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Research review paper

Anaerobic digestion integrated with microbial electrolysis cell to enhance biogas production and upgrading in situ

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ABSTRACT:

Anaerobic digestion (AD) is an effective and applicable technology for treating organic wastes to recover bioenergy, but it is limited by various drawbacks, such as long start-up time for establishing a stable process, the toxicity of accumulated volatile fatty acids and ammonia nitrogen to methanogens resulting in extremely low biogas productivities, and a large amount of impurities in biogas for upgrading thereafter with high cost. Microbial electrolysis cell (MEC) is a device developed for electrosynthesis from organic wastes by electroactive microorganisms, but MEC alone is not practical for production at large scales. When AD is integrated with MEC, not only can biogas production be enhanced substantially, but also upgrading of the biogas product performed in situ. In this critical review, the state-of-the-art progress in developing AD-MEC systems is commented, and fundamentals underlying methanogenesis and bioelectrochemical reactions, technological innovations with electrode materials and configurations, designs and applications of AD-MEC systems, and strategies for their enhancement, such as driving the MEC device by electricity that is generated by burning the biogas to improve their energy efficiencies, are specifically addressed. Moreover, perspectives and challenges for the scale up of AD-MEC systems are highlighted for in-depth studies in the future to further improve their performance.

1. Introduction

Anaerobic digestion (AD) has been employed with a long history as an effective and practical technology to degrade organic wastes with biogas produced as a renewable and green energy product for developing circular economy (Wei et al., 2024; Subbarao et al., 2023). While AD owns advantages, such as low operating cost and net energy output, challenges still need to be addressed for improving its efficiency. For example, long time for establishing a stable and robust microbial community is one of them, and extended hydraulic retention time (HRT) is another, which consequently compromise organic loading rate (OLR) for anaerobic digestors to be built with large volume and relatively high capital investment (Wei et al., 2024). On the other hand, raw biogas is a mixture of CH_4, a large amount of CO_2 up to 40%, and small amounts of other impurities (Calbry-Muzyka et al., 2022). Such a characteristic of raw biogas compromises its energy density, rising a necessity for upgrading with significant cost (Aghel et al., 2022). When AD is integrated with microbial electrolysis cell (MEC), not only is the biogas production enhanced, but also the raw biogas can be upgraded in situ.

MEC has been developed rather late for degrading organic wastes to produce H_2 and other value-added products with electroactive microorganisms (Liu et al., 2005; Kong et al., 2020). A typical MEC is composed of an anode, a cathode, and an external circuit for power supply, and the anode and cathode are separated by an ion-exchange membrane to selectively transport ions (Murugaiyan et al., 2022). The anode takes electrons that are released from the oxidization of organic wastes, and these electrons are transported by the external power through the circuit to the cathode for reducing H^+ to H_2, which can further reduce CO_2 to produce CH_4 or other chemicals (Kong et al., 2020).

A notable drawback of MEC is the loss of carbon resources in the form of CO_2 at the anode. Meanwhile, the overall volumetric productivity of MEC is much lower, since the electrodes cannot provide enough surface for electrochemical reactions to perform efficiently, needless to say high cost for its construction with two chambers that are separated by an ion-exchange membrane. However, when MEC is combined with

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AD, those intrinsic disadvantages can be overcome properly (Fig. 1).

When AD and MEC are integrated, electroactive bacteria can be enriched on the electrode surfaces as well as in the bulk solution to make electron transfer more efficient and effective, which consequently enhances the AD process for biogas production (Wang et al., 2022). In addition, with the assistance of electrotrophic and hydrogenotrophic methanogens, more CO_2 can be reduced to CH_4 for upgrading the raw biogas in situ to save capital investment on upgrading facilities with conventional AD processes (Ning et al., 2021). However, no commercial applications of AD-MEC systems are available at present, although intensive studies have been performed.

This critical review aims at assessing the state-of-the-art progress in AD-MEC systems, focusing on underlying fundamentals with AD and MEC, materials and configurations for fabricating the electrodes, designs for integrating the two units, and potential applications of these integration systems. Meanwhile, challenges and strategies for their solutions are also highlighted.

2. Fundamentals of AD and MEC

2.1. Microbial syntrophy and synergy in AD

AD is a complicated process through which microbial consortia degrade organic wastes synergistically. As shown in Fig. 2, AD involves four major stages: hydrolysis, acidogenesis, acetogenesis, and methanogenesis, through which a gaseous mixture composed predominately of CH_4 and CO_2 is produced (Van et al., 2020).

2.1.1. Hydrolysis

Organic wastes including proteins, polysaccharides, and lipids are insoluble, and thus cannot be digested directly by microorganisms (Li et al., 2019). During hydrolysis, these organic wastes are hydrolyzed

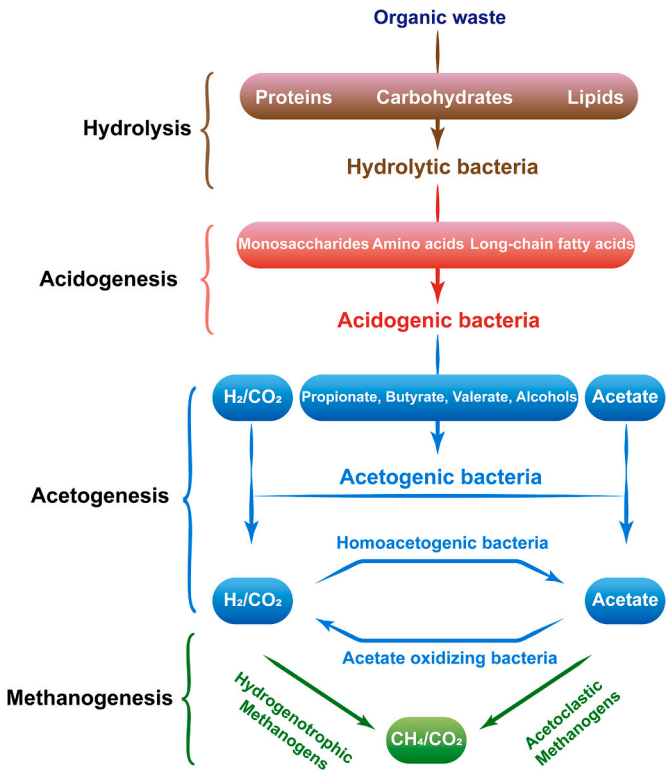

Fig. 2. Major reactions with AD processes (hydrolysis, acidogenesis, acetogenesis, and methanogenesis).

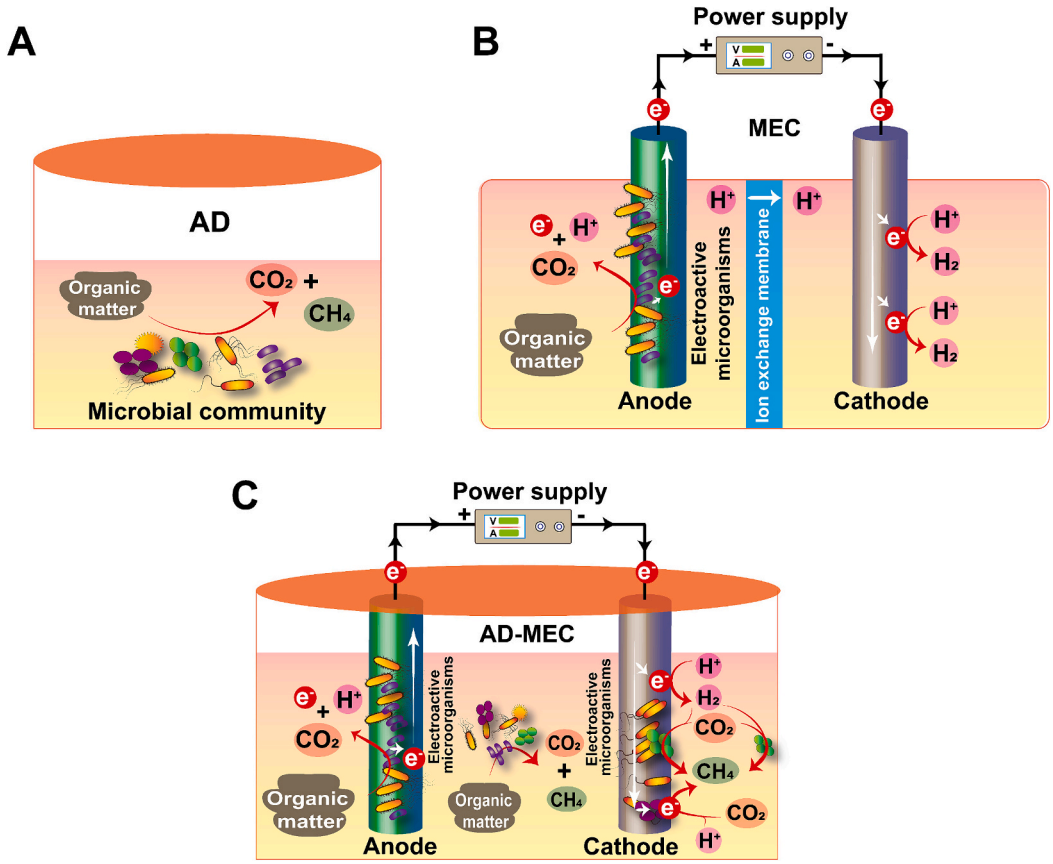

Fig. 1. Schematic diagrams for AD (A), MEC (B), and AD-MEC (C).

