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Anaerobic membrane bioreactor towards biowaste biorefinery and chemical energy harvest: Recent progress, membrane fouling and future perspectives

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ABSTRACT

Anaerobic membrane bioreactor (AnMBR) holds great promise to treat a broad range of waste streams for concurrent pollutants transformation and biofuels harvest while producing less digestate residuals. In this review, recent research advances, new discoveries and commercial application status of AnMBR technique were summarized and reported. A comprehensive comparison analysis designed herein demonstrated its fascinating superiorities over the conventional activated sludge-based processes with regards to good permeate quality, less digestate residuals, low operational costs, net profit/energy output, and outstanding economic and environmental benefits. Despite the great progress achieved previously, there are still numerous challenges head for AnMBR platform applications to be tackled, particularly for severe membrane fouling, low methane content in biogas, highly dissolved methane, poor ammonia removal and phosphorus recovery, etc. To address the above problems, a new-generation process, i.e. so-called "Integrated Multistage Bio-Process (IMBP)" constituted of solar-driven bioelectrochemical system (BES)-AnMBR, partial nitritation/anammox (PN/A), nitrate reduction via anaerobic oxidation of methane (AOM) and biological/chemical phosphorus precipitation units, was proposed in this article, with versatile capabilities in simultaneous biowastes valorization, CO_2 electromethanogenesis and simultaneous biogas upgrading, *in-situ* fouling control, ammonia removal, dissolved methane reutilization, and phosphorus recover as hydroxyapatite-rich nutrients. Despite the uncertainties about whether this approach possesses the powerful potential to dominate the future, but most surely, this hybrid concept will enhance the deployment and industrial competitiveness of AnMBR-based technologies in real-world scenarios, facilitating the establishment of the energy-sustainable and low-carbon society. Of course, more efforts are still required to demonstrate the feasibility of this integrated biorefinery approach. Nonetheless, this review opens up new research opportunities to integrate with other newly emerging processes to develop robust, multifunctional and sustainable AnMBR-based technologies towards biowaste biorefinery, chemical energy harvest and green, carbon-neutral society.

1. Introduction

Rapid industrialization and ever-rising energy demand have urged the need of exploring alternative power sources to alleviate energy shortage and crisis [1]. Biowastes, generated during the processes of people's life and production activities, represent a typical renewable source of energy in consideration of high contents of nutrients and readily biodegradable organic substances. To achieve its resource utilization and safe disposal is a major livelihood issue related to environmental safety and the healthy development of human society. Thus, considerable interest has been attracted concerning the reuse of biowastes to produce bioenergy [2,3]. Anaerobic digestion (AD) is

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List of abbreviations

AD anaerobic digestion
AeMBR Aerobic membrane bioreactor
ANAMMOX anaerobic ammonium oxidation
AnMBR anaerobic membrane bioreactor
AOB aerobic ammonium oxidizers
AOM anaerobic oxidation of methane
BES bioelectrochemical system
CAS conventional activated sludge process
CEPS chemically enhanced primary settling
COD chemical oxygen demand
CSTR conventional continuous stirred tank reactor
DS dry solids
EPS extracellular polymeric substances
ES excess sludge
HRT hydraulic retention time
IMBP integrated multistage bio-process
LB-EPS loosely bound EPS
MCRT microorganisms retention time
MF microfiltration
NOB nitrite oxidizing bacteria
OLR organic loading rate
PN/A partial nitritation/anammox
PS primary sludge
S-EPS soluble EPS
SMP soluble microbial products
SRT solids retention time
SS suspended solids
TB-EPS tightly bound EPS
TH thermal hydrolysis
TMP transmembrane pressure
TS total solids
UF ultrafiltration
VS volatile Solids

Units

Temperature °C
Time day (d), hour (h)
Biogas (methane) yield L/g-VS_{added}, L/g-COD_{added}
Transmembrane pressure kPa
Flux $L/m^2/h$
Membrane resistance m^{-1}
Energy consumption MJ/d

Nomenclature

J the permeate flux
V the volume of permeate
A the membrane area
$\triangle P$ the transmembrane pressure
μ the viscosity of water
R_t the total membrane resistance
R_m the intrinsic membrane resistance
R_c the cake layer resistance
$R_{p\text{-}org}$ the organic pore blocking resistance
$R_{p\text{-}inorg}$ the inorganic pore blocking resistance
Y the yield coefficient of activated sludge

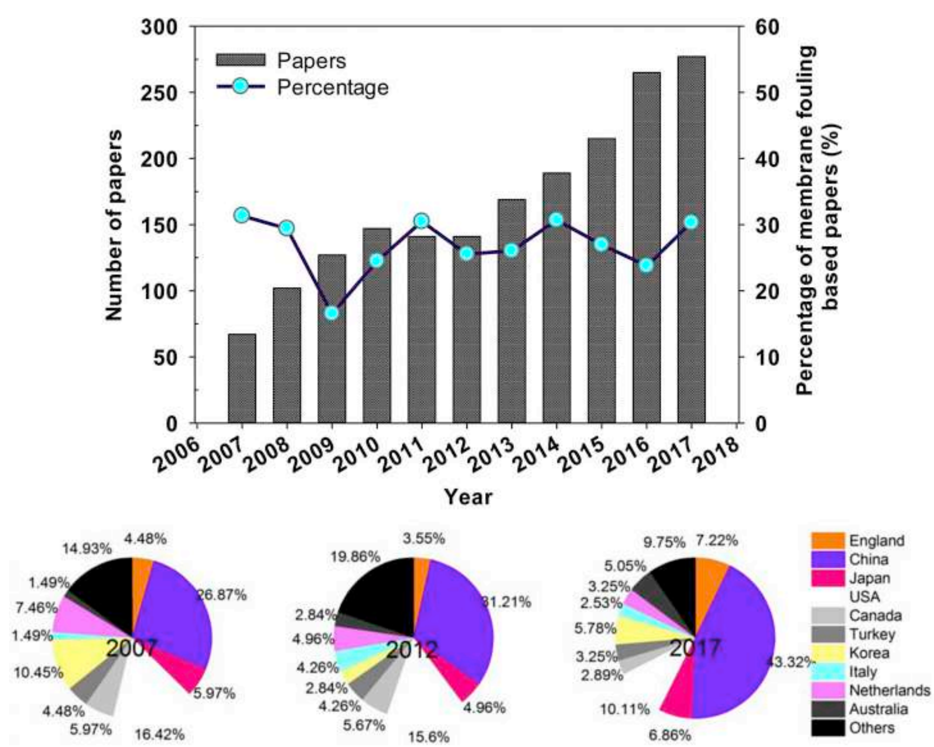

Fig. 1. Web of Science bibliometric study with the topics "AnMBR and membrane fouling", and proportion of research papers associated with AnMBR studies published in different countries (China, USA, England, Japan, etc.).

currently one of the most promising processes used for the treatment of various organic waste-streams (i.e. industrial and municipal wastewaters, waste activated sludge, food waste, livestock manure, algae, agricultural crops, etc.) [4,5]. It offers important advantages over the conventional aerobic biological treatment such as ease of operation, low sludge production, high treatment efficiencies, low energy demand, and energy production in the form of methane-rich biogas [6–10]. However, the biggest difficulty for AD process lies in the poor retention of slow-growing anaerobic biomass [11,12]. During this process, solids retention time (SRT), microorganisms retention time (MCRT) and hydraulic retention time (HRT) cannot be completely separated, which leads to the loss of biomass, slow-growth of key anaerobic microorganisms, low energy conversion efficiencies [8,13,14], and a longer time required to achieve full biostabilization [15]. Long SRT (i.e. 20–30 d [5]) is usually needed to ensure the proliferations of anaerobic biomass and full degradation of organics, which inevitably requires larger reactor volume [16,17]. Reversely, shortening SRT will cause the loss of anaerobic microorganisms, affecting the overall operational efficiency. Also, AD process is more sensitive to variations in the operating circumstance (e.g. temperature, pH, C/N ratio, salt concentrations and certain accumulating chemicals such as VFAs, ammonium ion (NH_4^+) and free ammonia (NH_3)) [18,19]. In addition, digestate has been also proven to be problematic due to its large quantities, deteriorated dewaterability, formation of odor gas (i.e. H_2S, and NH_3) and limited utility for land application [3,20]. Therefore, the application of anaerobic process is not widespread in comparison with the aerobic process.

Aerobic membrane bioreactor (AeMBR), as a new-generation technology, has been commercially implemented in industrial/municipal wastewaters treatment. Anaerobic membrane bioreactor (AnMBR), derived from AeMBR, represents a new research direction. AnMBR can effectively retain key microorganisms (e.g. methanogens) inside the reactor by direct membrane interception [21], realizing the complete separation of SRT and HRT. Namely, AnMBR can operate at shorter HRT with longer SRT. Accordingly, AnMBR can provide many economic and environmental advantages over the conventional AD, such as smaller footprint, lower production of digestate, and especially higher biomass retention, which enable higher organic matter biodegradation efficiency, less sludge production, increased bioenergy recovery, and more excellent effluent quality [10,22]. Because of the superiority of AnMBR, this technology has attracted substantial interests in the past decades for the treatment of a variety of waste streams with a broad spectrum of organic loadings, such as wastewater, sewage sludge, food waste, coffee grounds, livestock manure and high-strength landfill leachate to recover resources or biofules. With the success of lab-scale studies, more and more pilot-scale AnMBRs studies have recently been undertaken [23]. However, the practical applications of AnMBR technology so far still remain a big challenge due to serious technical issues, especially membrane fouling [24–26]. Membrane fouling causes the permeate flux decline, increased trans-membrane pressure (TMPs) and poor effectiveness, and thus has drawn the ever-increasing attention of both academia and industry [22,26,27]. The Web of Science shows that the number of peer-reviewed publications with AnMBR as topic is on the rise over the past 10 years (Fig. 1), i.e. only 67 papers published in 2007, over 150 papers per year since 2013 and up to 277 papers in 2017. Among the publications, the proportion of papers related to membrane fouling/cleaning fluctuates between 20% and 40%. Obviously, membrane cleaning represents an essential part of the applications of AnMBR. Unfortunately, the current work can only mitigate the extent of membrane fouling, whereas the technical issue cannot be effectively ev + + aded due to the effects resulting from multiple factors such as operation conditions (HRT, SRT, temperature, flux, etc.), properties of waste streams (e.g. soluble microbial products (SMP), extracellular polymeric substances (EPS), particle size, surface charge, etc.), and characteristics of membrane (i.e. types, materials, pore size, etc.) [8,22,28]. Therefore, large numbers of researchers have been exploring the relationship between these factors and membrane fouling in order to determine the best operating conditions for membrane fouling control. The rapid development of AnMBR technology have resulted in a diversity of novel configurations and new findings, and thus a comprehensive and the state of the art overview could be valuable to update the recent research process. Also, the superiority of AnMBR in sludge reduction, energy demand and environmental cost-benefits yet needs to be well documented, especially over the conventional activated sludge (CAS) processes.

This review was an attempt to summarize recent advances and practical applications of AnMBR and identify the main filed that needs further research. The operation conditions and long-term performance of AnMBR in treating different substrates were reviewed and summarized. Meanwhile, the mechanisms of membrane fouling, key influential factors and current progress in control measures' development were illustrated, and the application status of AnMBR was described as well. In addition, AnMBR system was evaluated and compared with other four modified activated sludge (CAS) processes in terms of operational performance, bioenergy recovery, economic benefits and environmental impacts. Finally, a new-generation wastewater treatment process was developed and the main conclusions and the future perspectives were proposed.

2. AnMBR configuration

AnMBR, made up of two parts: conventional anaerobic biological reactor and membrane models [29,30], is often divided into three categories: (a) side-stream AnMBR, (b) internal/submerged AnMBR, and (c) external submerged AnMBR (Fig. 2). Excellent reviews regarding

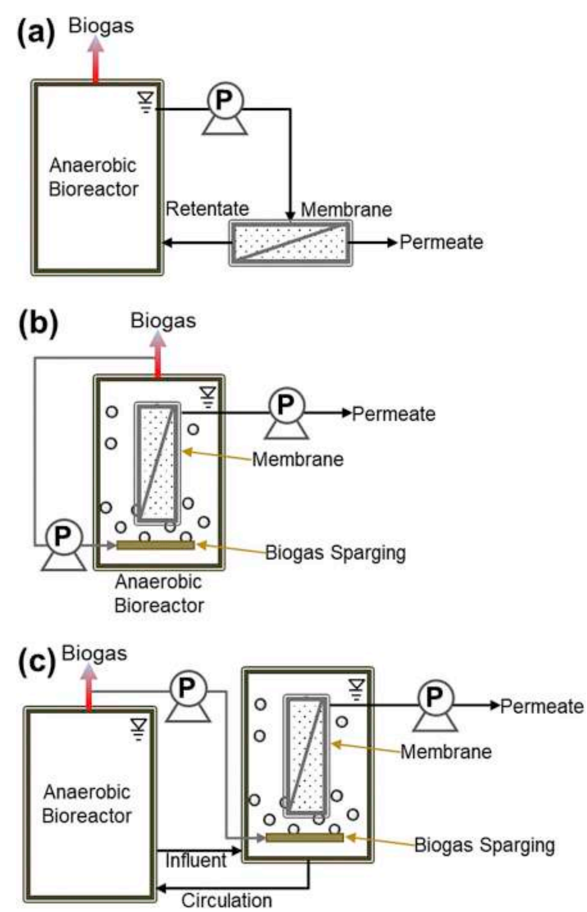

Fig. 2. Basic configurations and working principles of anaerobic membrane bioreactor (AnMBR): (a) external AnMBR, (b) internal/submerged AnMBR, and (c) external submerged AnMBR.

Table 1
Operational and performance data of different-scale AnMBRs fed with different feedstocks.

