Electronic Supplementary Material (ESI) for Environmental Science: Water Research & Technology.

Supporting information

A critical review on advanced anaerobic membrane bioreactors (AnMBR) for wastewater treatment: advanced membrane materials and energy demand

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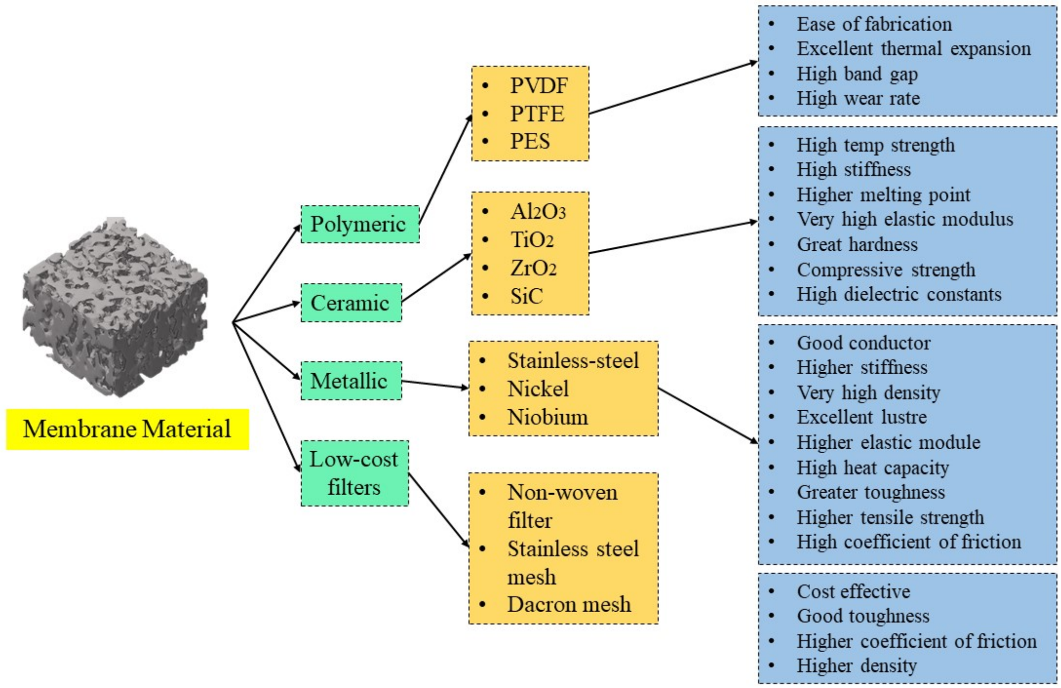

Figure S1. Membrane materials and their properties.

S1. Principle of AnMBR technology

S1.1 Configuration of AnMBR

The configurations of AnMBR are categorized according to the position of the membrane module in the system and the way of combination. The position of the membrane module determines the energy demand, footprints, and operation cost of the system [159]. The position of module differentiates three various types of AnMBR configuration (Figure S2), as mentioned below. In AnMBR, if the membrane module is fixed outside of the reactor and permeate is collected through a recirculation pump, the setup will be called side-stream AnMBR. In this format, the flow velocity ranging from 2-4 ms^{-1} is required for the optimal operation, otherwise it is easy to form a permanent cake layer on the membrane surface. In side-stream AnMBR, anti-fouling strategies such as cleaning, sludge treatment are convenient. However, in side-stream AnMBR, extra energy is required for maintaining volumetric flow, continuous cleaning, and transmembrane pressure [1], which is not economical as compared to other configurations. Submerged AnMBR, a possible alternative configuration with various pressure-driven crossflow membranes, demand less energy and mild operating conditions [2]. A mechanical stirrer is installed to avoid the quick formation of the cake layer on the membrane surface and biogas blockage. Less energy demand and less shear stress on microbial are the major advantages of submerged configuration. The membrane in submerged AnMBRs has high fouling potential and is competitive in treating low strength wastewater [3, 4]. Thus, most of the scientists adopt submerged AnMBRs for research purposes. In third case, the membrane module is positioned inside the tank (external tank or biological tank) and permeate is obtained by suction. In this type of AnMBRs, the aerators are placed adjacent to the module to reduce cake formation by turbulence. The membrane in external submerged AnMBR has less fouling potential; competitive in higher permeability and lower flux [5]. Moreover, this alternation yields higher stability, lower toxicity shocks and fewer energy demands (1 KWh/m^3) as compared with side-stream AnMBRs (6 KWh/m^3) [6]. Conversely, an extra pump in this configuration increases the energy consumption accompanied by modifying the sludge characteristics. To reduce the cost and membrane fouling, the AnMBRs process is generally integrated with primary-treatment systems. For instance, advanced oxidation processes and low-cost filters can be applied as influent pre-treatment methods for submerged AnMBR to overcome the fouling issues.

