldehyde dehydrogenase. Adapted from [160].

3.4. Product recovery

Due to the production of low-concentration products, product recovery is one of the most challenging steps in conventional/electricity driven CO/CO_2 fermentation processes. Acetic acid is the main product, although its production rate is considerably lower than those obtained through conventional sugar fermentation. In order to alleviate the product inhibition, in situ extraction techniques are suggested. pH is one of the important factors that affects the separation and fermentation process as mildly acidic conditions required for techniques such as membrane-based liquid–liquid extraction system (pertraction) could pose inhibition for the bacterium that functions at neutral pH [95]. Alternative VFA separation technology is electrodialysis, which has been tested for syngas fermentation and MES technologies [168].

Distillation following the dehydration step with molecular sieve is the most common technique to obtain ethanol with high purity. However, since the ethanol concentration is very low in the syngas fermentation broth, distillation is not economically viable due to the requirement of high-energy input. Pervaporation is another separation technique based on not only vaporization but also diffusivity and solubility of the products formed. Pervaporation can be implemented to the fermentation process for in-line separation of ethanol, which results in high conversion efficiency. From an economical point of view, hybrid separation processes such as membrane distillation, and vapor stripping-membrane have also been gaining attention [169,170]. Vapor stripping membrane processes are equipped with a vapor stripping column to recover ethanol from the feed liquid and they are used for ethanol recovery from the fermentation medium. In this system, an ethanol rich overhead vapor is compressed and then fed to a vapor permeation

system. The vapor permeation membrane system is a water selective membrane system to produce a retentate of ethanol rich product and water rich permeate. The stripping vapor in the column is formed by returning the permeate vapor from the membrane to the stripping column using either steam or reboiler heat [169]. Membrane distillation systems are an alternative for the mechanical vapor compression system. Recently more efficient membrane distillation systems consisting of a combination of distillation, vapor compression and membrane separation units have been developed. The overhead vapor from the distillation column is separated by compressing using a membrane vapor permeation unit. Thereafter, the enriched water is returned directly into the distillation column. Using this type of system, the ethanol product stream and the liquid reflux stream will be split [170].

3.5. Commercialization

Due to the increasing energy demand as a result of global industrialization, there has been a tremendous increase in CO_2 emissions since 1900 by almost 7.7 times, eventually reaching a final amount of 30.5 Gt by April 2020 (IEA, 2020: IEA, 2020. Chart: Global energy-related CO_2 emissions 1900–2020 (Available at: https://www.iea.org/data-and-statistics/charts/global-energy-related-co2-emissions-1900-2020). Under the Paris Climate Agreement, countries projected to mitigate their CO_2 emissions by 2020. Vast number of technologies have been proposed by both academia and industry to reach this goal. Accordingly, the pioneer company to commercialize the syngas fermentation is LanzaTech which was founded in New Zealand and currently has the headquarters in the US (LanzaTech, US, Accessed January 02, 2021, https://www.lanzatech.com). The corporation owns over 100 granted patents since 2009 (JUSTIA PATENTS, Accessed January 02, 2021, https://patents.justia.com/assignee/lanzatech-new-zealand-limited) regarding the C1 gas fermentation (including syngas and methanol) into 30 different products such as ethanol and other alcohols, ethylene, 2,3-butanediol, lactic acid, acetic acid, H_2, lipids, isobutylene, esters; fermentation media composition; carbon capture during fermentation; various bioreactor applications including coupled electrolysis, multi-stage processes and enhancement of the operational parameters; recombinant microorganisms for manipulated pathways and improved yields; maintenance of culture viability and gas dissolution in the liquid. The company introduced the term CarbonSmart™ for a circular economy. Under this context, continuous bioethanol production for 150 days was reported to reduce 100,000 tons of CO_2 in their first large scale commercial plant in China. Furthermore, due to syngas being the waste stream of steel industries, collaborations with the largest steel manufacturers in Asia and in Europe, Baosteel (China) and Arcelor Mittal (Belgium), respectively, were initiated. Both companies announced their successful applications in waste gas to bioethanol pre-commercialized plants. Accordingly, the EU funded Carbalyst® project from Belgium targets 80 million liters of bioethanol production. Another partnership of LanzaTech with Indian Oil to produce omega-3-fatty acids is projected to reach commercial scale by 2030. Another recent collaboration announcement in October 2020 with Total and L'Oréal introduced a multi-step