Feedstock	Sampling place	Properties	Configuration	Operational conditions	Performance	References
Sewage	Japan	TCOD 0.49 ± 0.11 g/L, TBOD 0.29 ± 0.09 g/L	Submerged, flat-sheet MF CPE membrane, working volume 6 L, surface area 0.116 m^2, pore size 0.2 µm	T 20 °C, sparging rate 5 L/min, HRT 12 h, SRT 650 d	COD removal 96%, BOD removal 97%, 269.5 mL-CH_4/g-COD_{added}	[74]
Pharmaceutical wastewater	China	TCOD 4.43 ± 0.83 g/L, TBOD 1.31 ± 0.24 g/L	External, total volume 188 L, working volume 180 L, PVDF hollow-fiber membrane, surface area 1 m^2, pore size 0.02 µm	T 14–38 °C, HRT 23.9 ± 1.2 h, SRT 250 d, OLR 4.46 ± 0.87 g-COD/L/d	TCOD removal 90.3 ± 1.5%, 262.7 ± 28.7 mL-biogas/g-COD_{added}, 117.0 ± 19.0 mL-CH_4/g-COD_{added}	[75]
Brewery wastewater	Canada	TCOD 11.1 ± 2.7 g/L, SCOD 10.2 ± 2.3 g/L, TSS 0.95 ± 0.45 g/L, VSS 0.77 ± 0.39 g/L	Submerged, working volume 15 L, hollow-fiber UF membrane, surface area 0.047 m^2, pore size 0.04 µm	T 35 °C, SRT 30 d, HRT 44 h, OLR 3.5–11.5 g-COD/L/d, scouring rate 15.3 m^3/h/m^2	COD removal > 98%, 524.7 ± 13.5 mL-biogas/g-COD_{added}, 309.6 ± 8.0 mL-CH_4/g-COD_{added}	[76]
Landfill leachate	Tunisia	TCOD 84.29 g/L, $TBOD_5$ 46.1 g/L, TSS 1.97 g/L, VSS 1.46 g/L, NH_4^+-N 2.8 g/L	External, working volume 50 L, flat-sheet Plexiglass UF membrane, surface area 1 m^2, pore size 0.1 µm	T 37 °C, HRT 7 d, OLR 7 g-COD/L/d	COD removal 93.97 ± 1.85%, 366.5 ± 7.2 mL-biogas/g-COD_{added}	[77]
Landfill leachate	Iran	TCOD 68.3 ± 8.0 g/L, TBOD 44.5 ± 3.0 g/L, NH_4^+-N 1.47 ± 0.09 g/L, PO_4^{3-}-P 130 ± 40 mg/L	Submerged, working volume 175 L, hollow-fiber membrane, pore size 0.1 µm	T 21 °C, SRT 55 d, HRT 15 d	COD removal 97.46%, NH_4^+-N removal 99%, PO_4^{3-}-P removal 90%	[52]
MBR excess sludge	Japan	TCOD 21.2 ± 1.3 g/L, TS 15.0 ± 1.2 g/L, VS 12.7 ± 1.0 g/L	External, working volume 20 L, PTFE tubular MF membrane, surface area 0.06 m^2	T 35 °C, CFV 0.7 m/L, SRT 700 d, HRT 34 d, OLR 2200 ± 200 mg-COD/L-reactor	COD removal 98%, 80 mL-biogas/g-COD_{added}, 51.6 ± 1.4 mL-CH_4/g-COD_{added}	[59]
Sewage sludge	USA	Sample 1#: TS 24.2 ± 3.3 g/L, VS 19.5 ± 3.4 g/L, TCOD 36.89 ± 11.68 g/L, SCOD 6.46 ± 1.30 g/L; sample 2#: TS 24.0 ± 3.2 g/L, VS 18.1 ± 0.4 g/L, TCOD 39.50 ± 5.94 g/L, SCOD 7.24 ± 0.88 g/L	Submerged, tubular MF membrane; acid-phase: total volume 2.5 L, working volume 1.5 L; methane-phase: total volume 14 L, working volume 12 L	Acid-phase: OLR 8.03 ± 0.5 g-COD/L/d, HRT 2 d, SRT 2 d, T 37 ± 3 °C; methane-phase: OLR 1.09 ± 0.2 g-COD/L/d, HRT 16 d, SRT 30 d, T 37 °C	COD removal > 90%, 261.6 ± 14.1 mL-CH_4/g-COD_{added}	[78]
Food waste	China	TCOD 2–15 g/L, SS 0.2–1 g/L	External, total volume 0.5 m^3, working volume 0.4 m^3, PES flat-sheet UF, surface area 0.32 m^2, MWCO 20000 Da, VLR 0–4.5 kg/m^3/d	T 37.0 ± 0.5 °C, HRT 60 h, SRT 50 d, CFV 1.02–1.09 m/s	COD removal 81.3–94.2%, 136 mL-biogas/g-COD_{added}	[79]
Food waste	South Korea	TS 45 ± 3 g/L, VS 43 ± 2 g/L, TCOD 52.7 ± 2.8 g/L, carbohydrate 17.7 ± 0.4 g/L, protein 11.4 ± 0.5 g/L, TN 2.22 ± 0.25 g/L, TP 176 ± 16 mg/L	Total volume 5 L, flat-sheet PE membrane, surface area 0.1 m^2, pore size 0.45 µm	T 55 ± 0.5 °C, biogas circulation rate 3.5 L/min, HRT 10.5 h, HRT/SRT 0.25, OLR 125.4 g-COD/L/d	Carbohydrate removal > 97%, protein removal 95.7%, 90.7 mL-H_2/g-COD_{added}	[61]
Coffee grounds	Japan	TS 347.0 ± 2.9 g/L, VS 344.0 ± 4.6 g/L, TCOD/TS 1.60 g/g	Submerged, total volume 15 L, working volume of 7 L, flat-sheet CPE MF membrane, surface area 0.116 m^2, pore size 0.2 µm	T 55–57 °C, flow rate 5 L/min, OLR 5.23 g-COD/L/d, HRT 30 d	382.4 mL-biogas/g-COD_{added}, 153.0 mL-CH_4/g-COD_{added}	[2]
Coffee grounds and sludge	Japan	Coffee ground: TS 347.0 ± 2.9 g/L, VS 344.0 ± 4.6 g/L, TCOD/TS 1.60 g/g;	Submerged, total volume 15 L, working volume 7 L, flat-sheet	T 55–57 °C, flow rate 5 L/min, OLR 23.6 g-COD/L/d, HRT 10 d, SRT 10 d	COD removal 44.5%, 245.8 mL-biogas/g-COD_{added}, 136.6 mL-CH_4/g-COD_{added}	[72]

(*continued on next page*)

Table 1 (*continued*)

Feedstock	Sampling place	Properties	Configuration	Operational conditions	Performance	References
		sludge: TS 158.0 ± 4.5 g/L, VS 124.0 ± 5.7 g/L, TCOD/TS 0.98 g/g	CPE MF membrane, surface area 0.116 m^2, pore size 0.2 μm			
Swine manure	China	TCOD 13.48 ± 2.24 g/L, SCOD 3.71 ± 0.24 g/L, TS 10.85 ± 2.64 g/L, VS 7.95 ± 2.16 g/L, TN 0.74 ± 0.02 g/L	Submerged, total volume 3.5 L, working volume 3.0 L, a PVDF membrane and an external hollow-fiber membrane, surface area 0.047 m^2, pore size 0.04 μm	T 35 ± 0.5 °C, biogas circulation 5.5 L/min, OLR 1.17 ± 0.19 g COD/L/d, SRT 45 d, HRT 13 d, flux 3–5 L/m^2/h	COD removal 96 ± 1.1%, 225.4 ± 74.9 mL-CH_4/g-COD_{added}	[21]
Liquid dairy manure	USA	TCOD 41.80 ± 3.50 g/L, TS 33 ± 2 g/L, VS 22 ± 1 g/L	External, working volume 105 L, tubular PVDF UF membranes, surface area 0.55 m^2, pore size 0.03 μm	CFV 4.3 m/s, SRT 24 d, HRT 12 d, cycle time 312 μHz	COD removal 41%, VS removal 36%, 132.1 mL-CH_4/g-COD_{added}	[73]

AnMBR configurations are available in the literature [31,32]. The side-stream configuration uses a recirculation pump to ensure trans-membrane pressure. The pressure-driven cross-flow is able to generate the surface shear, which disrupts the filtration cake for membrane fouling control [33]. Fouled membrane can be in-situ cleaned without interrupting system operation. The main drawbacks of side-stream configuration are more energy consumption as well as the rupture of sludge agglomerates and the liberation of EPS/SMP caused by high shear forces, which accelerates the membrane fouling and clogging [32]. In comparison, internal/submerged scheme, without use of a recirculation pump, is less energy-consuming. Due to the absence of the recirculation pump, surface shear forces are much lower, and thus the produced biogas is typically recirculated to remove filtration cake [24]. Biogas recirculation, however, can result in operation problems, especially the increase in dissolved methane in the effluent. In addition, chemical cleaning of this scheme is difficult since membrane module has to be taken out from reactor before cleaning. For external submerged AnMBR, the membrane is placed in an external membrane tank. Membrane fouling is mitigated mainly by concentrating the high-shear in the small external tank [34].

Furthermore, AnMBR configuration has a considerable effect on the treatment performance. It is reported that submerged anaerobic dynamic membrane bioreactor (SAnDMBR) required shorter time to form an effective dynamic membrane (DM) layer able to remove additional organic matters during filtration, ensuring a higher methane production rate and better permeate quality, compared with external anaerobic dynamic membrane bioreactor (EAnDMBR) [35]. Surely, a thicker DM layer in turn leads to higher filtration resistance and larger TMP values [36]. Recently, several novel configurations have been newly developed to maximize methane production and solids removal, such as anaerobic fluidized-bed membrane bioreactor (AnFMBR) [37], anaerobic electrochemical membrane bioreactor (AnEMBR) [38], anaerobic osmotic membrane bioreactor (AnOMBR) [39], anaerobic membrane distillation bioreactor (AnMDBR) [40], and granular anaerobic membrane bioreactor (GAnMBR) [41]. For more detailed information, readers may refer to Maaz et al. [24].

3. Main factors affecting the treatment performance of AnMBR

3.1. Type of waste streams treated

Up to date, AnMBR has been used for treating a variety of waste streams, such as wastewater, sewage sludge, food waste, coffee waste, manure and landfill leachate, as summarized in Table 1. For all kinds of wastewaters, membrane with pore size 100 KDa can retain almost all SS and microorganisms [42]. Domestic wastewater can be easily degraded and treated for methane production [43]; moreover, methane productivity increases with increasing organic loading rate (OLR) [16]. Comparatively, the application of AnMBR for treating highly concentrated industrial wastewater usually faces many technical problems, e.g. the toxicity to microorganisms [44]. Taking dyeing wastewater as an example, although AnMBR can give high organic removal (90–94%) and nearly complete decolorization (> 99%) [45,46], methane conversion is not always stable due to toxic inhibition on methanogens, accumulation of VFAs and pH drop caused by high-concentration azo dyes [47]. Similar observations were also reported in treating metalworking fluids [48], pharmaceutical wastewater [49], and high-strength landfill leachate (LFL) [50]. It should be emphasized that AnMBR technology allows enough retention time for anaerobic microbes to acclimatize to the toxic environments [51]. Therefore, in theory a wastewater can be anaerobically treated by AnMBR as long as it is amenable to anaerobic treatment [44]. In order to further promote the removals of contaminates, AnMBR technology usually needs to be combined with other processes, such as reverse osmosis [52] and anaerobic digester [53].

As a typical biomass waste, sewage sludge has the characteristics of high-solids content, numerous pathogens, odor, large energy potential, etc. [54,55]. Because of the special features, AnMBR system regards sewage sludge as valuable "energy source", simultaneously realizing bioenergy recovery and waste stabilization [56]. Decoupling HRT and SRT allows the system to operate with a higher OLR and longer SRT [30]. High SRT enhances the acclimatization of biomass, the hydrolysis of slowly degradable compounds, and the breakdown of complex particles, thereby inducing higher biodegradability, higher biogas production, and more stabilized and concentrated digested sludge [57]. Meanwhile, membrane filtration also shows the great promise for simultaneous sludge stabilization, thickening and dewatering [58,59]. Considering the presence of large particles (grit, branches, etc.), sewage sludge, in particular primary sludge, often needs pre-screening before being pumped to the digesters [57]. Because of the complex and rigid floc structure, methanization efficiency of sewage sludge still remains unsatisfactory. As a result, various pretreatment strategies (e.g. hyperthermophilic hydrolysis) have been integrated with AnMBR to improve solids removal and energy harvest [60]. Although the studies associated with the combination of pretreatment and AnMBR are still very few in the literature, the positive effect induced by pretreatment is obvious.

Similarly, AnMBR exhibits the superior and reliable performances in treating high-organic food waste (FW) than the continuous-flow continuous stirred tank reactor (CSTR) and self-agitated reactor (SAR) in terms of optimal OLRs, COD conversion and effluent quality [61–63]. It is worth noting that due to the high-solids content and poor fluidness, FW has to be highly diluted with tap water prior to feeding into AnMBR reactor to sustain the process stability. Another issue affecting the AnMBR treatment of FW slurry is the accumulation of long chain fatty acids (LCFAs), due to the presence of high-concentration lipids. LCFAs creates a physical barrier and retards the mass transfer by adsorbing onto anaerobic sludge, thereby inhibiting microbial activity and upsetting process stability [64]. Though the physio-chemical process via precipitation with divalent cations (e.g. Ca^{2+}) in FW can attenuate free LCFAs toxicity to a certain degree, the formation of low bioavailable LCFAs-Ca^{2+} precipitates tend to cause reactor clogging and membrane inorganic fouling, and impact digestion efficiency [65]. Due to the high-solids content, FW has also been anaerobically co-treated with other organic wastes, especially low-strength wastewater to maximize bioenergy recovery. It is reported that treating FW jointly with urban wastewater could accelerate the growth of *Archaea* population and fermentative genera (e.g. *Anaerolineaceae* and *Synergistaceae*) [66], and facilitate the conversion of complex components, methane production and sludge reduction. Obviously, a joint treatment with wastewater is able to offer a more robust, efficient, and eco-friendly process, reducing digestate production, energy consumption and subsequent treatment cost [67]. The mixing ratio of FW and wastewater is determined according to the Penetration Factor (PF) established, namely, the percentage of local households using FW disposers [68].

Besides, AnMBR system has also been employed for the treatment of other high-solids biowastes such as coffee grounds and livestock manure for biogas production [69–71]. Mono-digestion of coffee grounds shows poor performance due to the lack of nitrogen and trace metals [2], as well as the propionate build-up induced by retarded β-oxidation caused by the accumulation of hydrogen [72]. In contrast, livestock manure is more easily bio-converted for green energy conversion [73]. Additionally, the researchers have also investigated the feasibility of ammonia and phosphorus recovery as struvite from the permeate by crystallization process [21]. It is obvious that AnMBR can be a potential option for treating high-solids wastes despite still limited knowledge about the kinetic behaviors (e.g. microbial community, key metabolic intermediates, fermented residuals, etc.) occurring within the system.

In order to further explore effect of the types of treated waste streams relationship on the performance of AnMBR, box chart statistical analysis was performed (Fig. 3). As shown in Fig. 3a, methane yield shows less dependency on the type of substrates, and different waste streams has highly similar methane conversion efficiency. In sharp contrast, the optimal opertional conditions vary greatly. As expected, the longest SRT (i.e. hundreds of days) was observed for the treatment of wastewater and the SRT used for high-solids wastes is usually shorter than 50 days to sustain a high treatment capacity (Fig. 3b). The popular HRT for wastewater is hours to serveral days while it is 10–20 days for easily biodgeradable biowastes (such as sludge, food waste and manure) and reachs nearly one month for coffee grounds (Fig. 3b). For reference, the application range of AnMBR for treating various waste streams was summarized as well and is depicted in Fig. 4.

