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Advances in Energy-Producing Anaerobic Biotechnologies for Municipal Wastewater Treatment

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ABSTRACT

Municipal wastewater treatment has long been known as a high-cost and energy-intensive process that destroys most of the energy-containing molecules by spending energy and that leaves little energy and few nutrients available for reuse. Over the past few years, some wastewater treatment plants have tried to revamp themselves as "resource factories," enabled by new technologies and the upgrading of old technologies. In particular, there is an renewed interest in anaerobic biotechnologies, which can convert organic matter into usable energy and preserve nutrients for potential reuse. However, considerable technological and economic limitations still exist. Here, we provide an overview of recent advances in several cutting-edge anaerobic biotechnologies for wastewater treatment, including enhanced sidestream anaerobic sludge digestion, anaerobic membrane bioreactors, and microbial electrochemical systems, and discuss future challenges and opportunities for their applications. This review is intended to provide useful information to guide the future design and optimization of municipal wastewater treatment processes.

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1. Introduction

Municipal wastewater treatment plants (WWTPs) play a key role in wastewater sanitation and the protection of public health [1]. However, the high economic and energy costs and the pollution transfer issues (from water to solids and/or air) of activated sludge processes make them unsustainable and increasingly unaffordable, especially with today's ever-tightening water and air emission regulations. Despite substantial modifications in reactors and processes over the years such as the development of membrane bioreactors [2] and aerobic granular sludge systems [3] and the optimization of process operations [1], the core strategy of activated sludge processes (i.e., destroying energy-containing molecules by spending energy) remains unchanged. To make a fundamental change toward resource recovery [4,5], revolutionary technologies and processes will have to be implemented. Anaerobic technologies are considered to be one of the most promising solutions.

Shifting from aerobic to anaerobic treatment of municipal wastewater offers an exciting opportunity to turn municipal wastewater treatment facilities into self-sustained operators or even net energy producers [6,7]. In contrast to the activated sludge process, which is energy intensive and resource wasteful, anaerobic processes avoid the energy consumption of aeration and produce an energy output instead [8]. Moreover, the nutrients in wastewater can be preserved to allow subsequent reuse or recovery [4], thereby further increasing energy and economic benefits. In conventional treatment processes, the need for carbon consumption prohibits the utilization of all the organic matter for anaerobic energy production. It is notable that such usage is now becoming possible due to the emergence of carbon-independent nutrient-removal biotechnologies [9].

Anaerobic wastewater treatment is not new. It has long been practiced in treating high-strength industrial wastewaters and sewage sludge [10]. In such processes, complex biosolids can be efficiently broken down by anaerobic microorganisms in the

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absence of oxygen, generating a methane-rich biogas for energy recovery and yielding a stabilized sludge that is suitable for land use [1]. These sludge-derived products can partially offset the high cost of the activated sludge process for municipal wastewater treatment; however, energy recovery is usually very limited because most of the organic matter is still wasted in the water phase. Therefore, improving energy production requires either partitioning more organic matter to the sludge phase for anaero-bic digestion (side-stream treatment) or directly treating the low-strength water anaerobically (mainline treatment)—a process that faces different technological challenges.

Side-stream sludge treatment through anaerobic digestion has been practiced for years; however, enhancing this process requires new technologies to enrich the organic matter content in sludge and improve the conversion efficiency of the sludge biomass. For the mainline anaerobic process, the slow growth and poor activity of anaerobic microorganisms have become a critical issue. Municipal wastewater is characterized by low organic strength, a significant percentage of particulate organic content, and frequently psychrophilic conditions, which are unfavorable for the growth of methanogens [11]. Therefore, these characteristics hamper the hold-up of dense biomass in conventional anaerobic bioreactors such as up-flow anaerobic sludge blanket (UASB) and expanded granular sludge bed (EGSB) reactors due to easy biomass washout, and deteriorate the overall treatment performance. Better anaerobic technologies and more efficient reactors are needed to address these challenges.