production of bioethanol driven biobased packaging. In September 2020, another up to date integration with Mangalore Refinery and Petrochemicals Limited (India) was publicly announced, explaining the integrated process with the conversion of agricultural wastes into biochar and syngas by Ankur Scientific's (India) gasification technology. Apart from LanzaTech, Coskata, Inc (US) is another company that is assigned several numbers of patents for the conversion of C1 gases via fermentation (JUSTIA PATENTS, Accessed January 02, 2021, https://patents.justia.com/assignee/coskata-inc). However, the company ended their business by 2015 [171]. Similarly, the US based INEOS New and Planet BioEnergy is also another joint company that shut down its syngas driven bioethanol production plant in 2016 due to the problems occurred as a result of the impurities found in the gaseous substrate [172]. So far, there are no reports of pilot and commercial MES plants. Useful information and guidelines for scaling up MES can be obtained from other scaled-up BES (such as microbial fuel cells for wastewater treatment) [173].

4. Resource recovery from the conversion of VOCs and odorous compounds containing waste gases

VOCs and odorous compounds containing waste gases are among the most important industrial pollutants in terms of being harmful to public health and causing environmental nuisance. The emission of VOCs from industrial sources are commonly benzene, toluene, xylenes, methanol, dichloromethane, dichlorobenzene, chloroform, tetrachloro ethylene and tricloro ethylene [174] while odors include sulfur compounds, e.g., H_2S, mercaptans, dimethyl sulfide, and carbon disulfide; nitrogen compounds, e.g., NH_3 and amines [175,176].

Biological processes have been continuously developed to eliminate VOCs and odorous compounds from the polluted gas stream [[177], [178], [179]]. The bioreactors for the treatment of VOCs and odorous compounds have been mostly conducted under aerobic conditions which rely on purging air into the systems and the end-products consist of the cleaned gas stream, CO_2, and H_2O. For example, the VOCs emitted from petrochemical industries commonly have a low concentration of pollutants and the treated gas is preferable to be reused for inertization process in the absence of O_2 [180,181]. Similar to some biotechnologies for biogas cleaning, the presence of trace amounts of O_2 is problematic for downstream applications.

The anoxic/anaerobic bioreactors are promising technologies for resource recovery, e.g., VFAs and elemental sulfur (S^0) from the degradation of VOCs and odorous sulfur compounds, respectively (Fig. 5). Furthermore, the systems are allowed for simultaneous treatment processes with other pollutants, like nitrate/nitrite-nitrogen rich wastewater (Table 5). In biological desulfurization, S^0 is the most attractive end-product due to its high demands in industries, agricultural applications and the applications in wastewater technologies [182,183]. VFAs are in high demand as precursor chemicals in food, pharmaceutical and chemical industries; and their

recovery from bioprocesses encourages the paradigm shift towards bio-circular economy [184,185].