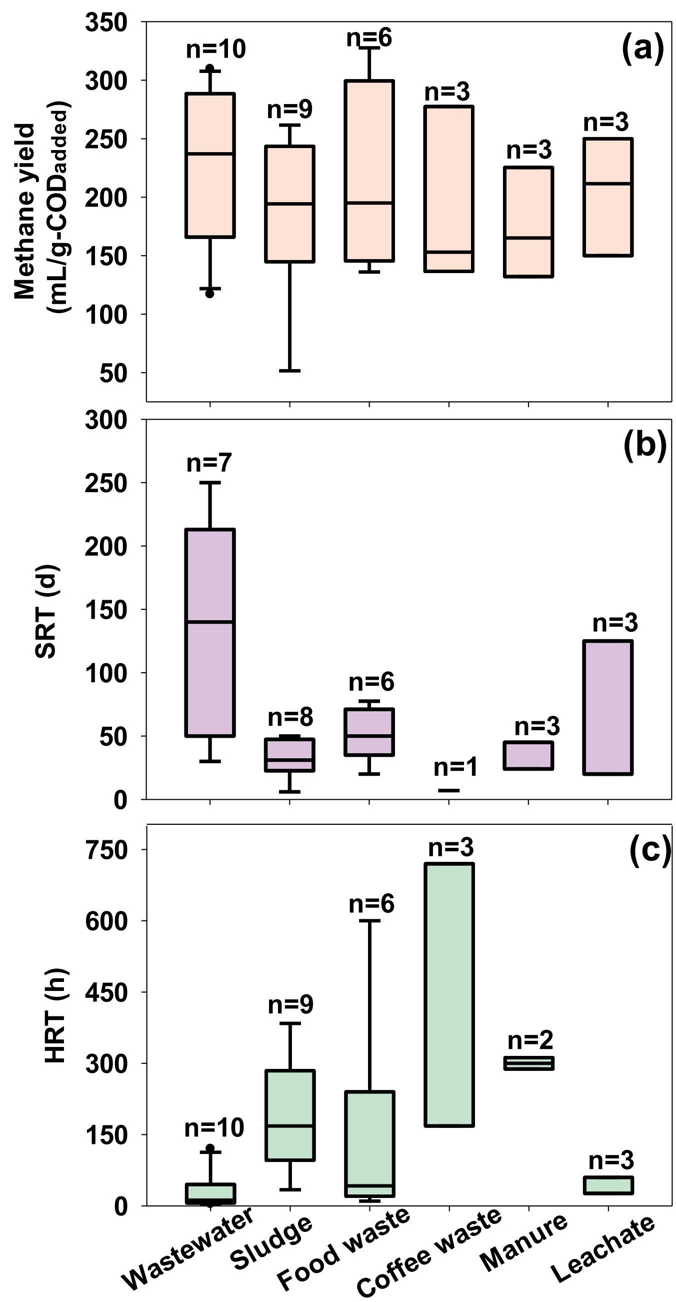

Fig. 3. Comparison of methane yield (a), SRT (b), and HRT (c) for treating various waste streams. Description of box plots: top and bottom of box = 75th and 25th percentiles; top and bottom whisker end = maximum and minimum; solid line in box = median value; gray dote = outlier. Note: wastewater [74–76,80–86]; sludge [30,57,59,78,87]; food waste [62,63,67,79,88]; coffee waste [2,71,72]; manure [21,89]; leachate [90,91].

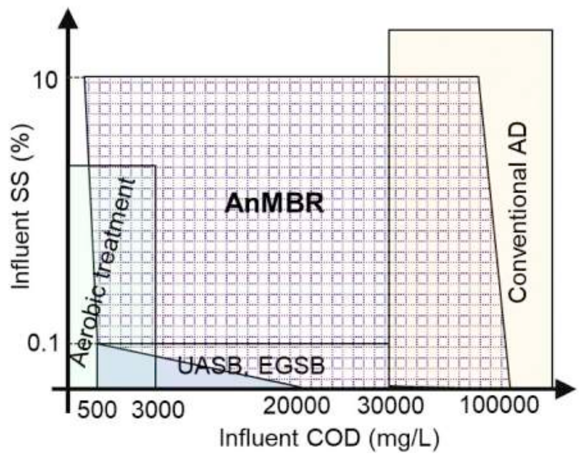

Fig. 4. Application range of AnMBR for treating waste streams (UASB: Up-flow Anaerobic Sludge Blanket; EGSB: Expanded Granular Sludge Blanket Reactor; AD: Anaerobic Digestion) [92,93].

3.2. Solids retention time (SRT)

SRT is one of the most important parameters affecting the performances of AnMBR. It is accepted that a long SRT is beneficial to get better COD removal and methane production for a specific biowaste (Fig. 4a). The difference between COD removal and CH_4 yield can be explained by the non-degraded particulate COD that is retained in the reactor. In some cases, as summarized in Table 1, most of the removed COD has been retained as particulate matter inside the AnMBR reactor rather than being degraded. It confirms that long SRT and high biomass concentration are the prerequisites of the full degradation of substrate. Huang et al. [8] studied the effect of different SRTs (30, 60 and infinite days) on treating domestic wastewater and observed the maximum specific methane productivity of 0.056 L-CH_4/g-MLVSS/d at the infinite SRT. Of course, it does not mean that the higher the SRT the better the overall performance. Too long or too short SRT are both unfavorable to the process stability [94]. Too short SRT, in addition to less methane output, leads to the substantial accumulation of SMP and the severe membrane fouling. Similarly, too long SRT accelerates membrane fouling and causes poor stability as well. Longer SRT can maintain higher MLSS concentration to upgrade organic biodegradation efficiency, but too high MLSS accelerates the formation of membrane fouling. In addition, too high SRT might also potentially lead to drop in permeate flux accordingly restricting bioreactor treatment capacity, despite that increasing SRT at a constant HRT increases the suspended mass concentration [95]. Therefore, achieving an optimal SRT is a prerequisite for a stable, and highly efficient AnMBR process. To sustain an appropriate SRT, a quantity of digestate biomass should be withdrawn periodically by peristaltic pump or manually. However, due to the differences in the properties of treated waste streams and operational conditions, the real SRT value or discharge frequency of biomass differs greatly (Fig. 3b).

3.3. Hydraulic retention time (HRT)

HRT is also a pivotal operational parameter of the AnMBR system (Fig. 5a). A suitable HRT cannot only gain a satisfactory treatment performance [75] but lessen membrane fouling. Ho and Sung [85] investigated the performance of AnMBR at different HRTs for treating synthetic municipal wastewater. It showed that the variable HRT from 12 to 6 h had no marked influence on COD removal (> 90%), and the observed methane yield was very close, falling within a narrow range of 0.22–0.24 L/g-COD_{added}. In parallel, Kunacheva et al. [80] observed the stable COD removal (> 94%) in a wider HRT range of 12–2 h. Surely, further decreasing HRT to 1 h, to a certain degree, led to the VFAs accumulation and the drop of COD removal, thereby deteriorating the performance. Slightly differing, Huang et al. [8] discovered that the decline of HRT (12, 10 and 8 h) facilitated the transformation of organic matter and methane production regardless of the operating SRT (30, 60 and infinite days).

Ulteriorly, a detailed statistical analysis on several representative AnMBR studies was made to establish the possible relationships among SRT, HRT, SRT/HRT and treatment performance. Although the data used for this purpose is not so sufficient, some interesting outcomes can be still discovered. The dependence of AnMBR performance on SRT rose at low HRT range, within which TCOD removal increased with increasing SRT while keeping the HRT constant (i.e. "SRT-dependent area") (Fig. 5a). Conversely, at higher HRT range, AnMBR performance became SRT-independent at SRT > 30 d; and further increasing SRT cannot induce additional promotion in TCOD removal (i.e. "SRT_{30}-independent area"). Therefore, a tradeoff could exist in controlling SRT

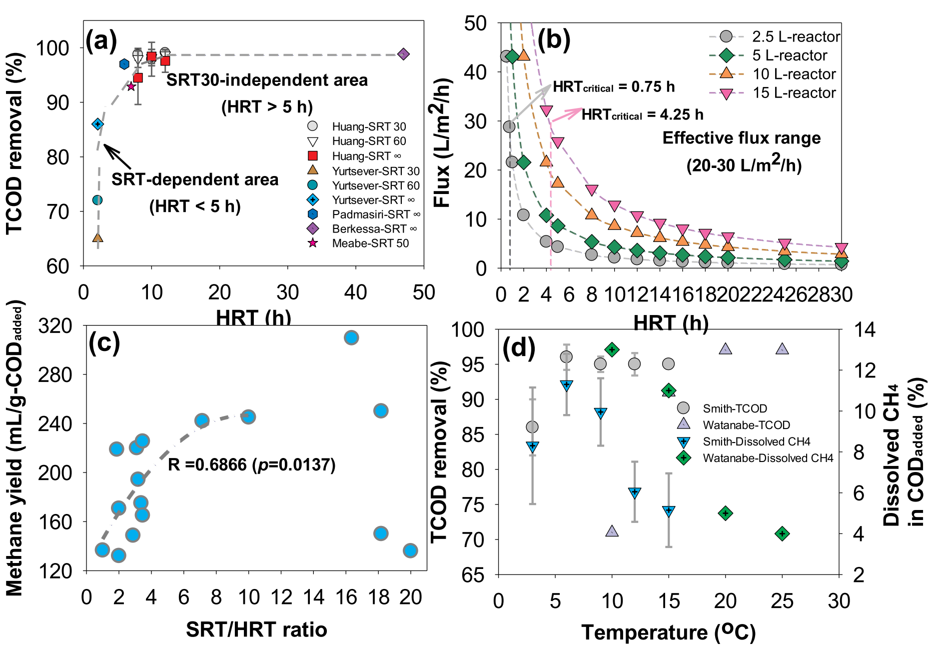

Fig. 5. (a) TCOD removal as a function of HRT observed in Huang et al. [8] (25–30 °C), Yurtsever et al. [102] (15 °C), Padmasiri et al. [103] (15 °C), Berkessa et al. [104] (37 ± 1 °C) and Meabe et al. [30] (55 °C); (b) relation between critical HRT and flux (KUBOTA flat-sheet membrane with filtration area of 0.116 m^2 and effective flux of 20–30 L/m^2/h was used for the calculation); (c) TCOD removal and dissolved methane percentage in the effluent as a function of operational temperature observed in Smith et al. [101] (HRT = 17, 19, 26, 29 h) and Watanabe et al. [74] (HRT = 12, 16 h).

and HRT to achieve high treatment capacity and maximal bioenergy generation. Meanwhile, Besides, for a specific membrane module, the selection of HRT strongly depends on the effective flux of used membrane, and reactor volume (i.e. Flux $(L/m^2/h) = V_{reactor}/(A_{membrane} \times HRT)$). Taking KUBOTA flat-sheet membrane with effective flux of 20–30 $L/m^2/h$ as an example, the relation between critical HRT and flux was established (Fig. 5b). In theory, the critical HRT can be shortened to as low as 0.75 h for a 2.5-L reactor equipped with one membrane; whereas it will be prolonged to 4.25 h if reactor is enlarged to 15 L. For the enlarged reactor, shortening HRT can be accomplished by applying more membrane modules. In reality, the applied HRT usually varies from days to hours according to the characteristics of treated biowastes. For instance, HRT was reported to be 5–20 d for hydrolyzed sludge [60], 13 d for swine manure [21], 18.5–22.7 h for food waste [96], and 1–4 h for wastewater [80]. The relation of HRT and flux becomes more complicated when some of the permeate is recycled [97]. Fig. 5c illustrates the possible correlation between SRT/HRT and methane yield. It is evident that SRT/HRT ratio has a significant influence on methane production. Methane conversion increases considerably with SRT/HRT ratio in low SRT/HRT range (< 10) whereas it sharply declines once SRT/HRT is higher than 16. The preliminary investigations reflect that SRT/HRT ratio should be considered as a key parameter during the operation of an AnMBR system, despite its unique merit of decoupling HRT and SRT.

3.4. Dissolved methane in low operational temperature

Anaerobic microorganisms especially the methanogenic bacteria are very sensitive to the environmental circumstances within the bioreactor, and their metabolic activity or proliferation is closely related to operational temperature. Normally, anaerobic reactions are controlled at mesophilic (30–40 °C) or thermophilic (50–60 °C) conditions to ensure rapid growth/diversity of anaerobes [98] or increase destruction rate of organic solids and eliminate pathogens [30]. Also, numerous studies have been carried out to assess the long-term performance of AnMBR at low temperatures (Fig. 5d). However, methane solubility is higher at lower temperatures, leading to increased energy loss through dissolved methane in the effluent [99]. Smith et al. [100], treating domestic wastewater at psychrophilic temperature (15 °C), observed COD removal as high as 92 ± 5%, but low temperature dissolved 40–50% of the total produced methane and caused low methane recovery. The decreased temperature reduced suspended biomass activity and led to the high SCOD in the bioreactor. Although the soluble organics could be effectively removed via the biofilm colonizing the membrane, this gave rise to a significant increase in dissolved methane oversaturation in the effluent, thereby leading to the secondary bioenergy loss [101]. Similar observation was reported by Martin-Ryals et al. [78], who reported approximately 39% decrease in methane yield from 37 °C to 21 °C due to the decreased microbial activity and the increased methane dissolution. In a separate research, Watanabe et al. [74] operated a sewage-fed AnMBR for 650 days with the decrease of temperature from 25 °C to 10 °C, noticing the decreased sewage treatment efficiency and CH_4 production below 15 °C. They attributed this mainly to high methane dissolution and inhibited hydrolysis and acidification processes at lower temperature. Besides, higher membrane fouling due to the secretion of additional SMP and EPS resulting from the microbial self-protection behavior was considered as another serious cause of poor sewage treatment at low temperature.

Based on the previous researches, it is apparent that highly dissolved methane and severe membrane fouling are the big challenges for psychrophilic AnMBR, and the development of low-energy and straightforward technologies for dissolved methane recovery and membrane fouling control are necessarily needful. The methods for dissolved methane removal proposed in the literature are biological oxidation (methane oxidizing bacteria), aeration or gas stripping, but they are often less efficient and moreover cause methane loss/devaluation rather than recovery as an energy product. It is worth noting that too high temperature might conversely lead to the release of smaller size particles, which likewise aggravates membrane fouling through accelerating the clogging of membrane pores [30].