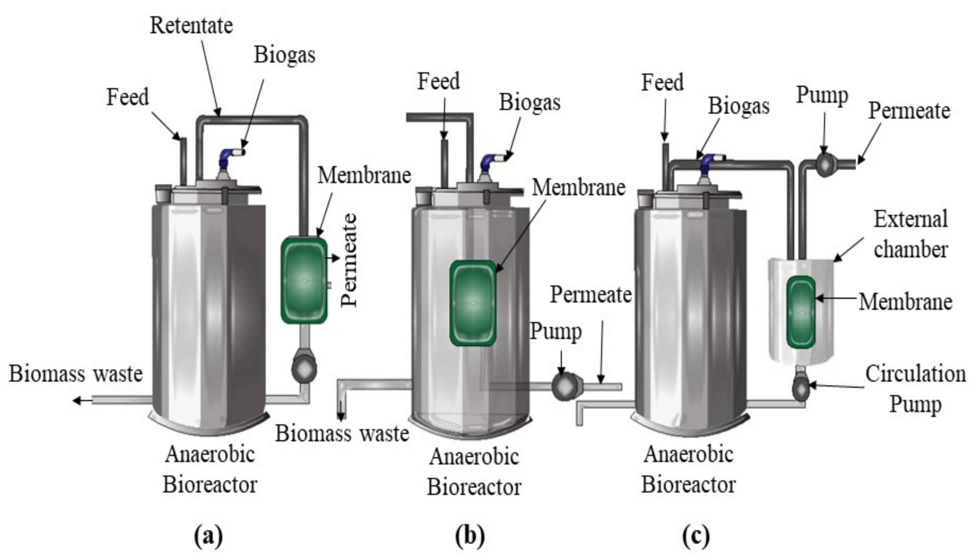

Figure S2. Different types of bioreactor configurations (a) side-stream AnMBR (b) submerged AnMBR (c) external submerged AnMBR.

Secondly, various unique combinations have been installed to minimize the fouling and decrease the energy demands, such as: (1) Anaerobic fluidized-bed membrane bioreactor

(AFMBR): A combination of biofilm carriers with biogas sparging was reported to control the biogas flow rate and to lessen the membrane fouling, as shown in Figure S3a [7]. Kwak et al. developed a novel AFMBR with a liquid recirculation particle-sparging system linked with membrane filters, as displayed in Figure S3b [8]. The process was unique, indicated by enhanced mass transfer characteristics along with excessive microbial species. The addition of GAC on the membrane surface is termed fluidized media, which controls the membrane biofouling and minimises energy demand [9, 10]. Energy required for fluidization in AFMBR was reported 0.028 kWh/m^3, which was significantly lower than conventional submerged AnMBRs; (2) Anaerobic submerged rotating membrane bioreactor (AnSRMBR): An innovative reactor was introduced to overcome this issue as shown in Figure S3d [11, 12]. An electric motor was installed to rotate the immersed membrane in a fixed axis to improve filtration capacity and reduce fouling through the shear force generated on the membrane surface. Moreover, cross flow velocity is produced by membrane module rotation. Thus, antifouling property due to rotary membrane, filtration system and achieved economical cross flow velocity enlist this configuration as significant in comparison with conventional cross flow systems; (3) Anaerobic dynamic membrane bioreactor (AnDMBR): The dynamic membrane technology was introduced to enhance membrane flux, and to reduce membrane fouling and the cost of membrane configuration [13]. The solid/liquid separation is accomplished by supported filter cake layers. In this system, the major issue of membrane fouling commutes into its beneficial. After being fouled, the formed cake layer is being replaced by new deposited layer, which makes it easier and cost-effective technique. Moreover, the lower CFV requires lesser input energy and resulting into improved methanogenesis activity and greater biogas production (0.34 $LCH_4/gCOD_{removed}$); (4) Anaerobic electrochemical membrane bioreactor (AnEMBR): A novel configuration of anaerobic filtration with microbial electrolysis cell (MEC) was recommended by integrating a nickel-based hollow fiber membrane (porous and highly conductive) [14]. The integrated porous membrane was featured to treat water in a single-stage and to recover biogas. This new configuration is defined anaerobic electrochemical membrane bioreactor (AnEMBR), as presented in Figure S3e. On applying higher voltage, the resultant higher rate of hydrogen evolution enhances gas bubble formation at membrane surface, resulting into lower membrane fouling. Moreover, the GAC as scouring media in fluidized bed membrane bio-electrochemical reactor (MBER) has been utilized to reduce the membrane fouling (Figure S3f) [15]. Thus, it can be stated that this technology has benefits of large bioenergy production and high effluent quality. To control membrane fouling, the anode oxidation of substrate (low organic loading) helps to minimize the fouling; (5) Anaerobic osmotic membrane bioreactors (AnOMBRs): osmotic differences drive the water molecules from feed solution with lower osmotic pressure permeating through a semi-permeable membrane into a solution with higher osmotic pressure [16]. This technique doesn't need any input energy to run filtration in comparison with conventional systems. A significant amount of biogas (0.25–0.3 $LCH_4/gCOD_{Removed}$) have been obtained through this technique. However, future research is still needed for this technique to resolve the effluent biogas problem; (6) Anaerobic membrane distillation bioreactors (AnMDBRs): combine the membrane distillation process with the bioreactor operating at the phenomenon of temperature difference (Figure S3h). Microporous and hydrophobic membrane is utilized for the transportation of water vapours to other side in the form of water. In this process, methane gas can be accomplished as input energy and remaining can be extract and recover easily. Thus, the permeate biogas is comparatively higher than other forms of AnMBRs, due to lower dissolved methane. Moreover, to control eutrophication, this system also has strength for completely deletion of phosphorus. However, further studies are required to deal with methane generation and other challenges.