into soluble molecules, such as amino acids, monosaccharides, and long-chain fatty acids by extracellular enzymes that are secreted predominately by hydrolytic bacteria at low levels (Wei et al., 2024). In general, hydrolysis is the rate-limiting step for AD, due to the heterogeneous characteristics of these enzymatic reactions and difficulties for different substrates to be accessible by the hydrolytic enzymes.

2.1.2. Acidogenesis

Soluble products released during the hydrolysis cannot be utilized directly by methanogens, but they can be metabolized by acidogenic bacteria to produce volatile fatty acids (VFAs) including acetic acid, propionic acid, butyric acid, valeric acid, etc., alcohols such as methanol and ethanol, and a small amount of H_2 and CO_2 (Li et al., 2019). Although acetic acid and H_2 can be assimilated by methanogens, other acidogenic products need to be further decomposed to acetic acid and H_2 through acetogenesis.

2.1.3. Acetogenesis

VFAs and alcohols can be further converted by hydrogen-producing acetogenic bacteria with H_2 and CO_2 produced (Van et al., 2020). Thermodynamically, the anaerobic oxidation of these compounds by hydrogen-producing acetogenic bacteria is not favorable under pure culture conditions with positive free energy as highlighted in Eqs. (1)–(4) (Wang et al., 1999; Zhang et al., 2023), but when they are co-cultured with hydrogen-consuming methanogens, syntrophy can develop to keep hydrogen partial pressure at extremely low levels, driving the acetogenesis forward.

$$CH_3CH_2COO^- + 3H_2O \rightarrow CH_3COO^- + H^+ + HCO_3^- + 3H_2, \Delta G^{0'} = 76.1 kJ/mol \quad (1)$$

$$CH_3CH_2CH_2COO^- + 2H_2O \rightarrow 2CH_3COO^- + H^+ + 2H_2, \ \Delta G^{\circ'} = 48.1 \ kJ/mol \quad (2)$$

$$CH_3CH_2CH_2CH_2COO^- + 2H_2O \rightarrow CH_3CH_2COO^- + CH_3COO^- + H^+ + 2H_2, \ \Delta G^{\circ'} = 25.1 \ kJ/mol \quad (3)$$

$$CH_3CH_2OH + H_2O \rightarrow CH_3COO^- + H^+ + 2H_2, \ \Delta G^{\circ'} = 9.6 \ kJ/mol \quad (4)$$

2.1.4. Methanogenesis

The last stage for AD is methanogenesis through which those intermediates released during acidogenesis and acetogenesis are assimilated by methanogens to produce CH_4 (Li et al., 2019). Generally, methanogens can be classified into three groups, which are highlighted in Eqs. (5)–(7).

$$\text{Hydrogenotrophic methanogenesis: } CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \quad (5)$$

$$\text{Acetoclastic methanogenesis: } CH_3COOH \rightarrow CH_4 + CO_2 \quad (6)$$

$$\text{Methylotrophic methanogenesis: } 4CH_3OH \rightarrow 3CH_4 + CO_2 + 2H_2O \quad (7)$$

The first is hydrogenotrophic methanogens such as *Methanothermobacter*, *Methanoculleus*, and *Methanobacterium*, which utilize H_2 as electron donor and CO_2 as electron acceptor to produce CH_4, and the second is acetoclastic including *Methanosarcina* and *Methanothrix*, which metabolize acetic acid into CO_2 and CH_4 (Demirel and Scherer, 2008). The last is methylotrophic such as *Methanohalophilus* that converts methyl groups with methylated compounds including methanol to CH_4 (L'Haridon et al., 2020).

Methanogens metabolize very limited substrates, and their growth rates are much lower than fermentative bacteria for acidogenesis and acetogenesis. Thus, methanogenesis could be another rate-limiting step (Van et al., 2020). Various parameters such as temperature, pH, HRT, OLR, and nutritional conditions affect methanogenesis. Appropriate temperatures for mesophilic and thermophilic methanogens are 30–40 °C and 50–60 °C, respectively (Kaur et al., 2024), and neutral pH values of 6.5–8.5 are needed for them, since their activities are inhibited under acidic or alkaline environments (Demirel and Scherer, 2008). Major metabolic pathways of hydrogenotrophic, acetotrophic, and methylotrophic methanogens are shown in Fig. 3.

2.2. Biosynthesis in MEC

MEC was developed initially by Liu et al. (2005) to produce H_2 from acetic acid on the cathode driven by an external voltage to address the challenge of fermentative production of hydrogen from sugars with a large amount of acetic acid produced as a byproduct. A typical MEC consists of two chambers separated by a proton exchange membrane, and each chamber contains an electrode (Pawar et al., 2020).

Firstly, electroactive bacteria enriched on the anode surface degrade acetic acid to release electrons, CO_2, and H^+ at an open circuit potential of -0.300 V. Then, the electrons are transferred directly onto the anode surface for being transferred further to the cathode surface via the external circuit. Finally, the H^+ selectively passes through the proton exchange membrane for being reduced on the cathode surface to produce H_2 under an open circuit potential of -0.414 V. Eqs. (8) and (9) highlight these electrochemical reactions that are occurred on the electrodes (Gautam et al., 2023).

$$\text{Anode: } CH_3COOH + 2H_2O \rightarrow 2CO_2 + 8e^- + 8H^+ \ (E_A = -0.300 \ V) \quad (8)$$

$$\text{Cathode: } 8H^+ + 8e^- \rightarrow 4H_2 \ (E_C = -0.414 \ V) \quad (9)$$

Theoretically, an external circuit voltage of 0.114 V can drive the hydrogen production, but in practice the applied voltage is higher than the theoretical value due to a necessity for overcoming the overpotential and internal resistance. Such a low voltage indicates that the consumption of electrical power for driving MEC is not intensive. However, there is an intrinsic disadvantage with MEC: hydrogen production occurs on the cathode surface, making such a device alone less, even not practical for applications at large scales, due to very limited surface areas provided by the electrode for those electrochemical reactions to be performed at acceptable volumetric rates.

2.3. Integration of AD with MEC

When MEC is integrated with AD, not only can its disadvantages be overcome, but also its potentials explored ultimately. The argument for such an integration is the consumption of electrical power to drive MEC. Reasons for MEC to enhance biogas production with AD, and in the meantime to improve the quality of this bioenergy product are commented below.

Firstly, the enrichment of electroactive bacteria in biofilms developed on the electrode surface and bulk solution stimulated by exerting an external voltage facilitates the hydrolysis of organic wastes and the acidogenesis as well, which consequently provides more assimilable substrates for methanogenesis (Min et al., 2020). Zhang et al. (2015) coupled an acidogenic reactor with Fe-C electrodes to establish an AD-MEC system, through which the acidogenic efficiency was improved significantly.

Secondly, electron transfer is a key for AD to produce biogas, and through supplying an external voltage, electron transfer from electroactive bacteria to the electrodes can be enhanced (Wang et al., 2022). Electron transfer is divided mainly into two categories: direct electron transfer (DET) mediated by pilus (nanowires) of electroactive bacteria and c-type cytochromes within their membranes and indirect electron transfer (IET) via mediators such as H_2 and ferredoxin (Fd), and both DET and IET can be enhanced when AD integrated with MEC (Yang et al., 2012).