4. Membrane materials and fouling control

4.1. Membrane materials

Membrane module is one of the core parts of AnMBR system. Membrane materials, pore sizes, surface charge, surface roughness, hydrophobicity, design/configurations and others all have significant effects on the membrane fouling rate and the performance of a reactor. The frequently reported membrane materials in the literature include polymers such as PVDF (polyvinylidene fluoride), PES (polyethylsulphone), PE (polyethylene), PP (polypropylene), PSF

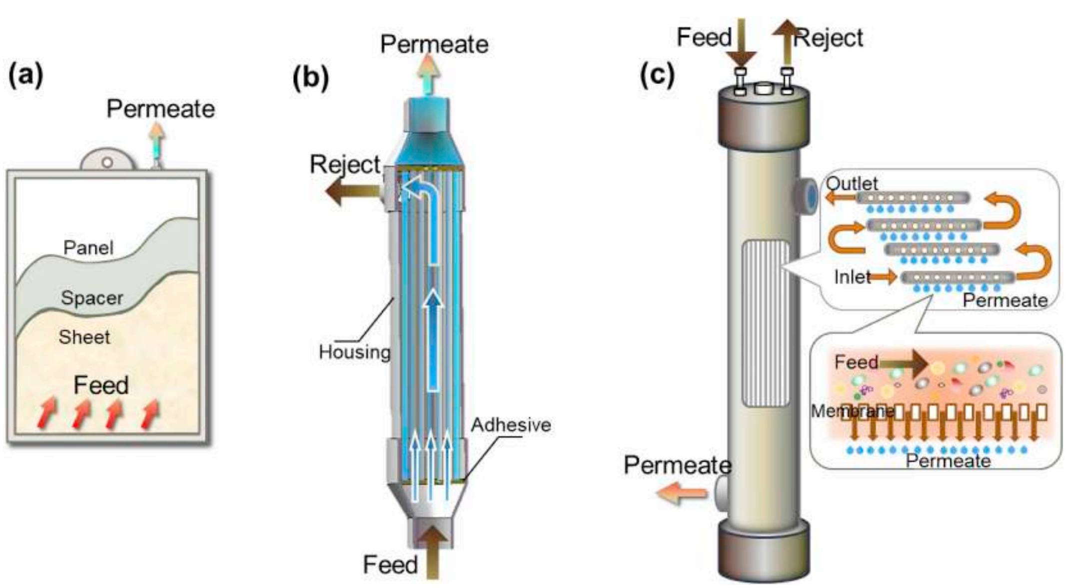

Fig. 6. Main types of membrane modules: (a) flat-sheet (Kubota Corporation, Japan), (b) external type hollow-fiber (Asahi Kasei Corporation, Japan), and (c) Tubular (Daicen Membrane-Systems Ltd., Japan).

(polysulfone), CPE (chlorinated polyethylene) and PTFE (polytetrafluoroethylene), and non-polymeric materials like ceramics [11,12,105]. Amongst them, PVDF membrane, as one kind of polymeric membranes, shows the highest popularity in lab-scale or industrial AnMBR construction [105] because of its excellent mechanical properties such as higher elongation at break and strong tolerance to frequent chemical cleaning due to the robust chemical resistance. It accounts for around 57% of the membrane products on the market, followed by PES [106]. This membrane material has a slower increase rate in irreversible resistance and a better performance in the irreversible fouling control than PE [107]. When modified with PFPE (perfluorinatedpolyether), PVDF membrane could better mitigate membrane fouling and achieve higher stripping flux, higher membrane mass transfer coefficient and more satisfactory methane recovery ability [108]. For more details of the production and modification of PVDF membranes, the readers are referred to Liu et al. [105]. Meanwhile, more attention has also been payed to the use of inorganic membranes, such as stainless steel membrane [109]. This membrane can get an excellent COD removal and higher permeate flux while simultaneously improving the nitration process. The main challenge for inorganic membranes is struvite precipitation [110], which tends to result in severe membrane fouling. Slightly differing, inorganic ceramic membrane possesses higher biomass concentration, longer service life, superior filtration performance and lower membrane fouling potential even compared with polymeric membranes [111,112]. The use of ceramic membrane also has some inherent limitations, in particular high manufacturing cost, restricting its real applications to a certain degree [113]. Nonetheless, with the improvement of manufacturing technology and the decrease of ceramic membrane price [114,115], the application of ceramic membrane will become ever-growing popular in the near future.

4.2. Membrane configurations

The common membranes applied for AnMBRs are MF (microfiltration) and UF (ultrafiltration) membranes, with three kinds of classical configurations: (a) flat-sheet, (b) hollow-fiber and (c) tubular (Fig. 6). Flat-sheet membrane has the merits of good stability, and the ease of cleaning and replacement, presenting a substantial promise for commercial applications. The most representative flat-sheet membrane can be KUBOTA Submerged Membrane Unit® developed by KUBOTA Corporation, Japan. The unit is a proven and reliable process that has been used in real AnMBR installation for the treatment of various types of biowastes since 2000 [116]. The main obstacle for KUBOTA flat-sheet membrane is relatively high cost (around 2500–3500 CNY/m^2, with effective flux of 20–30 L/m^2/h). Another commercially available flat-sheet membrane is supplied by Shanghai SINAP Membrane Science & Technology Co. Ltd., China. It has a comparatively lower cost of 350 CNY/m^2, with effective flux of 16–22 L/m^2/h and effective lifespan of 10 years [117] (for more detail see http://www.sh-sinap.com/Showpro.asp?id=3). Hollow-fiber membrane also shows a high attractiveness in research community for treating a broad range of biowastes due to their high membrane area per unit volume and cost-effectiveness [62,88,118]. The deficiencies of hollow-fiber membrane are the fast fouling rate and the requirement for high washing frequencies [119]. On the other hand, hollow-fiber membrane is cheaper in production and can withstand heavy backwashing [120]. Membrane price is mainly in the range of 150–200 CNY/m^2, with effective flux of 14–20 L/m^2/h and effective lifespan of 5–10 years [121]. Moreover, because of higher gas desorption rate, microporous hollow-fiber membranes have been frequently applied for degassing applications and dissolved methane recovery from AnMBR effluent [108,122–124]. Similar to flat-sheet membrane, tubular membrane also offers the advantages like high mechanical strength, high resistance to harsh environments, low fouling, long life, ease of cleaning and replacement as well as high convenience of handling SS and viscous fluids; however, the relatively high capital/operational cost, low packing density increase the difficulty for its commercial applications [12]. Compared with the former two, the application of tubular membrane appears to be more limited at present.

Regarding filtration, because of the discrepancy in the pore size or MWCO (molecular weight cut-off) of MF and UF membranes, they generally exhibit different membrane flux (filtration) and fouling behavior. The membrane pore size plays a pivotal role in the interception/retention efficiency of SS and microorganisms [125]. The smaller the membrane pore size, the more beneficial to the retention. Pore sizes of MF membranes usually are 0.05–0.45 μm and 0.002–0.05 μm for UF membranes [11,44]. Hence, all colloidal particulates and microbial cells in the influent can be retained completely by both units. Surely, the larger the membrane pore size, the higher flux and lower energy input. Another potential and effective option is forward osmotic (FO) process driven by the osmotic pressure difference, which retains solutes but allows water permeation through a semi-permeable membrane; other benefits of this process include high rejection of a wide range of contaminants, lower membrane fouling propensity [126,127]. Also, FO-AnMBR was demonstrated to have higher potentials in removing organic carbon, total phosphorus and ammonia-nitrogen than conventional pressure-driven AnMBR [128]. The limiting factors for FO membrane application are difficult cleaning [129] and high salinity situation.

4.3. Development and mechanisms of membrane fouling

Membrane fouling represents a major obstacle for the practical applications of AnMBRs [130]. In general, membrane fouling is classified under two categories: reversible fouling and irreversible fouling. Reversible fouling refers to the foulants that are easily removed by physical cleaning; and irreversible fouling is mainly associated with pore clogging/blocking and can only be eliminated through chemical cleaning. The commonly cited fouling mechanisms include pore blocking, adsorption or precipitation of organic/inorganic matters, concentration polarization, and cake layer formation [131]. The influent entering AnMBR usually contains a variety of substances such as particulate matters, colloids, soluble organic matters (EPS, SMP), inorganic matters, and microbial cells. Pore blocking occurs initially prior to cake layer formation, mainly caused by the accumulation of foluants with a size dimension smaller than or comparable to the pore size (colloids, solutes, microbial cells, etc.) [44,132], and is referred to as irreversible fouling. Cake layer formation results from the attachment of EPS/biomass, the deposition of small flocs and metal/struvite precipitation onto the membrane surface, usually belonging to reversible fouling [133]. The reversible fouling can transform into irreversible cake layer with cake consolidation in a long-term filtration process [11]. To date, different expressions have been developed to describe the membrane fouling resistance [113]. A classical, simple filtration test (Eq. (1)), proposed by Lee et al. [134], and further modified by Chae et al. [135] and Nie et al. [136], shows a substantial potential for this purpose since it can determine each resistance induced by the cake layer, pore blocking or the membrane itself.

$$J = \frac{1}{A}\frac{dV}{dt} = \frac{\Delta p}{\mu R_t} = \frac{\Delta P}{\mu (R_m + R_c + R_{P-org} + R_{P-inorg})} \tag{1}$$

where J is the permeate flux ($m^3/m^2/s$), V the volume of permeate (m^3), A the membrane area (m^2), $\triangle P$ the transmembrane pressure (TMP, Pa), μ the viscosity of water (0.8949×10^{-3} N s/m^2 at 25 °C), R_t the total membrane resistance (m^{-1}), R_m the intrinsic membrane resistance (m^{-1}), R_c the cake layer resistance (m^{-1}), $R_{p\text{-}org}$ the organic pore blocking resistance (m^{-1}), and $R_{p\text{-}inorg}$ the inorganic pore blocking resistance (m^{-1}) [132].

Fig. 7 illustrates the fouled membrane cleaning protocol extensively used for determining different membrane resistance distribution: (i)

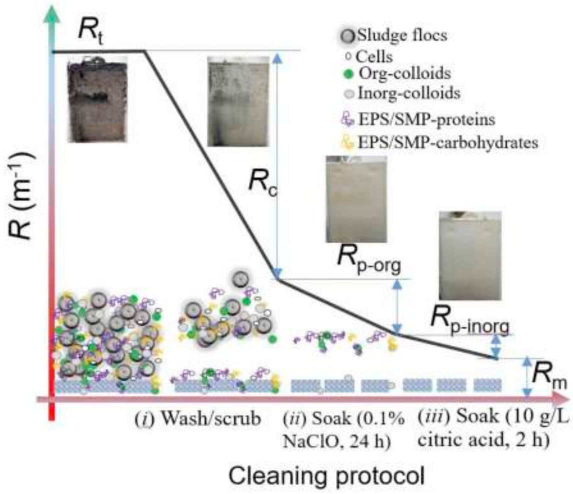

Fig. 7. Fouled membrane cleaning protocol used for determining membrane resistance distribution.

wash away the cake sludge on the membrane surface by a sponge, (ii) soak the membrane in 0.1% NaClO alkaline solution for 24 h to clear the organic matter in the membrane pores, and (iii) soak the membrane in 10 g/L citric acid solution to clear inorganic ions in the pores [132,136]. Through filtration test, it has been demonstrated that the faster formation of cake layer (R_c) is the main contributor to membrane fouling [28,137]. The continuous extension and thickening of cake layer leads to the decrease of membrane flux and the increase of TMP, eventually deteriorating the performance of reactor. Noted that the formation of cake layer is not necessarily harmful to AnMBR system as the initial layer might be beneficial to the purification of effluent through adsorbing or biodegrading the low-molecular substances or submicron colloidal particles [35,90,137,138].

The parameters affecting cake formation and membrane fouling include influent characteristics, SRT [102], temperature shock [139,140], and SMP/EPS [132]. It has been widely accepted that SMP/EPS are the major constituents inducing cake formation and pore blogging (Fig. 8a) [113,141]. EPS, as the high molecular weight polymeric materials, are secreted during the metabolisms or self-lysis of microbial cells with proteins, carbohydrates, humic substances and lipids as the main components [142,143]. EPS are dispersed in the bulk liquid, or adsorbed/combined in the cell surface [11]. Many physiochemical protocols have been proposed into the extraction of EPS, such as heating, formaldehyde + NaOH, EDTA, cation exchange resin and sonication [144]. Based on the spatial distribution, EPS can be divided into soluble EPS (S-EPS, or so-called SMP [136]), tightly bound EPS (TB-EPS) and loosely bound EPS (LB-EPS) [145]. Understanding the variation and distributions of EPS and SMP is of great significance to the investigation of membrane fouling mechanisms and the exploration of fouling control methods. The composition and complexity of EPS and SMP often change with the type of substrate, the concentration of substrate, the operational parameters (e.g. HRT, SRT, OLR, food-to-microorganism (F/M), temperature, etc.), duration time, and even the extraction methods (Fig. 8b). The occurrence of EPS-induced membrane

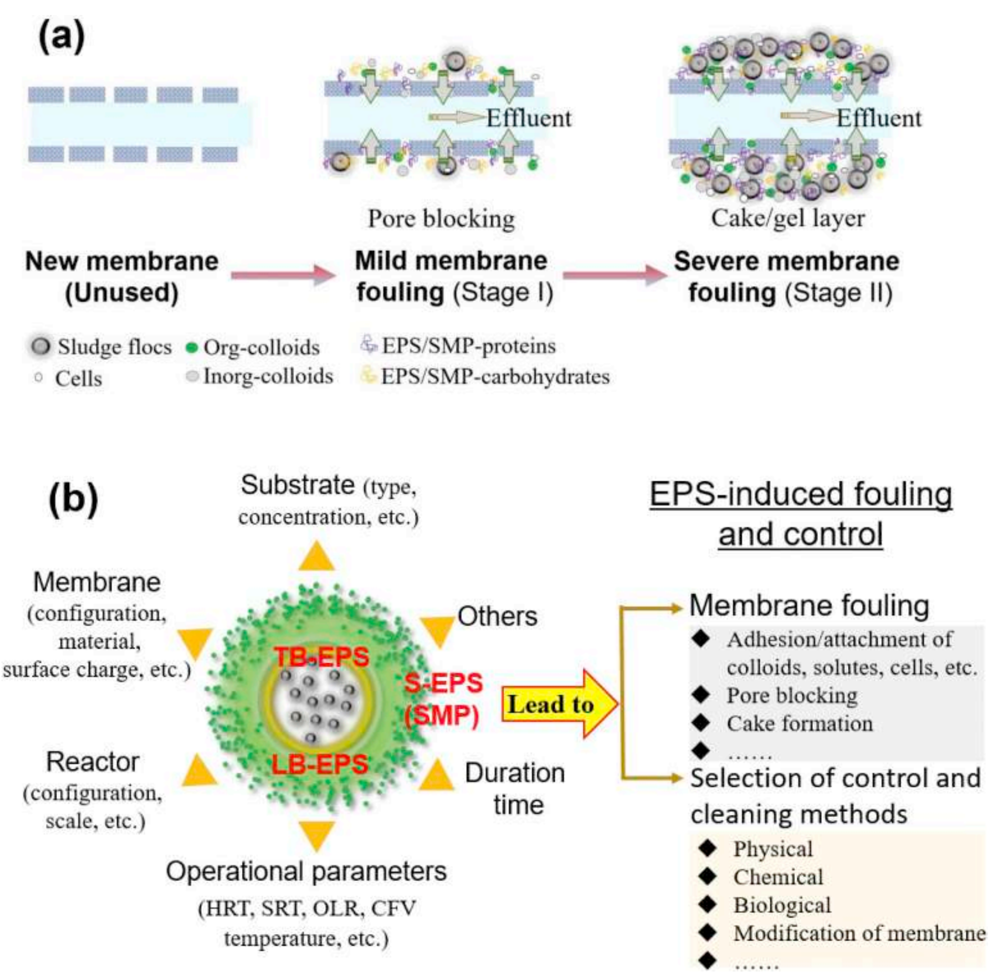

Fig. 8. (a) Adhesion/attachment of organic/inorganic foulants onto membrane surface/pores, cake formation and consolidation during long-term operation; (b) main factors governing secretion/production of EPS resulting in membrane fouling.

fouling is a highly complex, elusive bio-physicochemical process, Hence, how to unravel the fouling behaviors associated with SMP/EPS still remains a great challenge. Immense efforts have been devoted to characterize the chemical components and microstructure of SMP/EPS and to determine each role during foulant development [97,146]. It has been documented that the degree of membrane fouling and filtration resistance might be closely related to not only total EPS/SMP [136] but each EPS fraction, in particular LB-EPS [142,147].