Here, we outline several representative energy-producing anaerobic technologies for future municipal wastewater treatment: bio-concentration and enhanced anaerobic sludge digestion for side-stream treatment; and anaerobic membrane bioreactors (AnMBRs) and microbial electrochemical systems (MESs) for mainline treatment [9]. We summarize recent advances in these biotechnologies and highlight remaining challenges and required future developments for practical application. This paper focuses exclusively on energy production and relevant anaerobic biotechnologies. Progress in anaerobic platforms for integrated energy and resource recovery from wastewater can be found in other review papers [9,12,13]. This review may provide useful information to guide the future design and optimization of municipal wastewater treatment processes and is intended to encourage more thinking and research on anaerobic wastewater treatment biotechnologies.

2. Enhanced side-stream anaerobic sludge digestion

2.1. Technological advances

Enhancing side-stream energy recovery through bio-concentration and sludge digestion is a relatively mature and low-cost technology. The process flow is similar to that of conventional activated sludge treatment, but relies more on anaerobic than aerobic degradation of organic matter. There are two key steps in this process: ① up-concentration of organic matter into sludge biomass at a minimal energy consumption; ② high-rate anaerobic digestion of the carbon-laden sludge to produce energy-rich biogas, as shown in Fig. 1(a). The bio-concentration of organic matter can be readily achieved through the adsorption, assimilation, and accumulation of sludge biomass at a very short sludge age and moderate aeration [14], while anaerobic digestion of the resulting sludge biomass is favored by the raised carbon content. Such a process has been successfully practiced in several WWTPs, including the Strass WWTP in Austria. In this plant, the contact stabilization process is adopted to partition most of the influent organic matter into sludge for anaerobic digestion [15].

The energy efficiency of such a process is usually limited by a slow solubilization of the organics from the sludge biomass. Thus, pretreatment is commonly applied to make organic matter more amenable to utilization by acidogens and methanogens [16]. Many pretreatment methods such as hydrothermal, microwave irradiation, ultrasound, mechanical shearing, chemical, and biological (enzymatic) pretreatment are effective in breaking down the sludge biomass, but are energy or cost intensive [17]. Methods that can utilize locally available low-value energy and resources are preferable. In this respect, thermal hydrolysis offers a useful option, since it can directly utilize the lower-value heat generated from the co-generator or heat pumps [17]. This in situ waste heat utilization, together with the significantly decreased volume of sludge slurry relative to the bulk sewage, makes it possible to reach a high temperature with minimal or even zero extra energy input. Nevertheless, the performance of such a pretreatment depends strongly on the bio-concentration level, sludge properties, and availability of heat energy, which may vary significantly with operating conditions. The most successful application case so far is the Blue Plains WWTP in the US. This plant adopts a similar side-stream anaerobic process to that used in the Strass WWTP, but adds a Cambi thermal hydrolysis process (with raised temperatures and pressures) to enhance biomass solubilization [15]. This setup doubles the methane yield as compared with a conventional sludge-digestion process.

Other frequently encountered problems are the low organic content and unbalanced composition of the obtained sludge, both of which lower methane production. Co-digestion of the sludge with other organic-rich wastes (e.g., food wastes) provides a feasible solution [18]. This solution not only raises the available carbon concentration but also balances the carbon/nutrient ratio, leading to an improved biogas yield and energy balance [19]. In addition, the utilization efficiency of anaerobic digesters can be improved, partially offsetting reactor investment and maintenance costs. This strategy has been proved useful for improving biogas production and has been successfully applied for over eight years at the Strass WWTP.

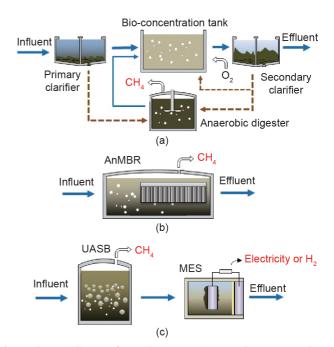


Fig. 1. Schematic diagram of several representative anaerobic energy-producing processes. (a) Enhanced side-stream anaerobic sludge digestion; (b) AnMBR; (c) the anaerobic digestion-MES integrated system.