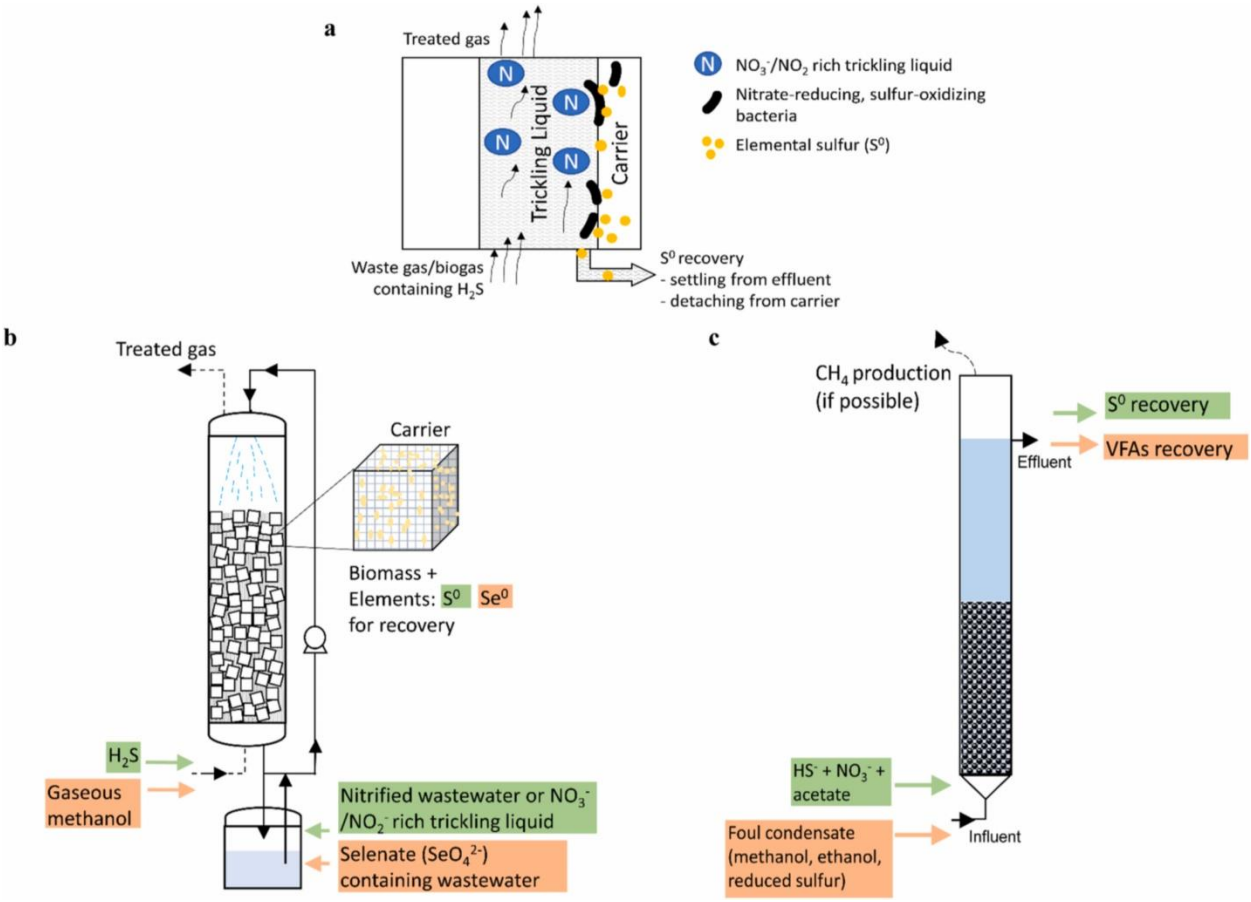


Fig. 5. Resource recovery in anoxic/anaerobic bioreactors used for the removal of VOCs (orange labels) and odorous sulfur compounds (green labels) (a) an example of the mechanism in the packing material in biofiltration of the simultaneous removal of H_2S and NO_3^- (b) anoxic biotrickling filter and (c) UASB. Dashed and continuous lines represent the gas and liquid flow, respectively.

4.1. Bioprocesses for resource recovery from odorous sulfur compounds

In bioprocesses, the removal of sulfur pollutants can be achieved by microorganisms (known as biocatalysts) under aerobic, anoxic, and anaerobic conditions (Eqs. (2), (3), (4), (5), (6), (7), (8)). The treatment of inorganic sulfur polluted gas streams, i.e., sulfide/H_2S, coupling with resource recovery, particularly S^0, is one of the most widely studied and applied in industrial scale. The end-products of bioprocesses for the removal of sulfur pollutants, i.e., S^0, SO_4^{2-} and H_2SO_4, can depend on the operational conditions including pH, temperature, electron acceptor types and the amount of the fed ratio between the sulfur pollutants and the electron acceptors (i.e., molar ratio of H_2S/O_2 or H_2S/NO_3^-) [186,187]:

Table 5. Bioreactor configurations used for the resource recovery from anoxic/anaerobic bioprocesses of VOCs and odorous compounds.

Bioreactor types	Main pollutants (MP)	electron acceptor/donor or co-pollutants	Inoculum	Packing material	EBRT (min)	Max. EC (g MP m^{-3} h^{-1})	%RE	Reference
Anoxic BTFs	H_2S	NO_3^-	Wastewater from municipal WWTP	Pall ring, PUF	2.4, 3	170	99.0	[187, 190]
Anoxic BTFs	H_2S	NO_3^-	The pure culture *Paracoccus versutus* MAL 1HM19	PUF	3	114	97.0	[193]
Anoxic BTFs	H_2S	NO_3^-	Consortium dominated *Thiobacillus* sp.	PUF	3	19	99.0	[192]
UASB	Sulfide-laden wastewater	NO_3^-, acetate	Sludge from the anaerobic sludge thickener	–	4 h (HRT)	29.2	100	[191]
Anaerobic BTF	Gaseous methanol	Thiosulfate	Anoxically incubated activated sludge from municipal WWTP	PUF mixed Pall ring	2.3	21	100	[201]
Anaerobic BTF	Gaseous methanol	Selenate	Activated sludge from municipal WWTP	PUF mixed Pall ring	4.6	46	>80.0	[202]
UASB	Foul condensate (methanol) as COD	Ethanol, acetone, total reduced sulfur	An anaerobic granular sludge from UASB	–	72 h (HRT)	360	42.0−46.0 (COD removal)	[203]