In spite of the great efforts sunk into EPS, till date the findings with respect to the relative contribution of each component in different EPS fractions to membrane fouling is still inconsistent and even contradictory in some cases. Several line of studies noted that carbohydrate-based EPS played a critical role in membrane fouling [140,148]; in sharp contrast, some researchers asserted that the main foulant should be proteins fraction [8,149]. Nie et al. [136] proven that instead of carbohydrate, protein EPS was responsible to the increased pore blocking and reduced membrane permeability, in line with the observations of Lin et al. [28], Ramesh et al. [142], and Chen et al. [132]. Proteins determine hydrophobic characteristics and carbohydrates, hydrophilic [150]. The EPS with high hydrophobic proteins would have a strong adhesion forces, and thus tend to adhere more to membrane surface/pore. Buntner et al. [151] found that the existence of protein caused a drastic drop in filtration performance and they even highlighted the positive role of carbohydrate fraction in improving the filtration performance. These conflicting perspectives might be induced by the discrepancies in operating conditions and substrates. High concentration of EPS can induce severe membrane fouling, and low concentration of EPS also accelerate the membrane fouling at longer SRT due to the drop of the flocculation of particulates and particle sizes [8]. Just as, the relationship between EPS concentration and membrane fouling is difficult to explain [152]. Apparently, more works still should be performed in this aspect. Apart from the severe impacts on membrane fouling, EPS are also deemed as a critical barrier hindering sludge dewatering, hydrolysis, and subsequent valuable resource recovery [54,145,153].

4.4. Strategies for eliminating/minimizing membrane fouling

Fig. 9 depicts various fouling mitigation approaches and corresponding working principles in AnMBRs, and Table 2 summaries and compares the performances of different cleaning protocols used. Membrane cleaning methods include physical, chemical, biological, and combined protocols. One of the most commonly used physical methods is backwashing, which is reversing the flow with liquid or air to wash the membrane. Utilizing higher backflush fluxes is marginally better than longer backflush durations for fouling mitigation [154]. Other physical strategies such as relaxation [62,154], sub-critical flux operation [155], biogas sparging [88], and aeration [29] can also alleviate membrane fouling. Apart from those, many novel and effective physical cleaning methods have been developed. For example, online ultrasonication could utilize the vibration of sound waves to clean the membrane [156]. However, the decontamination effect of online

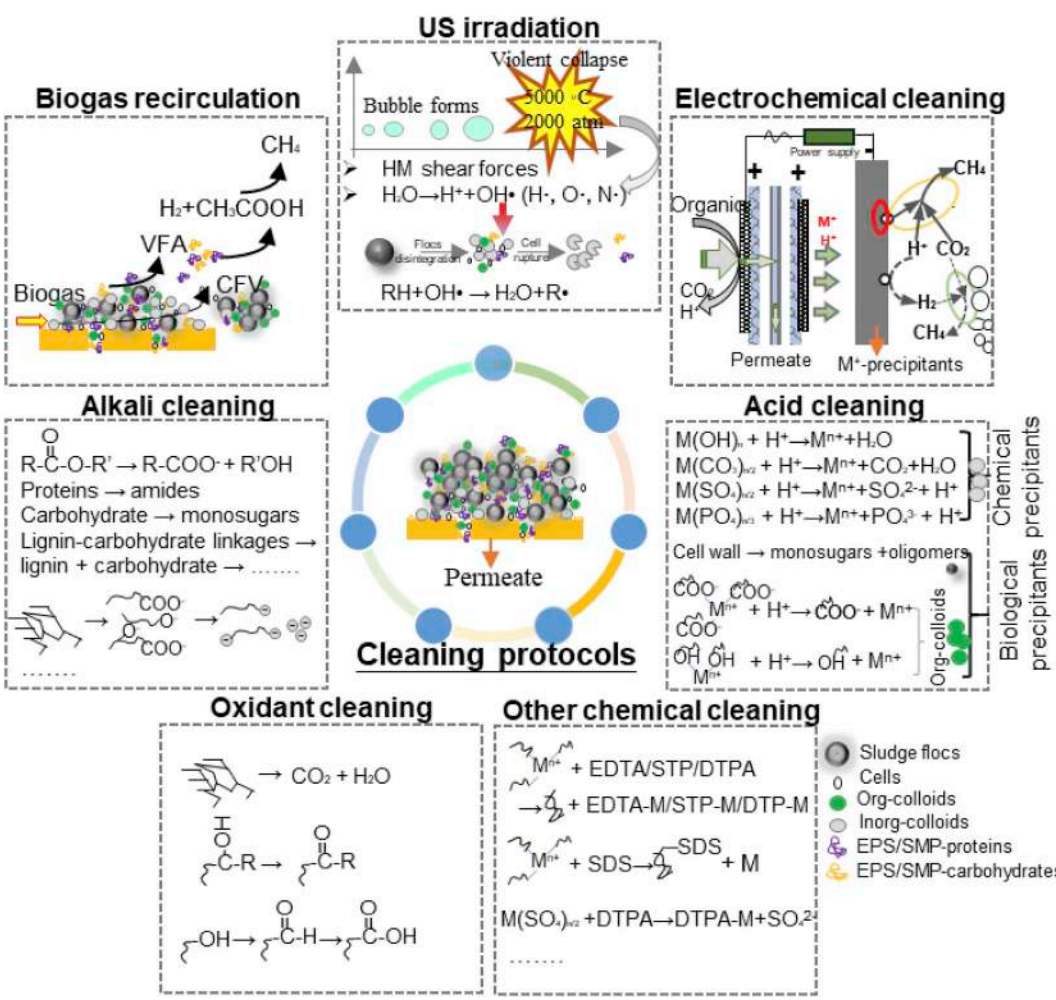

Fig. 9. Working principles of different protocols for membrane cleaning and fouling control (redrawn mainly based on Zhen et al. [54], Pilli et al. [196], and Wang et al. [177]). HM: Hydromechanical; EDTA: ethylene diamine tetraacetic acid; STP: sodium tripolyphosphate; DTPA: diethylenetrinitrilopentaacetic acid; SDS: sodium dodecyl sulfate.

Table 2
Membrane cleaning protocols proposed in different scale AnMBRs (unless otherwise stated).

Membrane	Supplier	Feed	Cleaning protocol	Characteristics	References
Physical					
Hollow-fiber PE (0.4 µm)	Mitsubishi Rayon	WAS	Irradiate with a US frequency of 28 kHz and an adjustable power output of 0–300 W	*In-situ*; mainly remove loose cake layer; Slightly affect the gel layer; no negative effect on the anaerobic microorganisms; greater sludge disintegration; $R_c = -80.4\%$	[156]
Flat-sheet PVDF (0.1 µm)	Mitsubishi Rayon	Diluted WAS	Apply intermittent electric field with a intensity of 50 V/cm and one cycle of 4 min on and 4 min off	*In-situ*; rejection of negatively charged foulants on membrane surface; complete permeability recover (100%)	[167]
Hollow-fiber PVDF (0.1 µm)	Zenon	Synthetic wastewater	Current density of 12.5 A/m^2, a period of 2, and an exposure mode of 415 s OFF–185 s ON	*In-situ*; higher sludge activity; increased floc size; decreasd SMP; lower fouling and enhanced anode electrodissolution	[168]
Hollow-fiber PVDF (0.04 µm)	GE Water & Process Technologies, USA	Domestic wastewater	Membrane rotation during the backwashing/relaxation phases	*In-situ*; enhance cake re-dispersion; reduce external fouling (56 − 18%)	[197]
Tubular PVDF (~0.018 µm)	Fareham, UK	Synthetic primary effluent wastewater	Fluidized GAC and CFV of 0.018–0.024 m/s	*In-situ*; lower cake layer formation rate; low recycle pumping rates; additional headloss; physical scouring for foulants, no GAC adsorption effect; 55–120% longer run-time; 70–100% energy saving	[162]
Chemical					
Flat-sheet CPE (0.2 µm)	Kubota	Synthetic wastewater	Wash away cake sludge and scrub by a sponge, soaked in 0.1% NaClO solution for 24 h and then 10 g/L citric acid solution for 2 h	*Ex-situ*; remove cake layer, organic matter and inorganic ions in membrane pores; high fouling flux recovery; $R_t = -90.4\%$	[132]
PVDF UF (MWCO 100 kDa)	Berghof, Germany	Kitchen waste slurry	Successive cleaning with tap water, 0.2% NaClO solution, pH 2 HCl solution and 1% EDTA	Low fouling flux recovery; periodical cleaning once the flux reduced by > 20%	[65]
Flat-sheet PE (0.4 µm)	Kubota	Synthetic leachate from OFMSW	Remove the external fouling by lukewarm water and a brush, soaked in 1% NaClO for 2 h and then in 1% oxalic acid for 2 h	*Ex-situ*; remove cake sludge on membrane surface, organic matter and inorganic ions in membrane pore; high fouling flux recovery (42.4%)	[97]
Tubular PES UF	Weir Envig, Paarl, South Africa	Swine manure	Sequential cleaning with 0.1 N HNO_3 (pH 2) and 0.5% EDTA-1% Na_3PO_4 (pH 10 adjusted with NaOH), 1 h each step	Remove gel layer and loosely attached foulants; remove Ca and P in membrane foulants; no effect on irreversible fouling; reduced resistance but incomplete flux recovery	[198]
Biological					
Hollow-fiber PVDF (0.1 µm, AeMBR)	Kolon, Korea	Simulated wastewater	Bacterial QQ using *Rhodococcus* sp. BH4 with CEB	*In-situ*; disrupt biofouling that cannot be achieved by chlorination; reduce the metal precipitates and EPS in biofilm; save filtration energy	[186,189]
Flat-sheet polyamide NF (AeMBR)	NE4040-90, WOONGJIN Corp., Korea	Synthetic wastewater	QQ acylase immobilization onto NF membrane surface by forming a chitosan − acylase matrix	*In-situ*; reduce secretion of EPS, prohibit the formation of mushroom-shaped biofilm; maintain a high flux after a 38-h operation	[184]
Combined					
Hollow-fiber UF	GE Water and Process Technologies	Meat-processing wastewater	Circulate biogas (0.9 L/min) and bulk liquid (1 L/min), maintenance cleaning once every week with citric acid (2000 mg/L, pH 2.5) and NaClO (200 mg/L, pH 8.75) at 30 LMH for 40 s then relaxing for 3 min in sequence of citric acid→tap water→NaClO→tap water	*In-situ*; weaken membrane fouling control effect over time; mitigate membrane fouling but do not recover flux	[88]
Hollow-fiber PTFE (0.2 µm)	Sumitomo Electric Industries Ltd.	FW slurry	Clean with biogas for 2 h, rinse with 1.5 L Milli-Q water and extendedly backwash at a flux of 60 mL/min with gas-aided for 2 h; backwash and desorb by 2000 mg/L NaClO with gas-aided for 24 h and then 3000 mg/L citric acid solution for 2 h	*Ex-situ*; removed cake layer, organic and inorganic foulants on gel layer or in pores; high fouling flux recovery (92.0%)	[62]
Hollow-fiber PVDF (0.1 µm, AeMBR)	Kolon, Korea	Synthetic wastewater	QQ sheet entrapping QQ bacteria (*Rhodococcus* sp. BH4, 200 mg); UV photolysis with a photon flux of 4 mW/cm^2 at 315–400 nm	*In-situ*; delay TMP build-up; greater QQ efficacy; no negative impact on the main biological performance	[188]

ultrasonic is limited, which can only get rid of the outer loose cake layer and has little effect on the tight cake layer. The effect of ultrasonic on microbial activity and membrane structure is also a problem [157].

Addition of granular materials is also an effective means [158], and it can reduce up to 25% of membrane cleaning and replacement costs [159]. Larger specific surface area of granular activated carbon (GAC) can effectively adsorb liquid colloidal particles and macromolecular organic compounds, reduce TMP, improve membrane flux and COD removal [160]. Also, the mechanical scouring due to the fluidization of GAC particles can further control membrane fouling [161], and the AnMBR with fluidized GAC as scouring agent is called anaerobic fluidized-bed membrane bioreactor (AnFMBR) [24]. The reactor with fluidized GAC offers many benefits, such as longer membrane run-time, energy saving, and low effluent BOD_5 [162]. In addition, fluidized GAC can provide a more selective environment for microbes (particularly methanogens), facilitating propionate-degradation, aceticlastic/DIET-dependent CO_2 reduction for methane production [163]. Dosing too much powdered activated carbon could cause a rise in viscosity that is harmful to the stability of system [91]. Other organic coagulants such as polyaluminum chloride, polyacrylamide [164], and ferrihydrite [165] were also chosen for membrane fouling control. The applications of coagulants can promote the aggregation of colloidal particles and tiny suspended solids in the solution, which can form larger particle size materials. Most recently, a new method by coupling electric field in MBRs has gained much attention. Coupling a small electric field is able to enhance effluent quality and mitigate membrane fouling via electrocoagulation, electrophoresis, and electrochemical oxidation [166]. High-intensity electric field can effectively strip membrane surface