2.2. Limitations and challenges

Although the feasibility of the side-stream anaerobic process has been demonstrated in full-scale WWTPs, its widespread application is still limited by several technological and economic barriers. First, the bio-concentration process still consumes oxygen and inevitably wastes a small amount of the organic matter. Secondly, the short solid retention times may lead to a poor settleability of sludge [20], which necessitates the addition of coagulants for sludge thickening or the installation of a membrane to avoid sludge washout [21,22]. Thirdly, the digestate contains rich organic matter (usually with the chemical oxygen demand (COD) >100 mg·L⁻¹) and is prohibited from direct discharge. Thus, it is either returned to mainstream reactors or subjected to downstream polishing by processes such as activated sludge with sufficient aeration and microalgae cultivation [23]. Lastly, anaerobic digesters and pretreatment devices are expensive and prone to abrasion or corrosion under suboptimal operation [24]. All these barriers lower the energy recovery efficiency, increase costs, or incur process instability for the overall process.

3. Anaerobic membrane bioreactors

3.1. Characteristics of AnMBR operation

While side-stream anaerobic sludge digestion involves a complicated process of pre-concentration to biosolids and release of the organic matter, this process could become much simpler if the low-concentration organic matter in the water phase were directly converted into energy under anaerobic conditions. Such a mainline anaerobic treatment is enabled by the recent development of AnMBR technologies [6]. An AnMBR is a highly compacted bioreactor that plays a dual role of contaminant removal and sludge separation [25], as shown in Fig. 1(b). Its excellent retention of sludge and particulate organic matter gives it a much higher treatment efficiency than other anaerobic bioreactors [26,27]. Another unique advantage of AnMBR is its good process robustness under climate-temperature conditions. Performance deterioration at low temperatures is a common challenge for most anaerobic processes due to significantly decreased microbial activity for solid hydrolysis and methane production [28,29], but is not a major concern for AnMBR. AnMBR can sustain a high sludge biomass concentration, especially for slow-growing and hydrolytic bacteria and methanogens, in order to compensate for suppressed microbial activity, effectively reject fine particles for sufficient hydrolysis [26], and maintain a good effluent quality at water temperatures down to 6 °C [30–32].

Therefore, AnMBR is an attractive technology for the mainline anaerobic treatment of municipal wastewater. However, there are several significant limitations to the practical application of this technology, including membrane fouling and the loss of dissolved methane, both of which are usually further aggravated by a low water temperature [33]. These limitations have become a focus of recent studies.

3.2. Membrane-fouling control

Its high biomass concentration (typically >10 000 mg·L⁻¹) results in an AnMBR treatment efficiency that is comparable to that of an aerobic process. However, it also increases membrane fouling due to the raised fluid viscosity and the presence of more bulk microbial cells and biomolecules [34], especially under psychrophilic conditions [35]. For example, the contents of soluble microbial products (SMPs) and fine particles in an AnMBR were found to increase markedly when the water temperature dropped

from 25 °C to 15 °C, causing severe membrane fouling [28].

Gas sparging by providing pressurized biogas has been widely used as an effective strategy to mitigate membrane fouling. For example, by applying continuous biogas sparging at rates of 40–60 m³·(m²·h)⁻¹ coupled with regular chemical cleaning (once every 3–4 months), membrane fouling was effectively suppressed in a pilot-scale AnMBR and the system was stably operated for over three years when treating municipal wastewater [32]. However, such a biogas sparging consumes energy of over 0.4 kW·h·m⁻³, about a third more energy than the recoverable biogas energy at 15 °C [27]. The need for better fouling control in AnMBRs has inspired intensive research efforts in reactor optimization and the development of low-energy fouling control strategies.

The fouling performance of an AnMBR is highly associated with the reactor type. A number of reactor configurations have been tested for AnMBR operation so far, including the completely stirred tank reactor (CSTR), UASB and EGSB reactors, and the fluidized bed reactor (FBR) [33]. CSTR was first used because of its ease of construction and operation. However, directly exposing the membrane to bulk sludge in a CSTR leads to severe membrane fouling [34]. Later, attached-growth bioreactors such as UASB [36] and EGSB [37] were considered. With the formation of granular sludge and the efficient physical entrapment of particulate organics in the sludge bed, spatial separation between the biodegradation zone and the membrane module in these reactors favors reduced biocake formation on the membrane surface. A pilot-scale AnMBR, which consists of a UASB and an external membrane unit, has been stably operated for over three years so far, with infrequent chemical cleaning [38]. Nevertheless, energy-intensive biogas sparging or fluid recirculation (0.25-0.5 kW·h·m⁻³) is still needed in these reactors to provide the necessary hydraulic shearing for fouling control [33,39]. Another concern is the instability of sludge granules during long-term operation, because the introduced membrane may eliminate the hydraulic selection pressure required for granulation and floc sludge washout [40].