Finally, organic compounds are oxidized within the electroactive biofilm that is developed on the anode surface, and H_2 can be produced

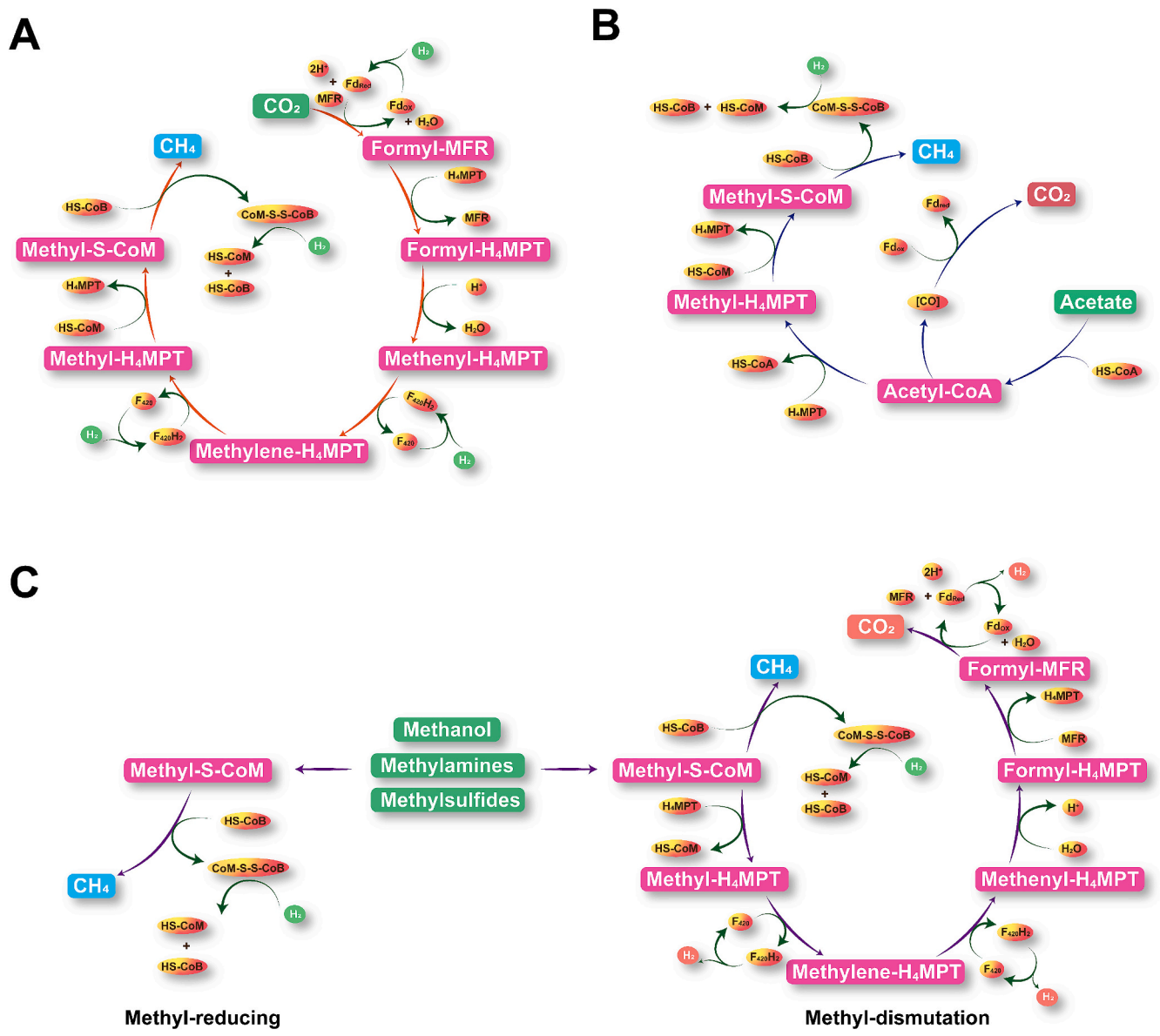

Fig. 3. Metabolic pathways of hydrogenotrophic methanogenesis (A), acetoclastic methanogenesis (B), and methylotrophic methanogenesis (C). MFR: methanofuran; F_d: ferredoxin; H_4MPT: tetrahydromethanopterin; HS-CoM: sulfhydryl coenzyme M; HS-CoB: sulfhydryl coenzyme B; CoM-SS-CoB: heterodisulfide coenzymes CoM and CoB.

at the cathode surface when H^+ takes electron to reduce CO_2 with CH_4 produced, leading to in situ upgrading of biogas. Liu et al. (2021) developed an AD-MEC system by supplementing acetic acid into artificial wastewater as the sole electron donor, and experimental results showed that the purity of CH_4 was increased to 97%, comparable to biogas that is upgraded through a physiochemical process with high cost.

As shown in Fig. 4, two dominant reactions can occur on the cathode surface (Huang et al., 2020): a) electroactive methanogens take electrons, converting H^+ and CO_2 directly to CH_4, and b) H^+ is reduced to H_2 through biotic and abiotic reactions to be assimilated together with CO_2 by hydrogenotrophic methanogens to produce CH_4.

It has been verified that, for total CH_4 production through AD, hydrogenotrophic methanogenesis contributes about 30% (Litti et al., 2022). When AD is integrated with MEC, CH_4 production by hydrogenotrophic methanogens can be enhanced substantially, since more H_2 is available in the cathodic environment for reducing CO_2, making the hydrogenotrophic methanogens more productive to upgrade the biogas in situ (Aryal et al., 2022). Moreover, the enrichment of hydrogenotrophic methanogens in the cathode biofilm can improve their growth and tolerance to environmental stresses such as toxicity from ammonia and organic acids as well as temperature fluctuations to further enhance biogas production and upgrading (Wu et al., 2021).

Zhi et al. (2022) equipped AD with a carbon brush anode and a hybrid Ti/RuO_2-graphite felt cathode to treat activated sludge waste, in which CH_4 production was improved significantly with a maximum yield of 16.4 mL/L achieved under the applied voltage of 1.2 V and solid retention time of 15 d, and analysis on the microbial community showed that *Methanobacterium* was dominant in the AD-MEC system.

Compared with conventional AD processes, microbial community in the bulk solution of AD-MEC systems can also be enriched with more electroactive microorganisms to enhance interspecies electron transfer, and the planktonic electroactive bacteria and methanogens enriched by externally applied voltage in the bulk solution of AD-MEC systems further accelerate the degradation of organic wastes and CH_4 production (Feng et al., 2018). Similar to methanogenesis in the biofilm, hydrogenotrophic methanogens are predominated in the bulk solution. Park et al. (2020) investigated the effect of an external voltage on the

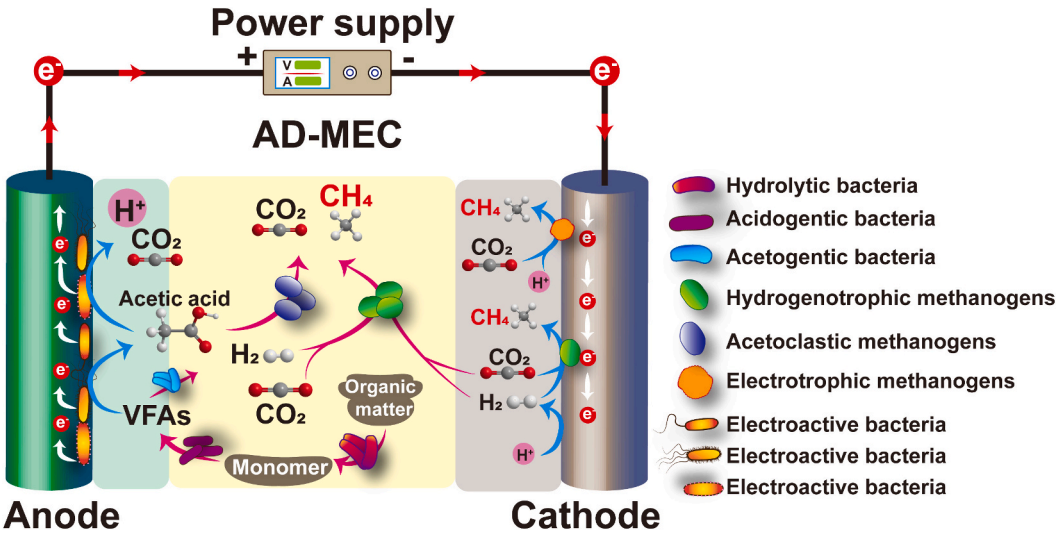

Fig. 4. Electromethanogenesis in AD-MEC systems.

performance of an AD-MEC system that was fed with food wastes, and proved that the current density in the bulk sludge was enhanced in comparison with the control, indicating that the electroactive bacteria was enriched, and the electron transfer between species was improved. Feng et al. (2018) claimed that microbial community enriched with electroactive bacteria in the bulk solution contributed even more to the increase of CH_4 yield.