Aerobic desulfurization:

$$H_2S + 2O_2 \rightarrow SO_4^{2-} + H_2O \quad (2)$$

$$H_2S + 0.5O_2 \rightarrow S^0 + H_2O \quad (3)$$

$$S^0 + H_2O + 1.5O_2 \rightarrow H_2SO_4 \quad (4)$$

Anoxic desulfurization:

$$5H_2S + 2NO_3^- \rightarrow 5S^0 + N_2 + 4H_2 + 2OH^- \quad (5)$$

$$5H_2S + 8NO_3^- \rightarrow 5SO_4^{2-} + 4N_2 + 4H_2O + 2H^+ \quad (6)$$

In conventional bioreactors, the S^0 recovery has been successful under aerobic conditions in industrial scale biofilters/BTF under various trademarks, e.g., Shell-Paques/Thiopaq, BioSulfurex, and Biopuric [188,189]. With these technologies, 4–10% air is directly supplied to the inlet gas stream to provide O_2 for the sulfur oxidation process. The recovery of S^0 greater than 90% has been achieved.

Anoxic desulfurization relies on the denitrification processes occurring in the absence of free-O_2 where NO_3^- and/or NO_2^- are used as electron acceptors (Eqs. (5) and (6)). The anoxic desulfurization coupled with S^0 recovery has been conducted with various bioreactor configurations, e.g., biofilters/BTF, CSTR and upflow anaerobic sludge blanket (UASB) in both batch and continuous operation modes [[190], [191], [192], [193], [194], [195]]. The operational conditions of the anoxic bioreactor were commonly achieved at neutral pH (6.8–8.0) and ambient temperature (20–30 °C). At pH below 6.0 or over 9.0, the end products produced during sulfide oxidation caused inhibition to denitrification [196].

Anoxic desulfurization is carried out by autotrophic denitrifying bacteria using inorganic compounds as the carbon and energy sources (Fig. 5a and b). The process causes low biomass production that serves as an advantage to the fixed-film bioreactors like biofilters/BTF to avoid high pressure drop from the overgrowth of biomass. Moreover, the ability of soluble electron acceptor (NO_3^-/NO_2^-) could reduce the limitation of gas-liquid mass transfer of O_2 in aerobic systems [197,198]. The anoxic BTFs have been tested for biogas desulfurization using NO_3^- solution and charge water from WWTP as trickling liquid in laboratory and pilot scales performing with high elimination capacity (EC) of 127–171 g S m^{-3} h^{-1} (at inlet H_2S 1400–14600 ppm_v) [[187], [190], [199]]. The recovery of S^0 was obtained up to 70% at a molar N/S ratio of the influent at 0.5 that was similar to the theoretical value in Eq. (5). Zeng et al. [200] applied the anoxic BTF for H_2S removal from real biogas (67% CH_4, 10% CO_2, 15% N_2 and 0.5% H_2S) using nitrified biogas digestion slurry as the trickling liquid. This integrated process achieved the maximum H_2S EC at 81.29 g H_2S m^{-3} h^{-1} and effectively treated nitrogen containing pollutants (i.e., NH_3, NO_3^- and NO_2^-) from the effluent of the anaerobic system.

Furthermore, the CH_4 content increased by ~1% after the anoxic desulfurization and the S^0 had been observed to be attached on the carrier material which was possible to be further recovered.