One important recent breakthrough is the adoption of FBRs in AnMBR operation [41]. Here, instead of self-formed microbial granules, granular activated carbon (GAC)-supported granular sludge is used to reduce bulk floc sludge and improve membrane performance [42]. In addition, the fluidized GACs themselves can provide direct physical scouring to the membrane surfaces and reduce the foulants (e.g., SMP and extracellular polymeric substances (EPS)) in the bulk solution via adsorption, thereby further contributing to fouling mitigation. In this system, the energy consumption (mainly for GACs fluidization and bulk liquid recirculation) significantly decreased, down to about 0.02 kW·h·m⁻³ [43]. A pilot-scale AnMBR with GACs has been successfully and stably operated for almost two years to treat municipal wastewater, with no chemical cleaning [44]. In addition to FBRs, the anaerobic baffled reactor (ABR) was also reported to impart good anti-fouling performance to AnMBR. An ABR is composed of a series of horizontally connected UASB cells through which the wastewater traverses the whole reactor in a reciprocating pathway, while the solids are fully rejected when passing through the sludge blanket. Thus, the supernatant of an ABR contains few suspended particles, especially in the later cells [45]. This creates an ideal particlefree environment to minimize biocake development, significantly mitigating membrane fouling in an AnMBR when treating municipal wastewater [46].

Although reactor optimization in combination with appropriate operating modes can help mitigate fouling, during long-term operation, small-sized foulants still gradually build up on membrane surface [44]. Thus, several other low-energy fouling control approaches have also been developed, including the adoption of a shear-enhanced membrane design [47], the addition of

flocculants [48], enzyme augmentation [49], and electrochemical approaches.

The most straightforward approach for membrane-fouling control is physical cleaning, which can be realized by not only enhancing fluid turbulence but also adopting a new design of membrane modules. Kim et al. [50] designed an AnMBR with a rotary disk of sponge that can clean the membrane surface during the disk rotation. A vibratory membrane system offers another attractive option for low-fouling operation. By moving the membrane in a transverse direction to the fiber axis at a moderate vibration frequency, a high local shear rate and turbulence of the fluid can be created near the membrane surface to restrict cake formation [47]. Other shear-enhanced membrane designs include oscillation or rotation of the membrane [51]. For example, Ruigómez et al. [52] reported a novel rotating hollow-fiber membrane that showed more significant fouling mitigation than gas sparging (93%–96% versus 41%–44%). Nevertheless, providing the extra shear or membrane movement still requires considerable energy input. In addition, although such a shear enhancement can effectively reduce the deposition of large particles, it is less effective for colloids and soluble foulants.

Electrochemical and biological approaches were proposed to complement the reactor optimization for effective fouling control. In particular, electrochemical intervention offers an easily controllable and environmentally benign way to suppress membrane fouling compared with chemical approaches [53]. To make it simpler, Katuri et al. [54] directly coupled an MES into an AnMBR design by using electrically conductive hollow-fiber membranes as the cathode for hydrogen evolution reaction and as the membrane for the filtration of UASB effluent. With an electric energy input of 0.27 kW·h·m⁻³, the system yielded methane-rich biogas (83% methane), which was attributed to improved methane production stimulated by the hydrogen evolved at the cathode. Meanwhile, membrane fouling was significantly reduced as a result of scouring by the generated hydrogen gas bubbles as well as by a reduced accumulation of negatively charged bio-foulants at the low-potential cathode surface [55]. The fouling mitigation and energy balance were further improved by applying a graphene-coated membrane with a new rectangular reactor configuration to increase the hydrogen production [56].