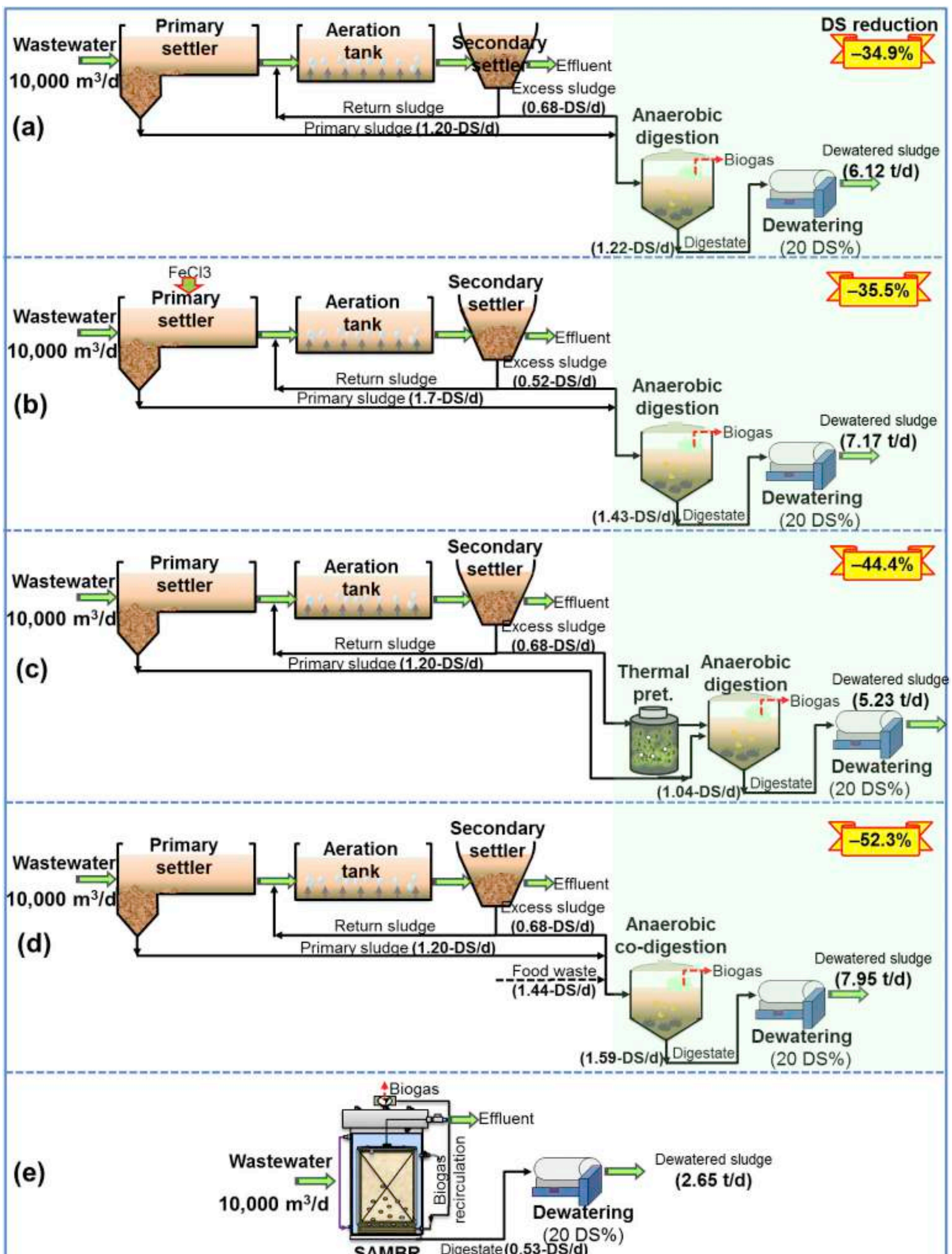

Fig. 10. Schematic diagrams of wastewater treatment processes compared: (A) the conventional activated sludge process coupled with sludge anaerobic digestion ("CAS + AD"), (B) the chemically enhanced primary settling-integrated CAS process coupled with sludge AD ("CEPS-CAS + AD"), (C) the CAS coupled with sludge thermal hydrolysis and AD ("CAS + TH-AD"), (D) the CAS coupled with sludge and food waste anaerobic co-digestion ("CAS + co-AD"), and (E) the AnMBR process.

pollutants to achieve the purpose of decontamination and antifouling [167]. Introduction of an external electric field cannot only be conducive to the prevention and control of membrane fouling, but also can strengthen the removal of COD, ammonia nitrogen, and phosphorus [168]. By operating a combined electrochemical-AnMBR system, Hou et al. [39] obtained the efficient removal of organics (> 93%) and phosphorus (> 99%), and the recovery of valuable resources (65% of PO_4^{3-} and 45% of NH_4^+). More details associated with membrane fouling mitigation by applying electric field can be found in Ho et al. [169] and Li et al. [170].

Physical cleaning can mainly remove the removable fouling, and chemical cleaning is an effective option for the removal of irremovable fouling. A variety of chemical reagents, such as acid solutions (HCl, H_2SO_4, H_3NO_4, citric acid, etc.), alkalis (NaOH), metal chelating agents (EDTA) and oxidants (NaClO, H_2O_2) have been used for chemical cleaning [12]. Other oxidants such as ferrate (VI) [171], ozone (O_3) [172] and electro-oxidation (•OH) [173] were also reported for membrane fouling mitigation. Another classical oxidization process is Fe(II)-activated persulfate (Fe(II)–$S_2O_8^{2-}$) reaction, which can generate powerful sulfate free radicals (SO_4^{-}•) with the redox potential (+2.60 V) similar to •OH. As a strong oxidant, SO_4^{-}• can easily disintegrate EPS and bacterial cells and liberate/decompose intracellular substances. Because of the powerful oxidizing ability, Fe(II)–$S_2O_8^{2-}$, since launched by Zhen et al. [174] in 2012, has been widely adopted for enhancing sludge dewaterability [54,153,175]. Although there is no any study reported in literature relation to its use in membrane fouling to date, the good performance of Fe(II)–$S_2O_8^{2-}$ for membrane cleaning still can be anticipated. Due to the ease of operation, membrane cleaning *in-situ* is more popular than cleaning *ex-situ*. The most common practice is a maintenance cleaning once a week and an intensive cleaning in 6–12 months [176]. There is not completely positive correlation between duration time of cleaning and cleaning efficiency. Prolonged or frequent use of chemical agents could cause the damage/degradation to the membrane [177]. Meanwhile, high concentration also is unnecessary, and 0.01–1% NaClO, dependent upon experimental conditions and membrane materials, is sufficient to achieve a high permeability recovery. Too high dosage during *in-situ* cleaning is more prone to damage cell integrity [178] and inhibit methanogenic activity [179]. In some cases, the cleaning efficiency achieved by a single chemical reagent is limited, and the combination of several chemical strategies is usually performed to achieve a higher cleaning efficiency [180]. Besides, more attentions have been also paid to the combination of different physical and chemical cleaning methods, such as low-dosage UV/chlorine pre-oxidation [181] and a integrated strategy of intermittent permeation, biogas and bulk liquid recirculation and maintenance cleaning [88]. Other combined processes, such as chemically enhanced backflush (CEB) [154] and ultrasound and alkaline solution of EDTA [182] were also found to greatly improve the effectiveness of membrane cleaning.

Biological cleaning achieves the control of membrane fouling mainly through the three methods, i.e. enzymatic approach, energy uncoupling, and bacterial quorum quenching (QQ) [177,183]. As an emerging anti-biofouling strategy, the enzymatic QQ retards biofouling development by degrading the quorum sensing signal molecules such as N-acetyl homoserine lactone (AHL) autoinducer using a QQ enzyme (acylase) [184]. In this context, the implementation of QQ can suppress the EPS secretion, mitigate the rate of TMP build-up, and reduce energy consumption by reducing coarse bubble aeration without compromising biological treatment performances [185,186]. Combining QQ with GAC, UV photolysis or chlorine injection tends to have synergistic effects [187,188] while saving substantial filtration energy [189]. Meanwhile, the QQ approach offers other benefits, such as lower aeration demand, less membrane cleaning frequency while saving energy, and reduced labor and chemical costs [190]. In spite of the great promise in improving biofouling mitigation, there are still several technical problems limiting the practical applications of QQ strategies, in particular the lack of robust and durable QQ media [190]. It is worth noting that up to now QQ strategies have been mainly used in AeMBR systems, the information available on their applications for AnMBR biofouling control is still very limited. Other strategies, such as chemical modification of membrane [191,192], development of multifunctional membrane [193,194], and design of new AnMBR configuration [41,195] were also proposed to improve the anti-fouling properties.

5. Biological processes for simultaneous wastewater treatment, sludge reduction and chemical energy harvest-which is the best?

In order to better understand its strength and superiority, five different wastewater treatment processes were designed and the advantages and limitations of each were evaluated in terms of sludge production/reduction, energy input and recovery, economic benefits and environmental impacts for comparison.

Fig. 10 illustrates the five different biological processes. Treatment Process A represents the conventional activated sludge process (CAS), followed by an anaerobic digester (AD) for mixed sludge treatment (i.e. primary sludge (PS) and excess sludge (ES)) ("CAS + AD", Fig. 10a). Treatment Process B includes chemically enhanced primary settling (CEPS) in primary clarifier with $FeCl_3{\cdot}6H_2O$ as the flocculant (~10 mg/L, 260–303 US $/t) ("CEPS-CAS + AD", Fig. 10b) [199] to remove biodegradable organics in the influent [200]. Treatment Process C employs thermal hydrolysis (TH, CambiTHP™ process) to pretreat ES prior to entering anaerobic digester to increase its biodegradability ("CAS + TH-AD", Fig. 10c). Treatment Process D co-digests sewage sludge with food waste (FW) based on dry VS ratio of 1.0: 1.0 instead of TH pretreatment ("CAS + co-AD", Fig. 10d). Treatment Process E represents the AnMBR process, a newly emerging technology being explored universally ("AnMBR", Fig. 10e). Typical wastewater compositions, according to the actual wastewater surveyed in Japan, were assumed and are listed in Table 3. A treatment capacity of 10,000 m^3/d (Q^3) was taken while assuming the effluent qualities reaches BOD_5 10 mg/L, and SS 10 mg/L, approved in Japan [201]. Note that N removal was not considered during the comparison considering the poor NH_4^+-N removal ability of AnMBR [202]. Sewage sludge (and FW) was mesophilically digested, dewatered mechanically and then transported a long distance of 50 km for land application [54]. Of particular importance for each process studied are energy input (i.e. aeration, thermal hydrolysis pretreatment, AD stirring/heating, as well as digested biosolids mechanical dewatering, transport and land use), biomethane production, digested residual reduction and chemical conditioner dosage added for the subsequent deep-dewatering. The general parametric values for the calculations associated with sewage sludge (and FW) anaerobic treatment, bioenergy recovery and final disposal are involved in Table 3. Other parameters and calculation procedures are described in the Supporting Information.

5.1. Sludge production, mass reduction and digestate deep-dewatering

The sludge amounts in different scenarios were calculated and are illustrated in Fig. 11a. The results reveal that the CAS process in Treatment Processes A, C and D produced approximately 1.20 t-DS/d of PS and 0.67 t-DS/d of ES at Q^3 = 10,000 m^3/d. The use of CEPS in Treatment Process B promoted the removal of more SS (BOD_5) from influent. More SS were first settled and interpreted by primary clarifier, which reduced 17.9% ES production (i.e. 0.55 t-DS/d) but came with 41.7% increased PS (i.e. 1.70 t-DS/d). Meanwhile, although the CEPS is able to reduce the pollutant load on the downstream unit minimizing aeration requirements (or oxygen demand) [213], total sludge production in this case rose obviously [214]. In sharp contrast, AnMBR process (E) exhibited "zero" sludge discharge thanks to the omission of primary/secondary clarifier, which would substantially cut the overall operational costs.

Table 3
Base-case assumptions for wastewater properties and treatment efficiencies, and general parameters used in calculating mass-energy balance [54].

Parameters	Values	References
Wastewater characteristics assumed		
Treatment capacity (Q^3)	10,000 m^3/d	
BOD_5	200 mg/L	
COD_{Cr}	480 mg/L	
SS	200 mg/L	
TP	5 mg/L	
TN	40 mg/L	
Effluent quality	BOD_5 ≤10 mg/L, SS ≤ 10 mg/L	
Overall energy consumption	≤0.675 kWh/m^3	
Aeration energy consumption per $BOD_{removed}$	1 kWh/kg-BOD_5	
Energy consumption for aeration	0.4 kWh/m^3	
Sewage sludge (food waste) treatment and bioenergy recovery		
(i) Properties of substrates		
Substrate temperature	12 °C	[203]
Primary sludge (PS)		
DS content	2–6% (4%)	[204]
VS (% of DS)	70%	[205]
Excess sludge (ES)		
DS content	0.6–1% (0.73%)	[204]
VS (% of DS)	75%	[205]
Food waste (FW)		
DS content	17.7%	
VS (% of DS)	93.5%	
(ii) Thermal hydrolysis of excess sludge		
Pretreatment conditions	150–165 °C, 20–30 min, 8–9 bar	CambiTHP™
Specific heat of sludge	4200 kJ/m^3/°C	[206]
(iii) Anaerobic digestion		
Mesophilic	35 °C	
SRT	20 d	
$SRT_{-TH.}$	10 d	
Energy consumed for mixing	0.005 kW/m^3, 20 min mixing/h	[207]
VS $removal_{PS}$	60%	
VS $removal_{ES}$	30%	
VS $removal_{PS+ES}$	50%	
VS $removal_{ES\text{-}TH.}$	~60%	CambiTHP™
CH_4 $yield_{PS}$	0.60 Nm^3/kg-$VS_{removed}$	
CH_4 $yield_{ES}$	0.50 Nm^3/kg-$VS_{removed}$	
CH_4 $content_{PS}$	67.2%	
CH_4 $content_{ES}$	62.5%	
VS $removal_{FW}$	75–85% (80%)	
CH_4 $yield_{FW}$	0.51 Nm^3/kg-$VS_{removed}$	
CH_4 $content_{FW}$	62.5%	
Heating value of CH_4	35.8 MJ/Nm^3-CH_4	[208]
Efficiency of generator	35%	
Efficiency of heat recovery	50%	
Heat loss during operation	150.84 kJ/d-m^3	[209]
(iv) Dewatering		
Chemical conditioner	0.1–0.2-DS%	
Price of chemicals	10 US $/kg	
Energy consumed for dewatering	101.4 kWh/10^3 kg-DS	[210]
DS $content_{control}$	20%	CambiTHP™
(v): Transportation		
Distance (WWTP → land application site)	50 km	[203]
Diesel consumed for transportation	35 L/100 km (3 axle semi-trailer vehicles), 14.1 tons of capacity	[203]
Heating value of diesel	38.4 MJ/L-diesel	
Price of diesel	2.558 US $/Gallon	
(vi): Land application		
Energy consumed for land application	351.68×10^{-3} kWh/kg-DS	[211]

Other assumptions.
(a) For the CAS process (Process A, C, and D): SS removal of 60%, and BOD_5 removal of 40% during primary settling.
(b) For the CEPS-CAS + AD process (Process B): SS removal of 80%, and BOD_5 removal of 50% during chemically enhanced primary settling.
(c) The empirical formula for PS, ES, and FW is $C_{11.2}H_{23.6}O_{5.3}N$, $C_5H_7O_2N$ and $C_{17}H_{29}O_{10}N$, respectively. The theoretical biogas yield and methane content are estimated via the following equation: $C_nH_aO_bN_c$ + [n – 0.25a – 0.5b + 1.75c] H_2O → [0.5n + 0.125a – 0.25b–0.375c]CH_4 + [0.5n – 0.125a +0.25b–0.625c]CO_2 + cNH_4 + cHCO_3^- [212].
(d) For AnMBR: 0.53 t-DS digested sludge production for treating 10,000 m^3 wastewater per day.