These studies indicate that not only can the integration of AD with

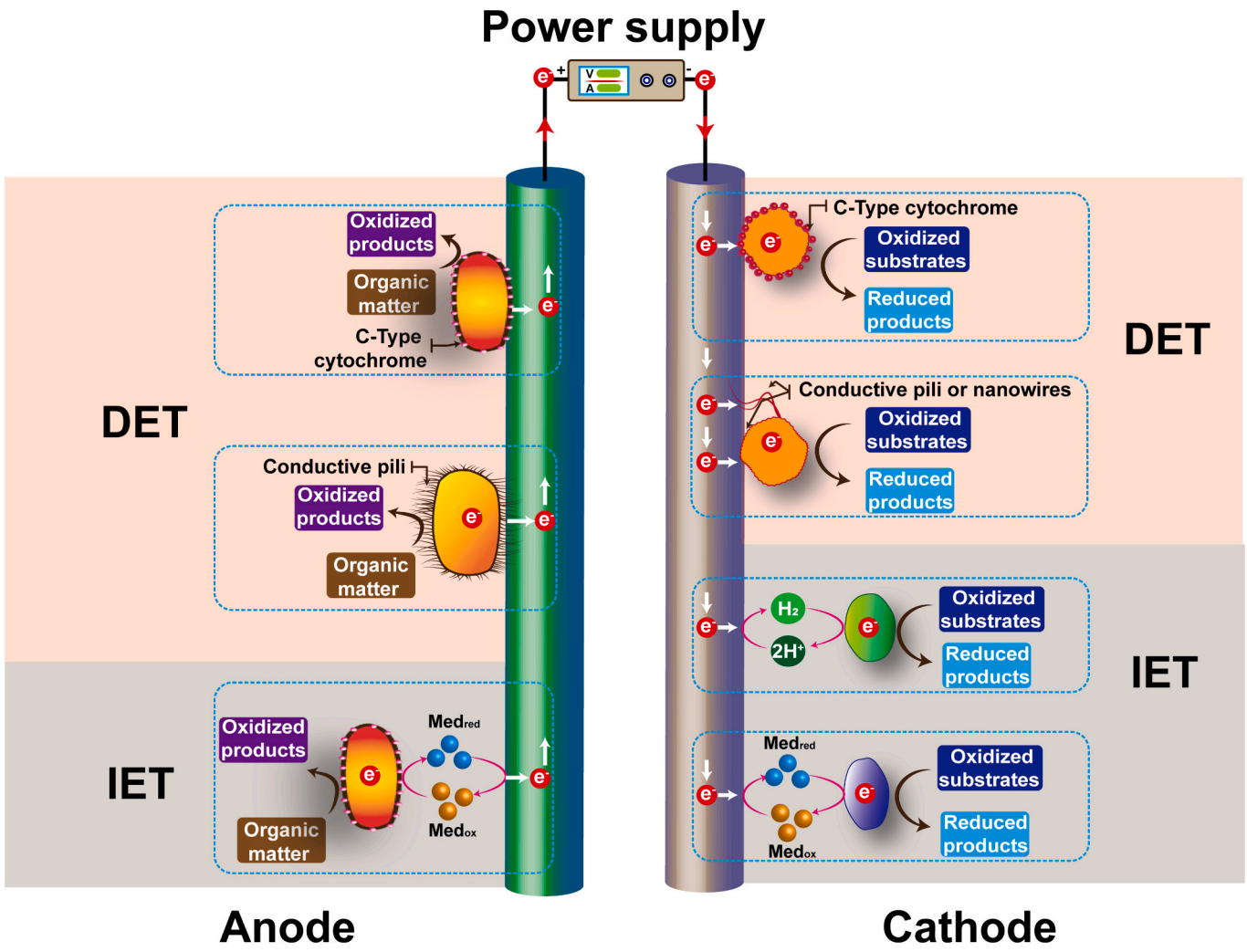

Fig. 5. Electron transfer from microbes to the anode and cathode surfaces via DET and IET.

MEC enhance reduction reactions on the cathode surface, but also improve biogas production in the bulk solution through enriching electroactive microorganisms and hydrogenotrophic methanogens.

2.4. Extracellular electron transfer in AD-MEC systems

There are two types of electroactive bacteria in AD-MEC systems (Fig. 5): electrotrophic microorganisms can transport electrons among cells, and exoelectrogens transport electrons from cells to other electron acceptors, but both of them can transfer electrons via DET and IET via the electrodes (Logan and Rabaey, 2012). While mechanisms underlying extracellular electron transfer remain to be further elucidated, possible pathways have been proposed (Pawar et al., 2020).

Basically, c-type cytochromes embedded into cell membranes and conductive pilus/nanowires are responsible for DET, and mediators or electron shuttles such as phenazines, flavins, H_2, neutral red, and methyl viologen are responsible for IET (Zhao et al., 2021).

C-type cytochromes are most common electron transfer media through DET, and most of them are located in the outer membrane of archaea, bacteria, and eumycophyta. For example, the membrane of *Shewanella oneidensis* contains c-type cytochromes and structural proteins, which have been proved to be the electron transfer conduit known as the Mtr respiratory pathway (Edwards et al., 2020). Ross et al. (2011) examined the feasibility of electron flow from the electrode to *S. oneidensis*, which is the reverse process of the Mtr respiratory pathway, and their results demonstrated that the Mtr pathway could also conduct reductive reactions, proving this electron transfer conduit is reversible.

The pilus of *Geobacter sulfurreducens* and *S. oneidensis* have been proved to act as biological nanowires for transferring electrons from cells to extracellular electron acceptors via DET, and these species are generally dominant for oxidizing organic matters to release electrons effectively onto the anode surface through DET (Clarke, 2022).

Some electrotrophic methanogens can also acquire electrons directly from the cathode surface via c-type cytochromes and conductive pilus/nanowires to reduce CO_2 and H^+ directly to CH_4 (Zakaria and Dhar, 2019). Liu et al. (2020a) investigated the effect of different cathode materials used for fabricating an AD-MEC bioreactor on biogas production and upgrading, and found that both biogas yield and purity were higher when carbon brush was employed compared to graphite plate. Besides, analysis on the microbial community indicated that *Methanothrix* could uptake electrons directly from the cathode surface via conductive pilus (Liu et al., 2019). Rowe et al. (2019) reported that *Methanosarcina barkeri*, a cytochrome-containing methanogen, participated in DET on the cathode surface of an AD-MEC system, and hydrogenase-mediated and free extracellular enzyme-independent modes were further characterized for the process.

CO_2 and H_2 serve as substrates for hydrogenotrophic methanogens to produce CH_4, but the low solubility of H_2 limits further improvement on the biogas production (Rabaey and Rozendal, 2010). Except for electron shuttles secreted by electroactive microorganisms, such as flavins and phenazines, some other redox compounds, including methyl viologen, viologen dyes, and neutral red, can also function well as electron shuttles for IET (Zhao et al., 2021). Although these electron shuttles can dissolve well in the bulk solution for being used repeatedly, a limit lies with their instability and toxicity to microorganisms (Noori et al., 2020). It is generally agreed that DET is more efficient than IET, since potential resistance with mass transfer for electron mediators with IET can be eliminated (Thapa et al., 2022).