Bio-fouling can also be dealt with by biological means such as adding enzymes and engineering the microbial interactions. The addition of exogenous hydrolases has been proved useful to improve the membrane performance of AnMBRs by providing structural disruptions of fouling layers and alteration of the sludge properties [57], but it is difficult to run sustainably. In general, dispersed hydrolases are prone to become deactivated or lost during long-term operation, while immobilized enzymes increase membrane resistance due to the accumulation of proteinaceous hydrolysis products in the immobilization layer [57]. Direct biological intervention may offer a better way by continuously generating enzymes or reducing biocake formation through quorum quenching [58]. This method has been successfully applied in an aerobic membrane bioreactor [59]. By adding quorum-quenching bacteriaentrapping beads, the energy consumption for membrane-fouling control was significantly reduced without compromising the effluent quality. Nevertheless, the feasibility of such biological control strategies for use with AnMBRs, which have different fouling mechanisms, still needs further investigation.

3.3. Dissolved methane recovery

Significant methane loss in the permeate presents another challenge for AnMBR operation [60]. The methane generated in anaerobic processes is only partially released into the gas phase,

while a considerable amount of methane (up to 38 mg·L⁻¹) remains in the liquid phase [44]. The methane loss in such a mainline treatment is more significant than that in side-stream digestion because more methane ends up in the effluent as a result of a lower methane production rate and lower water temperature. Such dissolved methane could count for as much as 88% of the total methane in an AnMBR (Fig. 2), resulting in severe energy loss and substantial greenhouse gas emission [61]. Thus, recovering this part of methane is essential.

A common way to remove methane from the water phase is bubbling with air or another gas in a bubble column aerator. Such an operation can remove the dissolved methane to a very low level, but it costs a great deal of energy and usually results in an over-diluted biogas that is unsuitable for power generation. In general, the methane fraction in the collected gas needs to be higher than 30% for practical electricity generation [62]. A more efficient way for methane stripping and recovery is applying a hollow-fiber membrane contactor. In this new design, a hydrophobic membrane is used to allow non-dispersive contact between the liquid and gas phases [63], leading to the significantly accelerated transfer of methane from the liquid to gas phases. With a very low energy input (< 0.002 kW·h·(m⁻³ water)), such systems can be operated at lower gas-to-liquid ratios to produce biogas with sufficient methane concentration (about 72%) for power generation [63]. In addition to using sweep gas as the driving force, vacuum extraction can also be used in combination with a hydrophobic hollow-fiber membrane to degasify the anaerobic effluent and recover burnable biogas [64]. The hydrophobic and nonporous membranes used in these systems can not only circumvent the membrane wetting problem that is easily induced by residual organic solutes but also reduce membrane module clogging caused by particulate matter [65]. However, they also suffer from a limited gas-transfer rate and thus need a long degassing time of up to 9.2 h, making their application constraining in practice.

Since the AnMBR penetrate contains a low concentration of organic solutes and is free of particulate matter, nonporous membranes seem to be unnecessary. Instead, a micro-porous membrane was found to be more suitable and efficient for treating AnMBR effluent, and was found to remove up to 97% of the dissolved methane [66] and yield a methane-rich biogas [61]. Thus, a micro-porous hollow-fiber membrane contactor may be a prom-

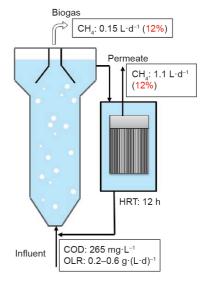


Fig. 2. Dissolved methane mass balance in an AnMBR [61]. HRT: hydraulic retention time; OLR: organic loading rate.

ising technology for addressing the dissolved methane issue for AnMBRs.

3.4. Challenges

Although the recent technology development implies that efficient dissolved methane recovery and membrane-fouling control can be achieved at low energy costs in lab-scale or pilot-scale AnMBRs, the practical feasibility, long-term performance, and economic aspects of these technologies for real municipal wastewater treatment at larger-scale facilities are still to be evaluated. Therefore, membrane-fouling control and dissolved methane recovery are still the key challenges for AnMBRs in wastewater treatment applications under climate conditions. In addition, our knowledge on AnMBR membrane-fouling mechanisms and influential factors are still very limited compared with those for aerobic systems. In particular, it remains unclear how the treatment performance and fouling behaviors of AnMBRs are affected by the microbial community and by sludge properties, which constrain system optimization.