The organic matters (e.g., obtained from wastewater) present in the system enabled to enhance heterotrophic denitrifying bacteria that could cause rapid growth of biomass and reduce the sulfide removal efficiency as NO_3^-/NO_2^- required by the denitrification process was depleted [192]. The process could be improved by applying appropriate bioreactor configurations handling high biomass concentration and providing sufficient NO_3^-/NO_2^- sources to the system. The anoxic desulfurization process with the addition of acetate has also been investigated in UASB for S^0 recovery (Fig. 5c), achieving >75.0% efficiency [180]. It was suggested that the ratio of acetate/nitrate/sulfide loading rates required to be optimized for high S^0 recovery efficiency. Yan et al. [194] studied an anoxic sequencing batch reactor (SBR) for the removal of odorous sulfur compounds from wastewater coupling with the anammox process (anaerobic ammonium oxidation). The latter process could enhance S^0 production by ~27% that was collected after the settling step of the SBR operation.

The anaerobic sulfide removal in photobioreactors coupled with S^0 recovery is likely to be a promising technology for resource recovery. Even though small numbers of works on this topic have been reported, the research on the field continues [[204], [205], [206], [207], [208], [209]]. The process has been carried out by green and purple sulfur bacteria (e.g., *Chlorobium* sp. and *Chloronema* sp.) using CO_2 as a terminal electron acceptor:

Anaerobic desulfurization:

$$2H_2S + CO_2 + \text{Light} \rightarrow 2S^0 + CH_2O + H_2O \quad (7)$$

$$H_2S + 2CO_2 + 2H_2O + \text{Light} \rightarrow SO_4^{2-} + 2CH_2O + H_2O \quad (8)$$

The concept of photobioreactor for H_2S removal is similar to the conventional bubble column photobioreactor for CO_2 fixation with an upflow feeding regime. Syed and Henshaw [204] have developed a laboratory scale photobioreactor equipped with LED light at a light intensity of 33 W m^{-2} incubated with pure culture of *Chlorobium limicola* using Na_2S added liquid medium as a sulfur source. The system could be promoted to receive sulfide loading rate up to 338 g m^{-3} h^{-1} and over 90% of the influent sulfide was converted to S^0 [204,205]. In practical applications, Luca et al. [207] applied the laboratory scale photobioreactor inoculated with *C. limicola* for H_2S removal from real biogas produced from the anaerobic digestion of food waste (containing 65% CH_4, 35% CO_2 and 400 ppm_v H_2S). The highest efficiency of the photobioreactor was observed at a H_2S elimination capacity of 17 g m^{-3} h^{-1}, biogas flow rate of 40 mL min^{-1} and 0.5 W m^{-2} light intensity. Anaerobic sulfide removal from the effluent of a pilot scale UASB (located at WWTP in Brazil) has been investigated in photobioreactors containing packing materials obtained light energy from natural sunlight [208,209]. The type of

packing material showed the effect on the reactor performance as the reactor packed with polyurethane foam showed 70–90.0% sulfide removal and 10–28% S^0 recovery efficiencies higher than the one packed with polypropylene ring at the maximum sulfide loading rate at 0.58 g S m^{-3} h^{-1}. Even though the latter performance was much lower than other bioreactors, those photobioreactor types showed the potential application of the bioprocesses for the simultaneous removal of sulfide and organic matter from the anaerobic effluent coupling with S^0 recovery.

4.2. Bioprocesses for resource recovery from VOCs

Anaerobic biodegradation has been used for the treatment of wastewater and waste gas stream contaminated with methanol, which is one of the major VOCs present in the pulp and paper industries, paint industries and petroleum refineries, coupled with the recovery of VFA [[201], [202], [203],210]. The removal of methanol was integrated with the bioreduction of selenate and thiosulfate in anaerobic BTFs achieving maximum methanol ECs at 21 and 46.4 g m^{-3} h^{-1}, respectively [201,202] (Fig. 5b). In the anaerobic BTF for methanol removal with thiosulfate reduction, CH_4 production was found to be negligible in the system (<0.2% *v/v*); however, H_2S present as an intermediate during operation should be considered for further treatment [201]. In this process, VFAs (i.e., ∼2000, 250 and 220 mg L^{-1} of acetate, propionate, and isovalerate, respectively) could be recovered from the effluent at the end of operation; however, the unregulated pH during operation could reduce the VFA production rates. VFA recovery has been achieved in UASB reactors treating foul condensate containing VOCs, including ∼11370 mg L^{-1} methanol, ∼500 mg L^{-1} ethanol and ∼600 mg L^{-1} acetone [203] (Fig. 5c). The carbon recovery as VFAs reached up to 98.0% at 38 °C. Besides the recovery of VFAs, the anaerobic bioprocesses of VOCs could be applied for the CH_4 production due to their high COD concentration without the formation of toxic end products. Lu et al. [211] investigated the biogas production in the UASB for the treatment of kraft pulp wastewater containing methanol (1.6–5.0 g L^{-1}) coupled with sulfate reduction. It was suggested that the conversion of methanol to CH_4 showed high efficiency and was stable at COD/SO_4^{2-} ratio ≥ 2.0.