Electron transfer in the bulk solution of AD-MEC systems also exists between electron-donating bacteria and methanogens via direct interspecies electron transfer (DIET) and indirect interspecies electron transfer (IIET). As shown in Fig. 6, enriched electroactive bacteria in the bulk solution can establish syntrophy with methanogens to degrade organic substances and transfer electrons to methanogens directly via conductive pilus/nanowires and other conductive materials or indirectly via mediators such as H_2 (Li et al., 2021).

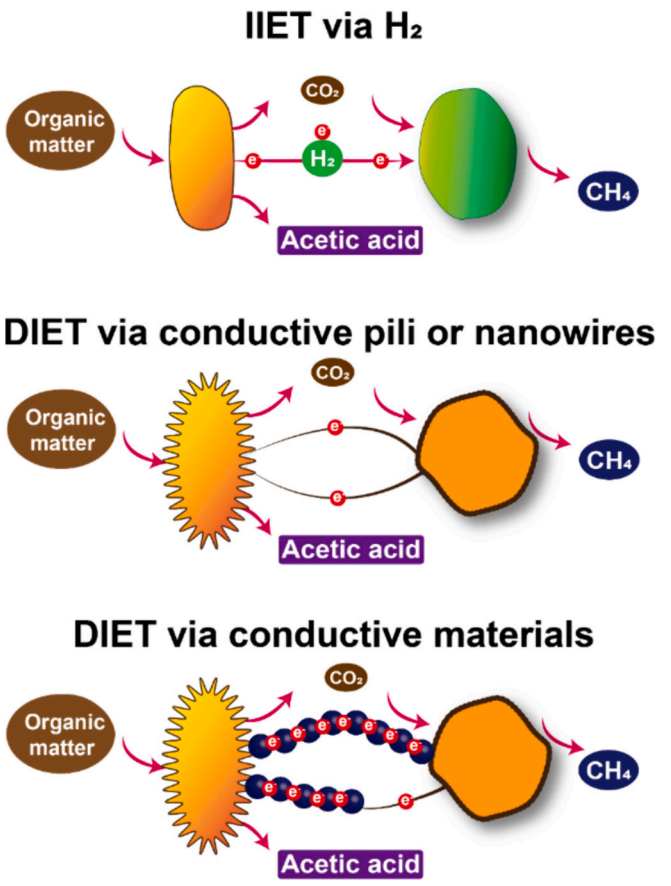

Fig. 6. IIET and DIET through conductive pilus/nanowires or conductive materials.

Rotaru et al. (2014) confirmed that *Geobacter* and *Methanosaeta* (*Methanothrix*) exchanged electrons via DIET, and when *G. metallireducens* and *M. harundinacea* were co-cultured in a defined medium, *M. harundinacea* took electrons via DIET to reduce CO_2 and H_2 with CH_4 produced. Another case is that *M. barkeri* was syntrophic with *G. hydrogenophilus* to accept electrons also via DIET to enhance CH_4 production (Rotaru et al., 2015).

The supplementation of a small amount of conductive materials is an effective strategy to improve DIET. Piao et al. (2019) verified the effectiveness of supplementing activated carbon as conductive materials for DIET between electroactive bacteria and electrophic methanogens in AD-MEC systems. Feng et al. (2020) investigated the effect of electrostatic field and the addition of activated carbon on AD-MEC systems, and concluded that the electrostatic field improved methane yield and production rate, and activated carbon further enhanced the overall performance via DIET.

Interspecies electron transfer among various microorganisms is vital for degrading organic substances and biogas upgrading in AD-MEC systems. Nonetheless, mechanisms underlying the electron transfer needs to be explored further for comprehensive understanding of this process to improve it more effectively.

3. Configurations of AD-MEC systems

Although the configuration highlighted in Fig. 1C is simple for developing AD-MEC systems, other designs have also been explored to improve their performance. Basically, two strategies have been proposed: integrating AD with MEC in situ where electrodes are embedded directly into AD reactors, and the sequential combination of AD with MEC for them to be operated separately.

3.1. Integrating AD with MEC in situ

This strategy is more economically competitive, since additional cost for constructing MEC reactors is minimized. As shown in Fig. 7, three configurations have been developed so far: single chamber, two chambers, and three chambers (Liu et al., 2017a; Zheng et al., 2021; Pan et al., 2021).

For a single-chamber AD-MEC reactor, no ionic membrane is installed between the anode and cathode to save capital investment. Besides, the internal resistance, which contributes predominately to energy loss with charge transfer, is maintained at a low level for transferring ions more efficiently from the anode to cathode (Park et al., 2020). Single-chamber AD-MEC reactors have been widely applied for electromethanogenesis. Kobayashi et al. (2017) designed a single-chamber reactor for inoculating with thermophilic methanogens to evaluate its electromethanogenesis performance, and observed that CH_4 production rate based on the cathode surface area was improved from 34.9 to 168.4 mmol/m^2/d when the applied voltage increased from 0.4 to 0.9 V. Bo et al. (2014) compared the performance of biogas upgrading through a single-chamber AD-MEC reactor with a traditional AD process, and an increase of 2.3 times in CH_4 productivity with purity as high as 98% was observed for the AD-MEC system. Guo et al. (2013) degraded sewage sludge in a single-chamber AD-MEC reactor, which enhanced CH_4 production of 11.4–13.6 times at the applied voltage of 1.4–1.8 V.

MEC has also been combined with AD reactors with other configurations in addition to continuously stirred tank reactor (CSTR) for applications in waste treatment to produce value-added products. Cui et al. (2016) conducted an AD-MEC system using an up-flow hybrid anaerobic digestion reactor with imbedded electrodes to investigate its effect on treating wastewater containing azo dyes, and found that when the electrodes were inserted into the liquid phase, more efficient degradation of azo dyes was observed. Wang et al. (2019) developed an up-flow anaerobic sludge blanket (UASB) coupled with electrodes embedded to treat wastewater containing acetyl pyrimidines, and their results demonstrated that a higher removal efficiency of 96.3% was obtained for acetyl pyrimidines in comparison with the UASB reactor alone. Anaerobic fluidized bed reactor has also been integrated with MEC to treat brewery wastewater, which showed improved effect on removing COD and total nitrogen (Asensio et al., 2021). Park et al. (2021) studied the efficiency of treating digestate in a trickling filter bed reactor combined with MEC for biogas upgrading, which improved CH_4 content substantially from 53% to 83%.