4. Microbial electrochemical systems

MESs are a relatively new but attractive anaerobic biotechnology [67] for wastewater treatment. Unlike anaerobic digesters that mainly produce methane, an MES produces electrical energy or hydrogen gas using wastewater as a fuel, as shown in Fig. 1(c). Electricity and hydrogen are cleaner and more valuable forms of energy than methane and are not plagued by the problem of dissolved methane [68]. In an MES, organic matter is anaerobically degraded in the anodic chamber; the released electrons can be stored or directly utilized as electric energy through appropriate electric devices [69]. Meanwhile, the MES can produce an effluent with a comparable quality to that of aerobic treatment if given sufficient treatment time [70]. Thus, the MES is widely envisaged as a promising technology to achieve the energy-neutral operation of wastewater treatment facilities.

4.1. Technology advances

The past decade has seen intensive studies and significant progress in improving the electrochemical performance of MESs by approaches such as optimization of reactor configuration, separator materials, electrode materials, and microbial community [71]. However, its practical implementation for municipal wastewater treatment is still limited by a low power density, relatively high cost, and difficulty in scaling up [72]. An MES typically has a lower energy output than a methanogenic digester; however, with a capital cost that is two to three orders of magnitude higher, the MES is economically uncompetitive in wastewater treatment applications. Scaling up an MES, either by increasing the geometric size of an individual cell or by connecting multiple cell stacks, usually leads to significantly increased energy losses and to power density decline [73]. For example, a 100 L MES was successfully run in England to treat raw municipal wastewater with simultaneous hydrogen production [74]. This system showed stable treatment performance over one year of continuous operation, but recovered less than half of the electrical energy input. In another pilot MES for municipal wastewater treatment, net electric energy production was achieved, but the power density was still too low to have any practical use [75]. Changing the carbon brush to a GAC-packed bed electrode was shown to further improve the power generation, due to enhanced biofilm growth and mass transfer-through [76]; however, the energy performance was still un-comparable with that of anaerobic digestion.

One important reason for the inferior performance of an MES compared with an anaerobic digestion lies in the different microorganisms. Unlike anaerobic digestion processes, where the efficient hydrolysis of organic solids is enabled by the hydrolytic bacteria abundantly present in anaerobic reactors, an MES selectively enriches exoelectrogens [69], which prefer soluble volatile acids as a substrate and which are incapable of particle hydrolysis [77]. Therefore, the available substrate for exoelectrogens in an MES is usually limited, and the slow mass transfer within the exoelectrogen biofilm further constrains the electrochemical performance—resulting in a low power density and effluent quality when treating raw municipal wastewater [78]. A possible solution is to combine MES with anaerobic digestion processes, thereby allowing a better play of its power-generating role while circumventing the inherent limitations [72,79].

The good synergy between MES and anaerobic digestion through the intimate collaboration of multiple microbial species for improved municipal wastewater treatment and methane production has been demonstrated in several recent studies [78,80]. Anaerobic digestion allows improved hydrolysis of the organic particles, providing more available substrate for the MES (Fig. 3). In turn, the MES process could prevent the accumulation of inhibitory intermediates, thereby releasing the feedback inhibition to acidogens and meanwhile obtaining electricity as an extra energy gain [72]. The introduction of an MES could even significantly improve the methane production from anaerobic wastewater digestion by 5.3-6.6 times [81], likely due to the extra electrochemical hydrogen evolution simulating the methane production [82]. To further improve effluent quality and process stability, membrane processes can also be incorporated [83]. These findings suggest that, instead of serving as a standalone technology, an MES might be better integrated with an anaerobic digestion process to maximize the energy recovery from municipal wastewater.

4.2. Challenges

Despite its success in laboratory-scale studies, in order for the MES to become a practical wastewater treatment technology, many of the economic and technological issues around its scaling up must be addressed [73,84]. Cost is another critical issue. The current cost of an MES, due to the use of expensive electrode materials, membranes, and reactors, is approximately 100 times that of a conventional anaerobic digester, making the generation of a small amount of electricity in such systems insufficient to justify their cost [85]. In addition, there are stability issues such as the clogging of electrodes and membrane fouling, during long-term operation for practical wastewater treatment [72].