The anoxic BTFs have been used for the removal of VOCs emission, e.g., benzene, toluene, ethylbenzene and xylene (BTEX), from petrochemical industries [180,181]:

$$aVOCs + bNO_3^- \rightarrow cN_2 + dCO_2 \quad (9)$$

Besides obtaining cleaned gas stream with free-O_2, VFAs (i.e., acetic acid) were detected as an intermediate anoxic biodegradation of toluene at loads of 3–34 g m^{-3} h^{-1} and they seemed to be immediately consumed during the process [180]. Akmirza et al. [181] investigated the application of anoxic BTF for BTEX abatement. The anoxic horizontal flow fixed film

bioreactor coupled with denitrification was investigated for the treatment of BTEX [212,213]. Ethanol was added into the system to enhance the BTEX solubility. The system showed carbon recovery as CH_4 production up to 65 mL d^{-1} at the end of the operation [212].

5. Microbial community and its distribution in different waste gas treatment systems

Biological process has several practical applications to treat waste gas due to its low investment and operating cost. It transfers waste gas into nutrients, energy, cellular components, and inorganic substances through microbe-mediated biological degradation of the pollutants. The microorganisms commonly present in waste gas treatment systems include autotrophic and heterotrophic bacteria and fungi, which can be cultivated as suspended growth in a well-defined liquid media or as attached growth on the packed bed depending on the configuration of the bioreactor. Biofilter, bioscrubber and BTF are the three most widely used biological process for waste gas treatment, where different microbial communities can be enriched while treating different types of waste gas. In these biofiltration systems, waste gas is treated in the biologically active filter bed which commonly contain a mixed consortia of biocatalysts. The inoculum used in the initial stage of different waste gas treatment bioprocess system also could also vary; however, the pollutant loadings characteristics and operational conditions could also affect the microbial community structure/distribution during long-term operations [192,214,215]. In some cases, even when the bioreactors were inoculated with the same biocatalyst and treating the same pollutant, after long term operation, the bioreactors would be characterized by a variety of different dominant bacteria species which would affect the overall performance of the bioreactor [215].

5.1. The inoculum used in different waste gas treatment systems

In most of the biological waste gas treatment processes, activated sludge were used as the inoculum. By feeding the bioreactor with the waste gas, a wide array of microbial communities could be enriched during the start-up stage [192,216]. During the start-up phase, the active microbial community would be low and they might not be sufficient for treating the target pollutants. This could lead to a long start-up period (e.g. 1-10 weeks), and low treatment efficiency. Forbiofilters treating special waste gases, which is difficult to enrich the degradation microbial community from common activated sludge, specific bacteria have to be cultured before the test. Lin et al. [217] investigated toluene removal in a BTF, wherein, a main toluene degrader *Pseudomonas* sp. YATO411 was inoculated. The microbial community results showed that the inoculated bacteria in the system was the dominant strain after various toluene loading tests in the BTF. Zhang et al. [218] applied a BTF to treat hydrophobic VOCs and *Cladophialophora* fungus was inoculated in the reactor. The filter packed with polydimethylsiloxane foam and ceramic composite carrier was started-up within 3 days and

restarted within 7 days after a starvation period of 1 month, where toluene elimination capacity of 264.4 $g\ m^{-3}\ h^{-1}$was achieved. Wu et al. [219] investigated the microbial community composition as a function of the VOC degradation characteristics, wherein the mixed culture enriched from activated sludge of a resin wastewater treatment plant was inoculated to the bioreactor. In that study, the activated sludge was acclimated for 3 months with less than 1 $g\ m^{-3}$ dichloroethane contaminated airflow and it was revealed that the dominant microbial community comprised of 62% *Xanthobacter* genus. Xia et al. [220] applied BTF to remove carbon disulfide and H_2S, wherein concentrated activated sludge from a wastewater treatment plant was cultured for 2 weeks before inoculation. Hence, inoculation with bacteria and fungi which has high performance in the treating the target VOC pollutant(s) are recommended during the start-up stage of BTF.