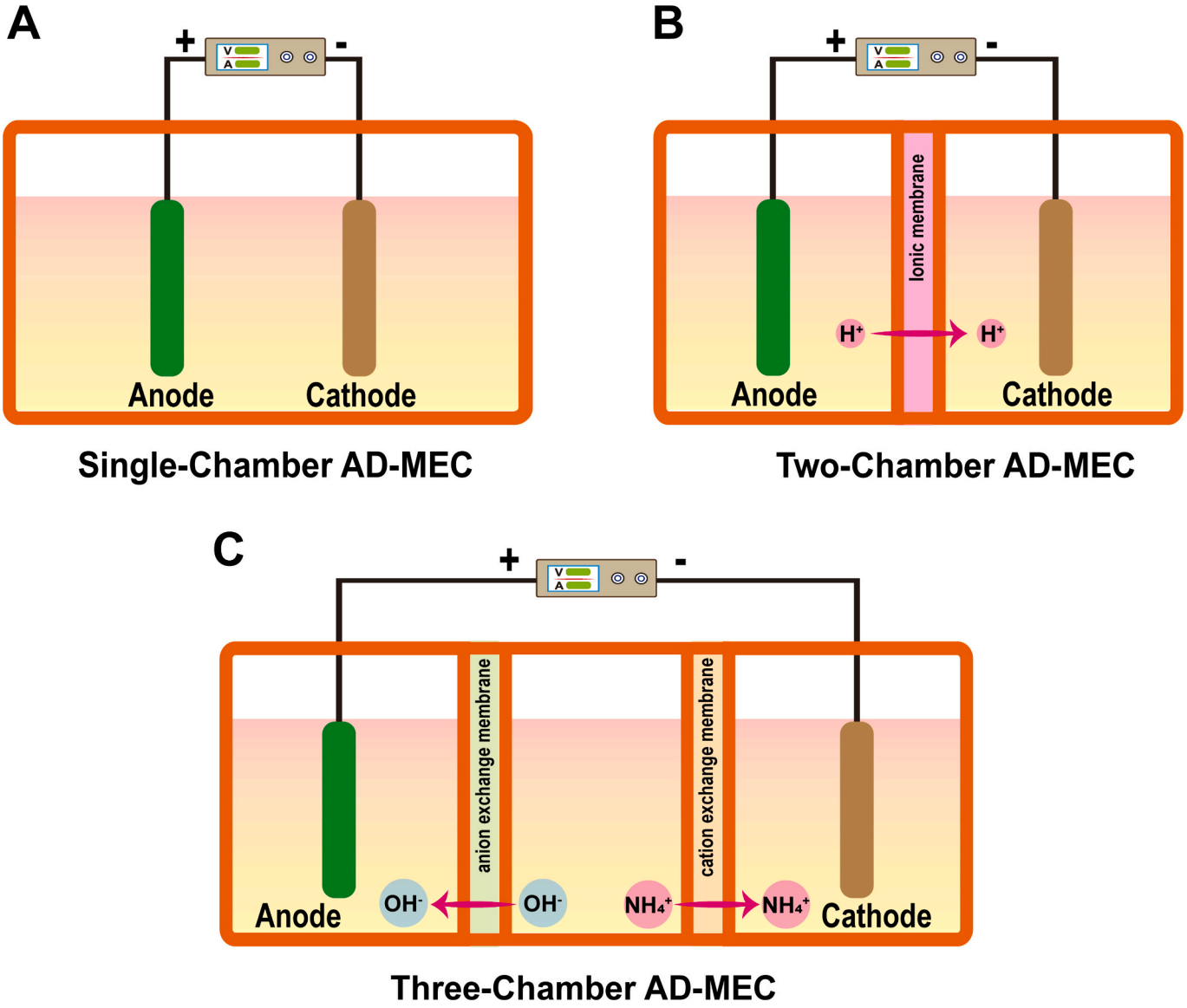

Fig. 7. Configurations of AD-MEC systems with single chamber (A), two chambers (B), and three chambers (C).

Although single-chamber AD-MEC systems have been widely studied, disadvantages, such as oxygen toxicity to obligate microbial consortia, particularly to methanogens, may occur to inhibit their activities. Two-chamber AD-MEC reactors with ionic membranes can prevent the exchange of oxidized and reduced products between the anode and cathode chambers (Ao et al., 2023).

Liu et al. (2017a) compared the performance of treating organic wastes by a single-chamber AD-MEC reactor and a two-chamber AD-MEC with a cation-exchange membrane, and found that the two-chamber system produced CH_4 more sustainably, since the membrane prevented VFAs from diffusing into the cathode side for toxicity to methanogens. Nonetheless, studies on two-chamber AD-MEC reactors are scarce in comparison with single-chamber systems, due to their high internal resistance for energy consumption, extra cost with ionic membranes, and frequent maintenance operations.

Three-chamber AD-MEC reactors with an additional chamber between the anolyte and catholyte have also been developed, through which excessive VFAs and ions generated and accumulated within the anode and cathode chambers, respectively, can be transferred to the third chamber through a cation exchange membrane (CEM) at the catholyte side and an anion exchange membrane (AEM) at the anolyte side.

Zheng et al. (2021) designed a three-chamber AD-MEC reactor to digest blue algae, a major contaminant in lakes, in its middle chamber, which not only removed ammonium nitrogen with the biomass, but also enhanced CH_4 production. Pan et al. (2021) applied a three-chamber bioelectrochemical reactor to achieve simultaneous wastewater treatment and the culture of *Chlorella vulgaris* as single cell proteins. However, more complicated configurations of three-chamber AD-MEC reactors require intensive capital investment, which consequently compromises their economic competitiveness.

3.2. Two-stage integration of AD with MEC

Two-stage integration of AD with MEC is usually applied for improving the digestibility of organic wastes to enhance energy production, in which the MEC reactor can function as a separate post-treatment unit for biogas production and upgrading.

Krishnan et al. (2019) used a two-stage AD-MEC system to treat effluent discharged from a palm oil mill, in which CSTR and MEC reactors were used for producing H_2 and CH_4, respectively, and experimental results indicate that the CSTR gave a yield of 205 mL/g (COD) for H_2 production, and subsequently, the digestate with byproducts acetic and propionic acids was fed into the MEC reactor operated with an external voltage of 0.5 V, gaining a yield of 290 mL/g (COD) for CH_4 production.

Barbosa et al. (2019) employed an AD reactor to digest urine and a MEC reactor to further degrade the digestate, and their experimental results indicate that the removal rate of COD (0.14 g/L/d), current density (218 mA/m^2), and coulombic efficiency (17%) were all improved in comparison with the MEC unit alone without such an AD reactor integrated. Cerrillo et al. (2018) used an AD reactor connected with a MEC device to digest pig waste, and such an integrated system was more stable when the organic and nitrogen loading rates were doubled.

Based on experiments conducted on lab studies, the two-stage combination of AD with MEC has been proved to be an enable concept, but its feasibility for applications at large scales is challenging, since technically these two units are difficult to be matched well in their treating capacities, and economically significant capital investment is needed for the MEC device.

4. Applications of AD-MEC systems

Growing effort has been paid on developing AD-MEC systems to address challenges with traditional AD processes: enhancing the degradation of organic wastes, improving biogas production, and upgrading the biogas product in situ for high purity.

4.1. Treating organic wastes

AD-MEC systems can enhance the treatment of organic wastes, especially for recalcitrant pollutants such as activated sludge, wastewater containing refractory compounds, and wastes from catering industry, food processing, and livestock farming with uncertain components.

Zhi et al. (2022) treated waste-activated sludge (WAS) via traditional AD, and drawbacks such as extremely long retention time for solids, toxicity of ammonia accumulated, and inhibition from VFAs substantially compromised its performance, but when MEC was integrated and operated with an applied voltage of 1.2 V, a shorter solid retention time of 15 d and a higher methane production rate, 8.6 times of that detected with the AD process, were observed for the AD-MEC system. Qin et al. (2021) also treated WAS with an AD-MEC reactor operated at an applied voltage of 0.8 V, which reduced the start-up time, and enhanced the process stability, making CH_4 production yield increased 31.7% compared to that achieved with the traditional AD system.

Azo dyes are major components in wastewater discharged by printing and dyeing industries, which are toxic to humans with health risks such as carcinogenicity, teratogenicity, and mutagenicity if it is released into environments inappropriately (Cui et al., 2019). Wang et al. (2019) developed a UASB-MEC system to treat acetyl pyrimidines-containing wastewater, which removed as high as 96.3% acetyl pyrimidines and 92.9% total organic carbon. Xie et al. (2021) evaluated the impact of cathode materials on treating wastewater containing *N*, *N*-dimethylacetamide, and found the AD-MEC reactor with stainless-steel mesh as cathode showed better COD removal efficiency and electrochemical performance.

The anaerobic treatment of food waste is difficult to be operated stably in traditional AD systems under high OLR conditions, because acidification can develop quickly (Xu et al., 2018). However, AD-MEC systems can facilitate the conversion of VFAs accumulated more efficiently to relieve the acidification (Chung and Dhar, 2021). Choi and Lee (2019) developed an AD-MEC system to treat food waste with an applied voltage of 1.2 V and the substrate loading of 2.4 g COD/L, which enhanced the yield and rate of CH_4 production at 20% and 30%, respectively, compared to the traditional AD process. Park et al. (2018) designed an AD-MEC reactor with a rotating impeller as anode to improve the performance of food waste treatment, which was operated stably at a high OLR of 6 kg COD/m^3/d.

AD-MEC systems have also been applied to degrade livestock waste including chicken manure and pig slurry that contains higher organic component and nitrogen concentrations (Huang et al., 2020). Ammonium in livestock manure was recovered via a two-chamber AD-MEC system with CEM for ammonium in the anode chamber to migrate into the cathode chamber, relieving its inhibition on the activities of electromethanogens to improve their electromethanogenesis (Cerrillo et al., 2021). Wagner et al. (2009) use a single chamber AD-MEC system with graphite-fiber brush as anode to treat swine slurry, producing H_2 and CH_4 simultaneously.

Generally, all organic wastes treated by traditional AD reactors can be treated by AD-MEC systems to improve their degradation. However, studies on AD-MEC systems have been performed predominately at lab scales, making them difficult to be scaled-up for applications at large scales, since cost-effective and biocompatible electrode materials are still limited.