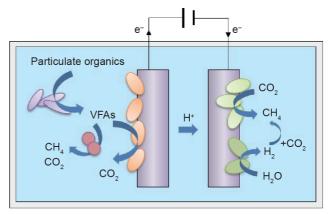


Fig. 3. Synergy between an MES and an anaerobic digestion process for maximized methane production with multiple microbial species involved. VFAs: volatile fatty acids

Another concern is the poor utility of the obtained bioelectricity: The power output from an MES is typically not high or sufficiently stable to drive a practical electronic device. Hence, it is insufficient to support the self-sustained operation of a WWTP [71]. It is essential to boost the power output to a usable level by applying more efficient power capture and storage devices [86] in order to find suitable niches for the in situ application of such low-power bioelectricity [87]. In a recent study, a capacitor-based circuit was incorporated into an MES; the circuit significantly raised the voltage output in order to successfully power intermittent pumping and aeration in a wastewater treatment bioreactor [88]. In addition, bioelectricity has been successfully used to mitigate membrane fouling in an MES-membrane bioreactor integrated system [89], to enhance photocatalytic decontamination by retarding the recombination of photo-excited electrons and holes [90], and to achieve heavy metal removal through cathodic reduction [91]. However, the energy efficiency of such hybrid systems is generally low and their application for real municipal wastewater treatment is still lacking.

5. Future perspectives

The extraordinary recent advances in anaerobic biotechnologies, together with other complementary low-energy treatment technologies, are causing energy-neutral and sustainable municipal wastewater treatment to approach reality. However, while side-stream anaerobic technologies are already in the early stages of practical application, other technologies such as AnMBR and MES are still in pilot-scale testing. In addition, many technological and economic challenges are yet to be addressed regarding full-scale widespread applications; these challenges call for further technological breakthroughs and for research efforts in the following directions.

The enhanced side-stream anaerobic sludge digestion process is a relatively mature technology that is likely to gain more widespread application in the next 5-10 years. At the core of this technology is the use of a contact stabilization process and an efficient dewatering system to obtain organic-rich, concentrated sludge for digestion; a co-generation system for burning the biogas to generate power and heat; and a thermal hydrolysis system for sludge thermal pretreatment by utilizing the in situ available heat. These processes bring about the multiple benefits of improved methane production, decreased volume and investment cost of anaerobic digesters, and better-quality sludge products. However, all these devices are expensive, and the processes involve considerable energy or chemical input. In particular, the complexity of municipal wastewater may make it difficult and costly to obtain high-quality sludge products and purified biogas purification for co-generation. Therefore, the development of low-cost devices and technologies will be the most important direction for boosting applications of the enhanced side-stream anaerobic sludge digestion process.

One promising way to improve energy production and economic return is to adopt co-digestion by adding external organic wastes into the digester. Such a strategy has been adopted by the Strass WWTP and has more than doubled the methane production rate. However, co-digestion may complicate the digestion process and may even introduce new problems if not kept under proper control. For example, the addition of many food wastes may lead to elevated concentrations of sulfur and hence to a higher fraction of hydrogen sulfide in biogas—necessitating extra treatment [18]. Another potential alternative for improving economic feasibility is to recover other higher-value products from sludge such as bio-oil, biochar, or other functional materials through pyrolysis [24,92]. Future investigation into these areas may bring about a new technological breakthrough.

AnMBR is a highly simple and compacted process that directly extracts wastewater energy and yields an effluent with low suspended solids and pathogens, making it suitable for decentralized municipal wastewater treatment and water-reuse systems [6]. However, its full-scale application has not been realized so far. Future development of this process may rely on the development of scalable and robust dissolved methane recovery technologies and better anti-fouling membrane and reactor systems.

With current technologies, around 50% of the organic energy in municipal wastewater can be converted to methane in an AnMBR; of this methane, half is lost in the effluent. Therefore, there is still plenty of room for energy recovery improvement. In particular, much work remains to be done to develop low-cost degassing technologies and to evaluate their performance in field studies. Another unaddressed issue is membrane-fouling control. Current ongoing research directions include: the incorporation of functional nanomaterials, such as carbon nanotubes, metal nanoparticles, and zeolites, into the membranes [93,94]; the application of quorum-quenching enzymes [95] or a turbulenceintensifying strategy [47]; and the utilization of bioelectricity to prohibit bio-fouling [89]. Future progress in these areas may ultimately allow for well-tailored membranes that not only efficiently separate contaminants from water but also actively clean themselves. In addition, replacing pressure-driven membranes with forward osmotic membranes presents another promising approach to address the membrane-fouling issue, and has already drawn considerable research interest [96].