5.2. The role of aerobic bacteria

Aerobic bacteria plays a major role in the removal of VOCs in a BTF. Li et al. [221] investigated aerobic denitrification and toluene biodegradation using nitrate as an electron acceptor and toluene as the electron donor in a BTF. Aerobic denitrifying bacteria, particularly *Bradyrhizobium*, *Comamonas*, *Cupriavidus*, *Pseudomonas*, *Pseuoxanthomonas*, and *Ralstonia Bradyrhizobium* were capable of achieving simultaneous toluene degradation and denitrification. Zhang et al. [222] investigated VOCs and nitrogen removal using an aerated vertical flow constructed wetland and the ammonia removal was improved. During aerobic biodegradation of VOCs, the VOCs are mineralized to carbon dioxide and water by microbes. The energy (ATP) produced during the biodegradation process was utilized by microbes for biomass production [178]. Long et al. [223] reported that H_2S was effectively reduced under aerobic conditions in a landfill and the abundance of sulfate-reducing bacteria also increased 30 times under aerobic conditions. This could suggest that under aerobic conditions, H_2S gas can be effectively removed in the BTF.

5.3. Role of anaerobic/anoxic bacteria

Biological process has been successfully applied in treating sulfur-containing waste gases, including H_2S, methyl sulfur and dimethyl sulfur etc. *Pseudomonas* has been identified as the dominant species of the bacterial population in different bioreactors used to remove nitrogen, H_2S and different types of VOCs. *Acidithiobacillus, Acidiphilium, and Metallibacterium* were the main strains that could effectively degrade carbon disulfide and H_2S [220]. Huan et al. [224] applied a BTF packed with polyhedral spheres to treat residual biogas containing H_2S and NH_3, wherein the removal efficiency was 98.2% and 88.6%, respectively. In that study, *Dokdonella* and *Thiobacillus* was the main contributor for H_2S and NH_3 removal. For H_2S removal, SOB are the main contributor in general and they are capable to survive under both aerobic and anaerobic conditions. Phototrophic SOB and chemotrophic SOB are the two main

types of bacteria used for treating H_2S, wherein S^0 or sulfate can be an end product. Under anoxic conditions, chemotrophic SOB can utilize H_2S as an electron donor and NO_3^- as electron acceptor. Then H_2S can be oxidized under anaerobic or anoxic conditions, where the products are sulfate, sulfur and nitrite or nitrogen [225,226]. The SOB species that can grow under anoxic conditions include *Thiobacillus denitrificans*, *Thiomicrospira denitrificans*, *Thiosphaera pantotropha* and *Paracoccus denitrificans*. For example, Khanongnuch et al. [192] applied an anoxic BTF to treat H_2S and NO_3^--containing wastewater, wherein H_2S removal efficiency reached over 99.0%.

5.4. Application of fungi for waste gas treatment

Fungal bioreactors for waste gas treatment have reported to exhibit higher efficiency than bacteria inoculated reactors. It is reported that the mycelium of fungi could uptake hydrophobic compounds faster than the bacteria dominant biofilm surface [227]. The advantage of fungi compared with bacteria is that fungi could completely mineralize hydrophobic VOCs under harsh environmental and process conditions, such as limited nutrients, pH fluctuation, shock pollutant loads, low EBRTs and less water contents [228,229]. Zhang et al. [230] reported that bio-trickling filter immobilized with fungi strains started-up within 7 days and could be restarted within less than 7 days after starvation. The average removal efficiency for toluene was greater than 92.5% at toluene loading rates less than 100.9 g m^{-3} h^{-1}. After a certain operational period, the dominant fungi species in the reactor shifted from *Fusarium* to *Paramicrosporidium saccamoebae*. Quan et al. [231] used static magnetic field in a fungal BTF to enhance gas-phase trichloroethylene (TCE) removal. It was reported that the magnetic field intensity could improve the phylum Ascomycota abundance, and thus the BTF also reached its highest removal performance. The application of fungi or bacteria and fungi mixture as inoculum has largely improved the removal of CH_4, toluene, n-hexane, xylene, and styrene [232]. The use of fungi and fungi and bacteria mixture, combined with extra physical or chemical process could have the potential to improve the removal of recalcitrant VOCs.