4.2. Enhancing biogas production and upgrading in situ

AD-MEC systems can accelerate methanogenesis via enhancing hydrolysis, enriching electroactive bacteria and methanogens, and improving interspecies electron transfer (Park et al., 2020), which are

promising for biogas production and upgrading, since these systems can promote the degradation efficiency of substrates and convert more CO_2 to CH_4 in situ almost without significant increase on capital investment and operating cost.

CH_4 content in biogas was increased to 76.9% when an AD-MEC system with an applied voltage of 0.3 V was used to treat sewage sludge, much higher than only 50–60% obtained via traditional AD reactors (Song et al., 2016). Another study with a single-chamber AD-MEC reactor made CH_4 content increased to 98%, indicating its excellent biogas upgrading performance (Bo et al., 2014). Tartakovsky et al. (2021) investigated the biogas upgrading performance of an AD-MEC reactor, and a high purity of CH_4 (85–90%) was produced. Park et al. (2021) treated anaerobic digestate with a trickling filter bed reactor integrated with MEC for biogas upgrading, and experimental results demonstrated that CH_4 content was increased significantly from 53% to 83%.

In addition to improving CH_4 content, many studies have been focused on improving CH_4 yield and productivity as well. Cai et al. (2016a) designed a two-chamber UASB-MEC reactor, which achieved a methane productivity of 70 mL/L·d, 2.6 times higher than that produced by a regular AD reactor. Furthermore, WAS was treated in a hybrid reactor coupled AD with MEC, which gained a productivity of 60 mL/L·d for CH_4 production, 2.0 folds higher than that achieved by the traditional AD reactor (Cai et al., 2016b). Gao et al. (2021) designed an AD-MEC system to improve wastewater treatment and biogas production simultaneously, which produced biogas with CH_4 yield increased 1.6-fold and purity improved to 90% from 55%.

4.3. Other applications

Ammonia inhibition frequently occurs during AD processes when treating substrates with high nitrogen contents, such as chicken manure, pig slurry, and blue algae. Integrating AD with MEC can improve the stability and robustness of AD processes for more efficient removal and recovery of ammonium (Cerrillo et al., 2018). Wang et al. (2021) developed a three-chamber AD-MEC reactor separated by AEM and CEM for digesting blue algae in the middle chamber so that ammonium released was migrated into the cathode chamber timely, which achieved a higher ammonia removal of 20.6% and CH_4 production of 14.6% compared to the traditional AD system. Cerrillo et al. (2021) coupled an AD-MEC system with ammonia recovery to digest pig slurry, and experimental results show that the combined system achieved a nitrogen removal efficiency of 31% and a productivity of 73 $L/m^3/d$ for CH_4 production. In addition to nitrogen recovery, the releasing rate of organic phosphorous in WAS could also be accelerated when MEC was integrated with AD for phosphorus removal and recovery (Zhou et al., 2019).

The concentration of VFAs is an indicator for the overall performance of AD, which are detected offline by gas chromatography or high-performance liquid chromatography with long response time and high cost. It is feasible for MEC to be integrated with AD as a biosensor for the real-time monitoring of VFAs, since VFAs released can be oxidized at the anode to release electrons to generate current for being detected immediately. Jin et al. (2017) developed a bio-electrolytic sensor to monitor VFAs online when synthetic wastewater was treated, and a good correlation ($R^2 = 0.99$) was observed between the current density and concentration of VFAs.

4.4. Economic performance analysis

No commercial applications have been reported so far for AD-MEC systems. Beegle and Borole (2018) analyzed the economic performance for the AD-MEC and AD systems using the water resource recovery facility at Oak Ridge National Laboratory with a capacity of 757 m^3/d to treat wastewater, which indicated that the AD-MEC system could be better with the 20-year net present value (NPV) of -$51,631.19 vs -$73,903.01 and the revenue of $1,904.14 vs $1,142.48, although its operating and maintenance cost of $5,007.19 would be slightly higher than that of $3,979.22 for the AD process.

However, capital investment on AD-MEC systems with bio-electrochemical devices is higher, and electricity is consumed to drive the MEC unit, which needs to be addressed properly.

5. Improvement on AD-MEC systems

AD-MEC systems are composed of MEC devices with electrodes and power supply as well as AD reactors, and improving their performance should be focused on enhancing the bioelectrochemical reactions through developing suitable materials for electrodes, fabricating efficient configurations for the electrodes, and employing applied voltages properly, together with the optimization of operating parameters for the anaerobic digestion such as OLR, HRT, pH, and temperature. Moreover, dynamics of the microbial community also needs to be considered, since it plays indispensable roles in improving the overall performance of AD-MEC systems.

5.1. Fabricating MEC

5.1.1. Electrode materials

The performance of AD-MEC systems is affected significantly by electrode materials, which should provide high surface area for electroactive bacteria to develop biofilms and perform catalytic behaviors mediated by the electron transfer more efficiently (Noori et al., 2020). Generally, electrode materials can be categorized into two groups: carbon- and metal-based materials (Park et al., 2020).

Carbon-based materials are dominated in the past decades because of their merits of excellent resistance to corrosion as well as large surface area and good biocompatibility for electroactive bacteria to be enriched as biofilms (Mier et al., 2021). Liu et al. (2020a) applied a carbon brush cathode for developing an AD-MEC system to treat wastewater, and evaluated its biogas production and upgrading performance, which obtained a high CO_2 reduction rate of 602 $mol/d/m^3$. Lin et al. (2019) investigated biogas production and phenanthrene biodegradation in the AD-MEC reactor with carbon paper as the cathode, and consequently, the maximum CH_4 yield of 113.5 L/kg (TS) and phenanthrene degradation rate of 52.4% were obtained, which are 30.5% and 83.6% higher, respectively, than that detected in the traditional AD system. Nonetheless, drawbacks of carbon-based electrode materials are poor mechanical strength for short durability, less conductive for significant resistance to electron transfer, and high cost for maintenance, which impede their utilization at large scales (Park et al., 2020).

Modifications have been made on traditional carbon-based materials to enhance their electrical conductivity, surface area, hydrophilicity, mechanical strength, and biocompatibility. For instance, Luo et al. (2014) utilized a carbon-black-modified cloth electrode for the bio-electrochemical system, reporting an increase of 2 folds in CH_4 production compared to the traditional graphite-fiber brush electrode. Redondo and Pumera (2021) modified the 3D-printed carbon-based electrode with archetypal MXene and Ti_3C_2, resulting in an enhancement of 3 folds in its capacitance.

Recently, metal materials, particularly stainless steel, have been developed as electrodes for AD-MEC systems. Major advantages of metal materials are low cost, good conductivity, and high mechanical strength for durability (Park et al., 2020). Xie et al. (2021) evaluated effect on treating *N*, *N*-dimethylacetamide wastewater by an AD-MEC system with different configurations of stainless-steel materials as the cathode, and found that the mesh with a size of 100 μm showed better COD removal efficiency, electrochemical performance, and biodegradability compared to the carbon cloth cathode. Liu et al. (2017b) studied heat-treated stainless steel belt as the cathode for an AD-MEC system, and compared to the graphite belt, the stainless steel belt gained better electrocatalytic properties for more efficient CH_4 production. In

addition to low cost and high durability, stainless-steel materials also own a merit for being fabricated into various configurations with high surface area for biofilms to be formed more effectively.

Major drawbacks of stainless-steel materials are corrosion in AD-MEC systems that are often operated with acidic environments, poor biocompatibility, and low surface area in comparison with carbon-based materials, which can be addressed by coating with carbon materials. An et al. (2020) developed a carbon-modified copper foam electrode by coating with multi-layer carbon nanotubes through electrophoretic deposition and screen-printing technology to improve the enrichment of electroactive bacteria and CH_4 production as well.

5.1.2. Configurations of the electrodes

The most important parameter for the electrodes is surface area, since high surface area can enhance the attachment of electroactive bacteria to form biofilms for more effective electron transfer. Arif et al. (2022) investigated the anaerobic performance of an AD-MEC system equipped with electrodes characterized by different surface area, and found that CH_4 production was increased from 316 to 361 mL/g (COD) when the electrode surface area was increased from 10 to 30 m^2/m^3 (reactor volume). Guo et al. (2016) investigated the effect of the ratio of cathode surface area over the volume of the anode on CH_4 production, and under the external voltage of 0.9 V, the CH_4 production of 0.14 $m^3/m^3/d$ was achieved when the ratio of 4 cm^2/cm^3 was applied for the stacked stainless steel mesh cathode and graphite fiber brush anode, which increased 1.8 folds compared to the system with the ratio of 1.0 cm^2/cm^3 for the electrodes.