Fig. 11b shows waste digestate production after the subsequent mesophilic AD. Digestate residual in Treatment Process A was estimated to be 1.22 t-DS/d. The CEPS in Treatment Process B enhanced the organic capture as PS, which mainly consists of abiotic organic matters possessing higher biodegradability and methane conversion efficiencies compared with ES [213]. But this advantage did not offset the economic-environmental issue triggered by high digestate generation. The amount of waste digested biosolids production in this case reached 1.45 t-DS/d, increases of 18.9% vs. A case. Differing from PS, ES contains large quantities of EPS and microbial cells [145,215], which present physical and chemical barriers to direct AD [54]. TH pretreatment is capable of rupturing gel-like EPS matrix and lysing cells. As a result, the digestate production in Treatment Process C with TH process showed a considerable decline, i.e. 1.04 t-DS/d. The Treatment Process D produced the most waste digestate of 1.58 t-DS/d, because of co-digestion with FW. Nonetheless, the cost-benefit offered by highly stimulated methane recovery could somewhat counteract the shortage. The least digestate production resulted during AnMBR process (around 0.53 t-DS/d [216]), 56.4% lower than Treatment Process A. A unique advantage is that AnMBR can accomplish wastewater stabilization and AD in the same reactor while simultaneously maximizing organics transformation into biomethane and simplifying operating procedure. Hence, for large/medium-sized cities with their shortage of land resources, AnMBR (process E) undoubtedly is superior to CAS process [217].

Dosage of chemical conditioners and corresponding costs for digestate deep-dewatering are closely related to the amount of waste digested biosolids produced. Costs for Treatment Processes A, B, C, D and E, were calculated to be 18.23, 21.81, 15.59, 23.69 and 7.95 US $/d, respectively (Fig. 11c). Compared with the Treatment Process A, the capitals of mechanical dewatering were reduced by 15.0% in the Process C and 56.7% in the AnMBR (Fig. 11d).

5.2. Energy balance and net profit analysis

The energy consumption is often the first priority for a wastewater treatment process. This consumption has been grouped into six categories: aeration, TH pretreatment, AD stirring/heating/heat loss, digested biosolids mechanical dewatering, transport and land application. Fig. 12a shows the energy input distribution in each process. Overall, for all CAS-based processes (Processes A, B, C, and D), aeration was the most important item (3600–4320 MJ/d), representing higher than one third of total energy input; the second energy-intensive item was AD heating, which contributed additional 35–41% to the total consumption depending on the processes. Differing from CAS-based processes, the main energy requirement in AnMBR system (Process E) was stirring/mixing through biogas recirculation (around 0.1 kWh/m^3 wastewater). Due to low membrane cleaning frequencies during low-strength domestic wastewater treatment process (i.e. (i) water back-flushing (1 time/3 months), (ii) citric acid solution back-flushing (riben1 time/6 months, 20 g-citric acid/L, pH 3.0–4.0 adjusted by ammonia solution), and (iii) 0.5-NaClO-Cl% solution back-flushing (1 time/1 year)), the energy consumption for membrane cleaning was ignored in the calculation. Land use was the third in energy requirement, whereas there were no distinct differences among the different treatment processes, except with AnMBR process (Process E). Energy input for other

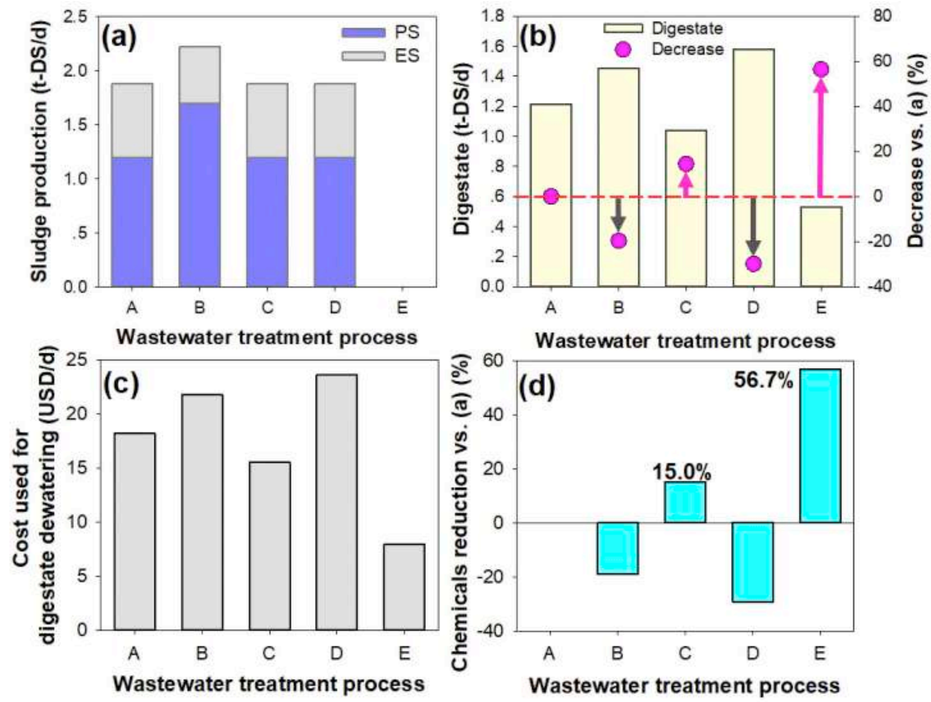

Fig. 11. (a) Primary (PS) and excess sludge (ES) production, (b) waste digestate production and its reduction efficiency relative to the Treatment Process A, (c) costs of chemical conditioners added for digestate deep-dewatering, and (d) chemicals reduction relative to the Treatment Process A.

operations like digestate dewatering, transport was similarly low.

Total energy consumption varied greatly upon the treatment processes. It was estimated to be 10216.1 MJ/d for Treatment Process A. While the CEPS in Treatment Process B reduced aeration energy, the energy of AD heating showed an obvious rise caused by higher sludge production. Thus, the total energy input in this process correspondingly increased to 10679.3 MJ/d. Co-digestion with FW required more energy for heating, which therefore led to the highest consumption of 11686.8 MJ/d for Process D. A very exciting phenomenon was that Treatment Process C with TH exhibited comparatively low energy demand (7190.0 MJ/d) although TH pretreatment required roughly 923.9 MJ/d energy for disintegrating ES flocs. This was mainly attributable to highly efficient waste heat reuse to pretreat the next batch of ES, shortened SRT, increased sludge bio-degradability and biogas production, no need of AD heating, and improved digestate dewaterability [54]. Besides, as expected AnMBR process (E) had the least energy consumption of 4591.0 MJ/d, about 1.57–2.54-fold lower than other processes.

Net energy output was then calculated based on the energy consumption and the energy benefits (heat + electricity) derived from methane production. As illustrated in Fig. 12b, capturing more SS for AD via the CEPS promoted methane production by 22.7%, comparable to by 22.8% of TH pretreatment. In a recent work conducted by McCarty [218], they calculated 28–30% increase in methane production by utilizing chemically enhanced primary removal to send 100% of influent SS into the digester in two nitrogen-removal treatment systems (i.e. recycle N/DN (Nitrification/Denitrification), and recycle N/DN plus side-stream). Increment of up to 186.4% in methane yield occurred in AnMBR process (Process E), followed by the Treatment Process D (by 145.1%). According to the energy balance results in Fig. 12c, all scenarios considered gave a positive net profit, indicating that the CAS process can become economically feasible while in conjuration with sludge AD. Sludge fermentation could produce sufficient bioenergy to satisfy the need for wastewater-sludge treatment. In spite of that, the most economically feasible should be AnMBR process (28425.6 MJ/d), with more than 70% increase in net profit than any other A through D system. The bioenergy recovered in the form of biogas can be utilized either at a power generating facilities or hot-water boiler, covering the for the WWTPs or the residents in the vicinity [219]. Apparently, from the energetic, economic and environmental perspective of view, AnMBR process is more sustainable and of greater promise for wastewater treatment. Of course, in order to make a more accurate quantification of cost-benefits derived from different wastewater treatment scenarios, other factors such as maintenance, local circumstance of labor, footprint, land price, and market for energy exchange and reclaimed water reuse should also be studied in greater detail.

5.3. Full-scale application status and development of a new-generation AnMBR platform towards biowaste refinery, environmental sustainability and low-carbon society

Up to date, the full-scale AnMBRs have already been built in some countries due to the great performance and the net energy income. Theoretical research and field experiments have corroborated the outstanding performance of AnMBR to harvest biomethane from diverse waste streams. One of the most popular AnMBR suppliers can be the Kubota Corporation of Japan. Since 2000, Kubota Corp. has established around 26 full-scale worldwide AnMBR installations for treating a wide range of biowastes [116,219,220]. In a project, two AnMBRs equipped with 150 Kubota flat-sheet membranes (0.8 m^2 each) in each reactor were constructed for digesting dairy processing wastewater in Kobe, Japan. Each AnMBR had an effective volume of 100 m^3 and a total treatment capacity of 30 t/d while producing 880 m^3/d of biogas. Biogas, after desulfurization, was used in boilers, saving 95000 L of A-grade oil [93,220]. Kubota AnMBR system offers many merits such as 2/3–4/5 reactor volume reduction, 3–5-fold concentrated biomass, low minor components (H_2S) in biogas, real-time discharge of methane fermentation inhibitors (ammonia) with the permeate and high process stability [219]. Another representative MBR manufactory can be ADIR Systems Inc. In early 2008, ADIR Systems Inc. designed and installed the first AnMBR system with a treatment capacity of 475 m^3/d for Ken's Foods company located in Massachusetts of USA, to treat the salad dressing and BBQ sauce wastewater [221]. In another project, ADIR Systems upgraded the anaerobic wastewater treatment system of a

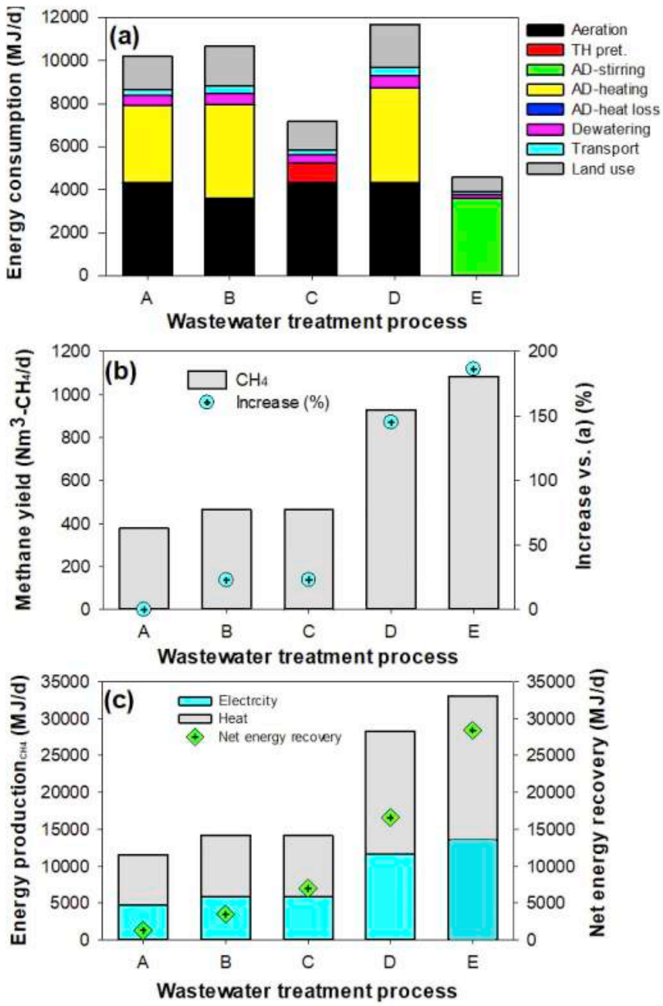

Fig. 12. Energy input distribution (a), biomethane production (b) and net energy recovery (c) in five different wastewater treatment processes.

confectionery manufacturing plant located in Pennsylvania of USA, to AnMBR to comply with discharge standards.

In spite of the superior performances and such progress in upscaling outlined above, the application of AnMBR, especially in full-scale mode, is still facing difficulties and grave challenges. The first and the most severe issue can be membrane fouling. While a myriad of approaches, such as *ex-/in-situ* physical, chemical, biological cleaning, membrane modification, and functional membrane fabrication, have been proposed as outlined before, membrane fouling still remains unresolved due to the complex biological, physiochemical processes (e.g. gelatin biopolymer secretion/adhesion, cell attachment, flocculation/coagulation of small flocs, and precipitation of inorganic crystals (e.g. $MgNH_4PO_4{\cdot}6H_2O$, $K_2NH_4PO_4$, $CaCO_3$, etc.)). Membrane fouling control continues to be the research priority in future [25,120,158]. An alternative fouling control means can be bioelectrochemical system (BES), a newly emerging platform technology proposed during the last decade, which has shown great promise for simultaneous waste biorefinery, chemicals synthesis, clean electrofuels generation and biosensors [222–225]. BES embodies versatile features such as accelerated degradation of organic substances provoked by anode-respiring bacteria in the anode (or exoelectrogen *Geobacter*), and value-added biomolecules production by electrotrophs in the cathode driven by a small external power source (i.e. DC power, solar, wind, etc.). Exoelectrogen in the anode can oxidize a wide range of substrates from simple short-chain fatty acids to real biowastes while generating electrons and protons [226–228]. For instance, BES has been employed in enhancing anaerobic digestion of various kinds biowastes such as pig slurry [229], sludge, food waste [230], *Egeria densa* [231], and beer wastewater [232], for bioenergy recovery. Meanwhile, the electrons and protons reach the cathode via an external circuit and combine to form H_2 gas or to form low-carbon electrofuels such as CH_4, acetate and formate by electro-reducing CO_2 with electrotrophs colonizing in the cathode as bioelectrocatalysts [233–235]. With CO_2 as the sole carbon source, Zhen et al. [236] observed a high methane production of 75.8 mL/L/d at the cathodic potential of -0.9 V vs. Ag/AgCl.