6. Outlook and future perspectives

The valorization of different forms of liquid and gaseous waste streams is one of the persisting problem all over the world. There is an urgent need to solve this problem from an environmental and economical point of view to provide better living conditions for humans and all life forms. The application of bioprocesses is proposed to be a promising approach due to their advantages such as less energy requirements and environmentally friendly end products.

Bioprocesses for the treatment of contaminated gas streams as well as syngas fermentation have wide application process industries. The main concerns while using gaseous substrates are the solubility in the bioreactor and the cost of the fermentation media. There are numerous ongoing

research focusing on the reduction of the costs by substitution of the medium components by their waste counterparts. On the other hand, novel bioreactor configurations has been proposed to improve the gas-liquid mass transfer of sparingly soluble gases [233]. In addition, maintenance of the temperature is a critical issue keeping in mind the decrease in gas solubility with increasing temperature. Although increased temperatures may be limiting for CO_2 solubility, the use of thermophilic cultures is also supported due to the possible prevention of contamination and less energy requirement to reduce the waste gas temperature. The utilization of waste gas streams for chain elongation by integrating with syngas fermentation has been the focus of research recently. Achieving optimal high ethanol/acetic ratio from syngas fermentation for subsequent chain elongation process is quite challenging as the production of alcohols obtained were much lower than acids. Besides, utilizing syngas fermentation effluents for the generation of other valuable metabolites such as lipids and biopolymers by combing with already established bioprocesses widen the opportunity to utilize waste gas. Enriched mixed cultures that use C1 gases have yielded higher productivity over pure cultures, leading to broadening of the product spectrum. However, detailed studies on the pathways and the metabolic characteristics of using mixed culture are still under development.

Electricity driven MES is a promising biotechnology to treat C1 gases. When this technology is combined with the production of renewable energy and other value added chemicals, it will further reduce the cost of the process. Hence, future research needs to focus on intensifying the process of MES for commercialization, e.g. the development of efficient electrode that allows enhanced biofilm growth and at the same time provide the possibility to create smaller gas bubbles, identify the electrode transfer mechanism at the biocathode surface and the use of real waste gas instead of artificial one. The use of above-mentioned processes for chain elongation driven from the real time waste gas streams is believed to be a novel perspective for environmental protection and promoting circular economy.

7. Conclusions

Bioprocesses for waste gas treatment are promising technologies to promote the concept of bio-circular economy. The industrial waste gas, e.g., VOCs, VICs and syngas, has the potential to serve as feedstock for the recovery of value-added products by combining different downstream processes. The current available technologies include the conventional waste gas treatment systems and hybrid/novel bioreactor configurations, such as biofiltration systems for VOCs and VICs treatment and microbial electrosynthesis for syngas conversion. Each of these bioreactors has its own biochemical mechanism, as well as operating advantages and limits. The performance of all these bioreactor configurations for treating the pollutants, as well as resource recovery from waste gas varies depending on the following parameters: type and source of the biocatalyst (mixed, pure culture) and the enzymes produced, mode of operation (up-flow, downflow mode), aerobic/anoxic/anaerobic treatment, and the process conditions such as the type of waste gas, its composition and concentration (single pollutant/mixture), the packing

material, the pH, temperature, nutrient concentration, gas flow rate and empty bed residence time, respectively. However, for some technologies, the major limitation of adopting a resource recovery approach is the low product concentration/yield, and the high downstream processing costs to recover the value-added product which should be optimized at the pilot and semi-industrial scales in order to successfully implement these technologies in practice.

Authorship contribution

Ramita Khanongnuch: Writing – original draft preparation, Writing – review & editing, Haris Nalakath Abubackar: Conceptualization, Methodology, Writing – review & editing, Supervision, Tugba Keskin: Conceptualization, Writing – original draft preparation, Writing – review & editing, Mine Gungormusler: Writing – original draft preparation, Writing – review & editing, Gozde Duman: Writing – original draft preparation, Writing – review & editing, Ayushi Aggarwal: Writing – original draft preparation, Writing – review & editing, Shishir Kumar Behera: Conceptualization, Methodology, Writing – review & editing, Supervision, Lu Li: Conceptualization, Methodology, Writing – review & editing, Büşra Bayar: Software, Writing – review & editing, Eldon R. Rene: Conceptualization, Methodology, Writing – original draft preparation, Writing – review & editing, 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.

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