Distance between the anode and cathode also plays a significant role in AD-MEC systems, since it affects the internal resistance by influencing the ionic diffusion (Park et al., 2020). Decreasing the distance would enhance CH_4 production and COD removal by reducing the internal resistance of AD-MEC systems, but when the electrodes are too close, short circuit may occur, and consequently compromises their performance (Hou et al., 2015). Rivera et al. (2017) maximized H_2 production and energetic performance for an AD-MEC system when a stainless-steel cathode of 71 cm^2 was installed with a distance of 4 cm to the anode.

5.1.3. External voltage

An external voltage can provide energy for AD-MEC systems, and thus electrochemical reactions that cannot occur spontaneously can proceed, since a suitable external voltage can increase DIET among the microbial community, enhancing the degradation of VFAs and other organic compounds to improve CH_4 production and biogas upgrading as well (Wang et al., 2022). However, negative effect can also be caused when higher voltage is applied, since the bulk solution near the anode can accumulate H^+ and acetate, leading to acidification, even a system collapse (Liu et al., 2020b).

Generally, a suitable voltage applied to AD-MEC systems ranges from 0.1 to 1.8 V (Park et al., 2020). Flores-Rodriguez et al. (2019) investigated the anaerobic performance of an AD-MEC system operated with an external voltage from 0.5 to 1.5 V, and a maximal CH_4 yield of 0.351 L/g (COD) was achieved at the applied voltage of 1.0 V. Choi et al. (2017) also found that among tested voltages of 0.5, 0.7, 1.0, and 1.5 V, the applied voltage of 1.0 V was optimal for CH_4 production at 408.3 mL/g (COD). Chen et al. (2016) treated WAS with an AD-MEC system operated at an external voltage from 0.3 to 1.5 V, and revealed that the applied voltage accelerated the hydrolysis and acidification of WAS, but the best performance was obtained at the applied voltage of 0.6 V. The optimal voltages reported by researchers for AD-MEC systems are different, due to differences with the inoculum levels, substrates, electrode materials, and reactor configurations.

5.2. Operating AD reactors

5.2.1. OLR and HRT

AD systems should be operated at high OLR and short HRT conditions to minimize their operating cost and improve efficiency. Nonetheless, traditional AD systems are prone to accumulate VFAs under high OLR conditions, which consequently decreases the pH value to inhibit the activity of methanogens severely. However, AD-MEC systems are more stable under high OLR conditions because produced VFAs can be utilized quickly through the external energy supply (Litti et al., 2022).

Lee et al. (2019) developed an AD-MEC system for being operated at different OLRs (0.25, 0.50, 1.00, and 1.25 g (glucose)/L/d), and experimental results showed that the maximal CH_4 yield of 0.34 L/g (COD) was achieved at the OLR of 0.5 g (glucose)/L/d. Huang et al. (2021) conducted an AD-MEC reactor to treat blackwater at different OLRs, and obtained a higher CH_4 yield at the OLR of 3 g COD/L/d. Park et al. (2019) investigated the effect of OLRs on CH_4 production in an AD-MEC system to treat food waste, which could operate stably at the OLR of 10 $kg/m^3/d$, 2.5 times higher than that of the traditional AD reactor.

Substrate can be degraded more completely through AD when HRT is sufficiently long. HRT is affected by many factors such as substrate composition and concentration, operating temperature, and reactor configuration (Khan et al., 2016). A disadvantage of traditional AD systems is an extremely long HRT of 15–40 d for different substrates, which consequently compromises their efficiency (Sathyan et al., 2022). Although the treating capacity of AD reactors can be enhanced by reducing HRT, the incomplete degradation of organic wastes can occur due to inadequate reaction time (Laçın et al., 2023). AD-MEC systems have been reported to be operated at shorter HRT in comparison with traditional AD, since the degradation of organic wastes can be enhanced by the supply of external electric energy, but the shorter HRT might result in a high flow rate that could lead to the damage of biofilms on the electrodes and the loss of functional microorganisms in the bulk solution as well.

5.2.2. pH and temperature

pH and temperature have a significant effect on the performance of AD-MEC systems. Normally, suitable pH for operating AD processes is from 6.5 to 8.5, because methanogens are sensitive to pH fluctuations, and most electroactive bacteria and methanogens are prone to thrive in neutral pH environments (Demirel and Scherer, 2008). While the accumulation of VFAs leads to a drop in pH for souring AD systems, AD-MEC systems have been reported to prevent the acidification through assimilating VFAs more efficiently, and thus could be operated at higher OLR conditions without significant acidification in comparison with traditional AD reactors (Huang et al., 2020).

Generally, AD-MEC systems are carried out under mesophilic temperature (25–40 °C) conditions (Huang et al., 2020). It had been proved that an applied voltage to mesophilic AD systems can shorten their HRT to increase OLR, which is partly due to the enrichment of hydrogenotrophic methanogens (Litti et al., 2022). For traditional AD reactors, hydrogenotrophic methanogens are thriving under thermophilic environments, and the better thriving of hydrogenotrophic methanogens under mesophilic conditions in AD-MEC systems make them possible to be operated at ambient temperature without additional heating cost to save energy consumption.

6. Prospects and challenges

AD-MEC systems have been developed for enhancing the degradation of organic wastes. Although numerous studies have been engaged to elucidate underlying mechanisms and optimize their configurations and operating parameters, bottlenecks of additional cost with the electrodes and energy consumption for driving AD-MEC systems have limited their applications at large scales.

Capital investment on AD-MEC systems with bioelectrochemical devices is higher than AD reactors, and electricity is consumed to drive their operation. Strategies have been proposed for addressing these challenges, such as developing cost-saving electrode materials, surface

modifications of the electrodes, and driving AD-MEC systems with cheap electricity.

Carbon-based materials and non-precious metals such as stainless steel are promising for developing cost-effective electrodes due to their tunable physical and chemical properties including resistance to corrosion, good conductivity, and biocompatibility. However, modifications through coating, etching with chemicals, or sintering at high temperature are needed for further increasing surface areas, and also creating rough surfaces and porous structures. As a result, electroactive bacteria can colonize more effectively onto the electrodes, even penetrate into their inner pores, to ultimately increase productivities of the MEC unit.

Moreover, mechanism underlying extracellular electron transfer within AD-MEC systems are still poorly understood, which is a prerequisite for AD-MEC systems to be operated stably and efficiently so that syntrophic bacteria can interact effectively with methanogens. Electron transfer between microorganisms and electrodes as well as among different species plays an important role for maintaining a syntrophic relationship within AD-MEC systems, but the complexity of microbial consortia, feeding substrates, and the fluctuation of operating conditions impedes research progress in this regard. Metaomics analyses at genome and transcriptome levels are expected to address this challenge in the near future.

AD and MEC can be integrated in situ as one reactor or separately as two-stage systems, but the in situ integration is more preferred, since such a design can enhance the AD process. Although energy consumption is needed for driving AD-MEC systems, it is not very intensive, since low voltage and current density are employed. Therefore, renewable energy provided by solar panels can be explored for such a purpose. In case biogas is co-fired for power generation, AD-MEC systems can be driven by the self-produced electricity to make their operation more economically competitive.

7. Conclusions

AD-MEC systems have been demonstrated in laboratories and pilot scales to be more effective and efficient than traditional AD processes in treating organic wastes, recovering energy through biogas production, and upgrading biogas in situ. Although commercial applications of AD-MEC systems have not been reported so far, model simulation and economic analysis indicate that their performance is better. With research progress in scientific fundamentals to further elucidate mechanisms underlying bioelectrochemical processes as well as technological innovations on developing novel electrode materials and configurations as well as bioreactor designs to optimize energy production with biogas and balance electricity consumption on driving the MEC unit, AD-MEC systems are expected to be improved continuously, and developed as a platform for treating organic wastes more efficiently and effectively to recover more energy through enhancing biogas production and upgrading the biogas product in situ.

Declaration of competing interest

The authors declare no conflicts of interest.

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