In consideration of those merits described, BES has been applied in a broad range, such as wastewater decontamination for bio-H_2 generation (electrohydrogenesis), CO_2 electroconversion to carbon-natural electrofuels or chemical commodities (bioelectrosynthesis) [237–239]. Also, BES has been successfully coupled with the conventional anaerobic continuous stirred tank reactor (BES-CSRT system) to mediate process stability and stimulate biomethane recovery from a myriad of waste streams (Fig. 13) [240–244]. It has been demonstrated that the combination with BES is able to regulate microbial community, eliminate the VFAs build-up and expedite organic removal and methane production [231,245] while simultaneously enhancing the digestate dewaterability [230]. In this regard, when BES is integrated into AnMBR system (i.e. BES-AnMBR), some exciting outcomes can be expected as well: (i) *in-situ* fouling control via anodic oxidation of gelatin biopolymers (EPS/SMP), (ii) enhanced biomethane productivity through syntrophic interactions between fermenting anaerobes and electroactive bacteria (e.g. *Geobacter*) (Fig. 14a), (iii) electroreduction of CO_2 in biogas and simultaneous methane purification via bioelectromethanogenesis [246,247] (e.g. hydrogenotrophic methanogens via (i) $CO_2 + 8H+ + 8e^- \rightarrow CH_4\ (g) + H_2O$, $E_{cat} = -0.244$ V vs. SHE [223,238,248]; and (ii) $2H^+ + 2e^- \rightarrow H_2\ (g)$, $E_{cat} = -0.414$ V vs. SHE,

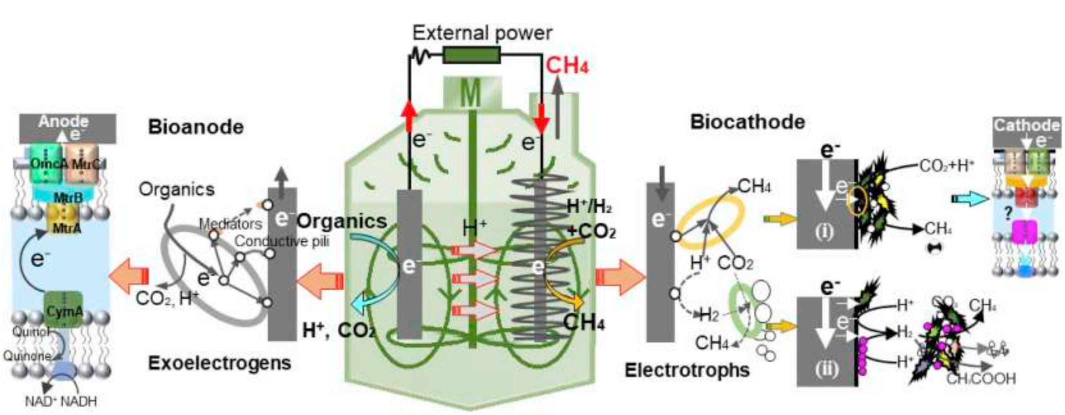

Fig. 13. Schematic illustration of possible metabolic pathways of organic compounds and extracellular electron transfer pathways involved in a combined BES-CSTR system [222,247].

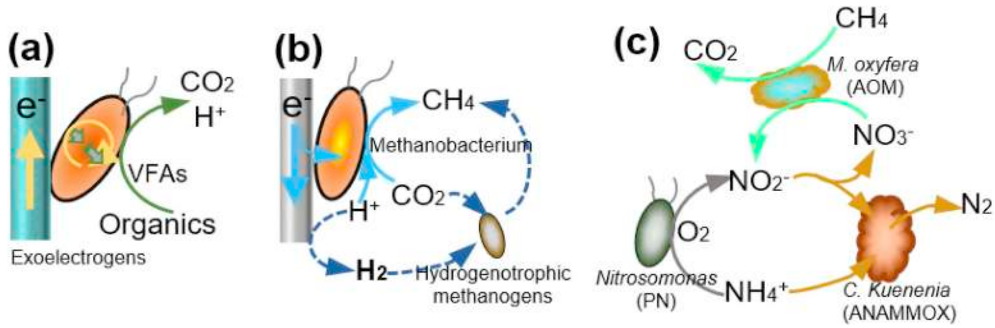

Fig. 14. Observed interactions between key populations involved in the BES (a–b), PN/A and AOM processes (c).

and $4H_2 + CO_2 \rightarrow CH_4$ (g) + $2H_2O$, $\triangle G^{0'} = -131$ kJ/mol) [249–251] (Fig. 14b). Besides, part of cations in digestate liquid can also migrate towards biocathode under a poised electric field and then be removed near to cathode through *in-situ* chemical precipitation. This cannot only accumulate and recover a diversity of valuable elements, such as struvite crystals [252,253], heavy metals (e.g. Cd, Co, Cu, Pb, and Zn) [254–256] and scale-forming ions (e.g. Ca^{2+}, and Mg^{2+}) [257], but alleviate inorganic pore blocking associated with chemical precipitators in AnMBR process. Obviously, the integrated BES-AnMBR system can be an attractive and environmentally sound option in the view of sustainability. However, so far very few studies have recognized the potential of BES in enriching specific functional microorganisms and/or remvoing some soluble by-products for enhancing methane recovery and addressing membrane fouling [38]. More importantly, the bioelectrocatalytic behaviors of the newer processes in resources recycling, biogas upgrading, waste digested solids reduction are still not well elucidated [39]. Further work to speed the development and practical applications of the newer and proof-of-concept processes would appear to be more urgent.

5.4. Post-treatment of AnMBR effluent for efficient nitrogen removal

Another concern to address is high effluent ammonia-nitrogen in AnMBR system due to the degradation of organic nitrogen and ammonification [258]. AnMBR usually possesses the poor removal efficiency of NH_4^+-N compared to anoxic/aerobic process [202,259]. In order to meet discharge standards, the permeate should be post-treated. One of the most innovative nitrogen removal processes can be Partial Nitritation/Anammox (PN/A), that is partial oxidization of ammonia first by aerobic ammonium oxidizers (AOB) to nitrite (e.g. genera *Nitrosomonas* and *Nitrospira*), followed by ANAMMOX (Anaerobic Ammonium Oxidation) (e.g. genera *Candidatus Kuenenia* and *Candidatus Brocadia*) by using the produced nitrite as electron acceptor to oxidize the remaining ammonia to dinitrogen gas [260,261] (Fig. 14c). The PN/A process entails many outstanding advantages compared to the traditional N/DN process such as less oxygen demand and reducing agent supply, and lower sludge production and N_2O emissions [260]. Moreover, when AOB and ANAMMOX take place in the same reactor (i.e. single-stage PN/A process), they can offer additional benefits like smaller footprint and less investment cost [262,263]. In view of that, the combination with single-stage PN/A process can be a good choice to cover the shortage of AnMBR system. Of course, there are still many critical challenges associated with the applications of single-stage PN/A process, particular with the urgent needs for the new methods to accelerate the growth of ANAMMOX bacteria, and for the efficient strategies to suppress or out-select nitrite oxidizing bacteria (NOB) while balancing AOB and ANAMMOX bacteria. To overcome such issues, several novel strategies have been proposed in the literature, such as adding hollow cylinder carrier made of hydrophobic polypropylene resin [264], fixing combined carriers consisting of plastic fibers, rings and center ropes [265], and developing syntrophic micro-granules [263], and they indeed upgraded the symbiosis of AOB and ANAMMOX bacteria and nitrogen removal. Nonetheless, how to apply those strategies in a real-world scenario remains a vital problem, and the efforts should be redoubled to confirm their real potentials.

Additionally, aside from dinitrogen gas ANAMMOX process also yields nitrate as the final product ($NH_4^+ + 1.32\ NO_2^- + 0.066HCO_3^- + 0.13H^+ \rightarrow 1.02N_2 + 0.26NO_3^- + 2.03H_2O + 0.066CH_2O_{0.5}N_{0.15}$) [266], leading to the incomplete nitrogen removal (< 90%). The removal of nitrate can be further provoked by AOM (Anaerobic oxidation of methane) followed by second-cycle ANAMMOX process, considering the fact that the *bacterium*

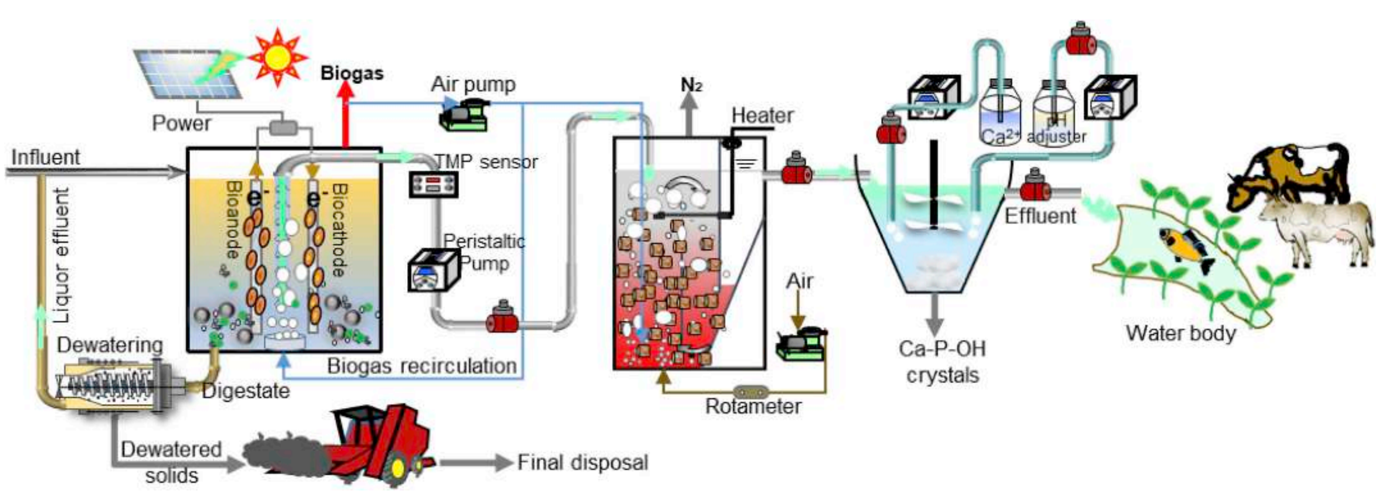

Fig. 15. Integrated Multistage Bio-Process (IMBP) consisting of solar-driven BES, AnMBR, PN/A, AOM and chemical precipitation.

Candidatus '*Methylomirabilis oxyfera*' is able to couple AOM to nitrate reduction to nitrite ($CH_4 + 4NO_3^- \rightarrow CO_2 + 4NO_2^- + 2H_2O$, $\triangle G^o = -503$ kJ/mol-CH_4) [267]. In a combined AnMBR-single-stage PN/A process, methane can be provided by recirculating biogas generated from AnMBR to drive AOM reaction (i.e. "integrated AnMBR-single-stage PN/A-AOM process") (Fig. 14c). In this context, nitrogen removal efficiency would be improved considerably and a theoretically complete removal can result while a dynamic balance is established among AOB, ANNOMOX and AOM. The inclusion of AOM also offers a unique merit, this is the reuse and recovery of the dissolved methane present in the AnMBR effluent. It is well-known that the high dissolution of methane in the effluent is an unavoidable technical issue for the AnMBR-based technologies [23,123,268]. Not only can a large fraction of effluent-dissolved methane reduce bioenergy recovery but exert a severe influence on global warming. Accordingly, the occurrence of AOM during N/A process will alleviate the environmental issues associated with the dissolved methane (e.g. greenhouse gas emissions, explosive risk), upgrade the biomethane recovery and simultaneously maximize nitrogen removal. Also, strategies to manage phosphorous are required to recycle the value-added substances from waste streams to the upmost extent [269,270]. Apart from the electrochemical crystallization by BEC as claimed before, phosphorous recovery can be accomplished as well by Polyphosphate Accumulating Organisms (PAO) [271], or via chemical precipitation as calcium phosphate (hydroxyapatite, $Ca_{10}(OH)_2(PO_4)_6$)Ca–P) and struvite ($MgNH_4PO_4 \cdot 6H_2O$) [272–275]. Considering the outstanding effectiveness of each process, it can be imagined that design and development of an Integrated Multistage Bio-Process (IMBP), consisting of BES, AnMBR, PN/A, AOM and biological/chemical precipitation, appears to be of great significance to concurrent waste biorefinery, membrane fouling control, biomethane generation, dissolved methane recovery/reuse, nitrogen removal and phosphorous recovery from the energetic, economic and environmental perspectives of view. Based on this, the new process, i.e. so-called IMBP, was proposed and is illustrated in Fig. 15. In view of current findings and state-of-the-art, the advancement of such novel concept will surely enhance the industrial competitiveness of AnMBR-based technologies in real-world scenarios, finally promoting the establishment of the energy-sustainable and low-carbon society. However, no information is still available regarding the real behaviors of the IMBP platform, and a comprehensive investigation is urgently needed. Deep dewatering and final disposal of waste digested biosolids will also have to be considered for their potentially negative impact on the eco-sustainability of the process.

6. Concluding remarks and future perspectives

In this review, recent research advances and new discoveries of AnMBR technique (i.e. types of substrates, operation conditions, long-term performance, membrane materials, membrane fouling mechanisms, key foulants, fouling control/cleaning measures, etc.), and current application status were systematically summarized and critically reviewed. AnMBR has the ability to treat a broad range of waste streams in an extremely high conversion efficiency while recovering bioenergy, and consequently has gained ever-increasing attention from researchers worldwide. Continuous production of massive biowastes and irresistible desire to green and carbon-less society in recent years greatly motivate the intensified research efforts in this area. Up to now, numerous attempts have been carried out to push forward the applications of such options from bench experiments to real-world scenarios. The recent commercial availability of the biorefinery approach represents a major advance for this option. Furthermore, five different conventional/unconventional wastewater treatment processes were designed and evaluated to determine their strengths, potentials and limitations. The comprehensive comparison analysis provides a substantially compelling evidence that AnMBR could exhibit versatile superiorities over CAS-based processes with regards to good permeate quality, less digestate residuals, low operational costs, net profit/energy output, and outstanding economic and environmental benefits.

Although the great progress has been made, there are still tough challenges that need to be confronted for this technology, particularly for severe membrane fouling, biogas upgrading, highly dissolved methane, effluent ammonia discharge, phosphorus loss, etc. To address the above problems, a new-generation process named "Integrated Multistage Bio-Process (IMBP)", which is constituted of BES, AnMBR, PN/A, AOM and biological/chemical precipitation units, was proposed in this article. Not only can such novel integration accelerate the transformation of biopolymeric components and CO_2 electro-methanogenesis by means of bioelectrochemical regulation, accomplish *in-situ* fouling control and upgrade biogas, but also remove ammonia to a greater extent by PN/A-AOM pathway while simultaneously re-utilizing dissolved methane, and recover phosphorus as HAP-rich nutrients. Despite the uncertainties about whether this approach possesses the powerful potential to dominate the future, but most surely, this hybrid concept will enhance the deployment and industrial competitiveness of AnMBR-based technologies in real-world scenarios, facilitating the establishment of the energy-sustainable and low-carbon society. Of course, new efforts dedicated is still a pressing need at present to demonstrate the feasibility of this integrated biorefinery approach. It can be anticipated that this review article will open up research opportunities to integrate with other newly emerging processes to develop robust, multifunctional and marketable AnMBR-based technologies and maximize biowastes valorization for biofuels.

Declarations of interest

None.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rser.2019.109392.

Note: BOD = biochemical oxygen demand; CFV = cross-flow velocity; COD = chemical oxygen demand; CPE = chlorinated polyethylene; FO = forward osmosis; MF = microfiltration; MWCO = molecular weight cut-off; OLR = organic loading rate; PE = polyethylene; PES = polysthersulfone; PTFE = poly-tetrafluoroethylene; SCOD = soluble chemical oxygen demand; T: temperature; TCOD = total chemical oxygen demand; TN = total nitrogen; TP = total phosphorus; TS = total solids; UF = ultrafiltration; VLR = volumetric loading rate; VS = volatile solids.

Note: CEB: chemically enhanced backflush; CFV: cross-flow velocity; EDTA: ethylene diamine tetraacetic acid; FW: food waste; GAC: granular activated carbon; HD: hydrodynamic; LMH: flux range, $L/m^2/h$; NF: nanofiltration; OFMSW: organic fraction of municipal solid waste; QQ: quorum quenching; SAD_m: specific aeration demand; US: ultrasound; WAS: waste activated sludge.

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