MES is likely to be utilized as a complementary treatment to anaerobic digestion for enhanced energy recovery or to other electrochemical/photochemical processes for enhanced pollutant removal [13]. However, to make it economically practical, efforts will be needed to further lower the material costs and improve the energy efficiency—especially in scaled-up systems. Future field studies may deliver key information to guide technological development toward real-world applications. Another compelling application would be to utilize bioelectricity for the generation of specific high-value products in a process called microbial electrosynthesis [82]. For example, with an MES, the volatile fatty acids produced in anaerobic digestion may be readily converted to methanol, a higher-value fuel that can be separated and transported more easily [97].

It is important to note that maximizing carbon utilization for energy recovery occurs when carbon-independent nutrient-removal/recovery processes become available. Therefore, in addition to advances in the energy-producing anaerobic biotechnologies themselves, advances in low-energy nutrient-removal/recovery technologies are of critical importance in ensuring successful implementation of the overall processes. These technologies include anaerobic ammonium oxidation [98]; denitrifying anaerobic methane oxidation [99,100]; and the sulfate reduction, autotrophic denitrification, and nitrification integrated process [101].

Realizing the technological development and process optimization described above entails a better understanding of the microbial ecology in different systems and an optimized process control. We currently have very limited knowledge of the fundamentals of these novel systems in biological processes such as the functional and spatial relationships among hydrolytic bacteria, acidogens, and methanogens in AnMBR; interactions between non-exoelectronic and exoelectronic microorganisms in MES; and microbial dynamics in response to environmental changes. For example, it is unknown how the enforced vibratory shear in an AnMBR or the applied electrode potential will affect microbial physiology, metabolism, and inter-species interactions. The use of "omics" approaches and other culture-independent techniques

may offer better insights into these biological processes and their links with environmental conditions [102,103]. In addition, advances in instrumentation and sensor technology, in combination with the development of specific process models, will be needed to provide *in situ* process monitoring and risk diagnosis, and to allow improved control strategies for preventing process upsets. In particular, models for the emerging anaerobic processes are still scarce.

Lastly, the social, cultural, and political constraints on the implementation of new technologies will have to be considered. These issues may include the safety of reclaimed water and other products, carbon footprints, and social impacts [104]. Thus, life-cycle assessments will be needed to evaluate and aid the design of each overall process, and it will be necessary to combine researchers' efforts with support from the government and the public in order to make these processes into a practical reality.

6. Conclusions

The goal of achieving energy self-sufficiency in municipal wastewater treatment has spurred tremendous research efforts to develop more efficient energy-producing anaerobic biotechnologies. There are currently two dominant anaerobic energy-producing platforms: the enhanced side-stream anaerobic sludge digestion and mainline treatment with an AnMBR or an MES. These cutting-edge biotechnologies, in combination with low-energy nutrient-removal/recovery processes, offer an exciting opportunity to realize truly sustainable municipal wastewater treatment. However, many of these technologies are still immature. Bringing them into practical application in WWTPs will require further advancements to make them efficient, reliable, cost-effective, and scalable, and they must also overcome social constraints.

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Compliance with ethics guidelines

Wen-Wei Li and Han-Qing Yu declare that they have no conflict of interest or financial conflicts to disclose.

Nomenclature

ABR anaerobic baffled reactor
AnMBR anaerobic membrane bioreactor
COD chemical oxygen demand
CSTR completely stirred tank reactor
EGSB expanded granular sludge bed
EPS extracellular polymeric substances
FBR fluidized bed reactor
GAC granular activated carbon
HRT hydraulic retention time
MES microbial electrochemical system
OLR organic loading rate
SMP soluble microbial product
UASB up-flow anaerobic sludge blanket
VFA volatile fatty acid

WWTP wastewater treatment plant

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