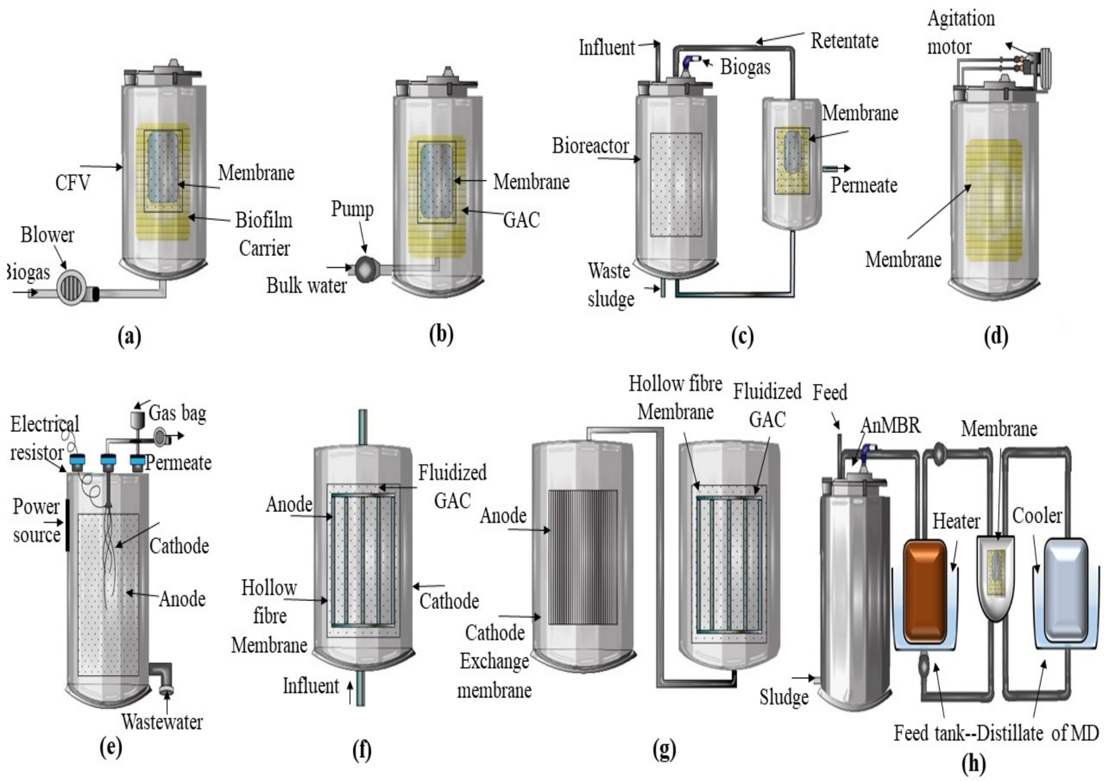

Figure S3. Unique AnMBRs combination to reduce energy demand and fouling: (a) Biogas-particle sparging; (b) Liquid recirculation particle-sparging; (c) Anaerobic electrochemical membrane bioreactor; (d) Anaerobic cross flow-particle sparging MBR; (e) Anaerobic rotating MBR; (f) Individual fluidized MBER; (g) Hybrid MFC-MBR system; and (h) Anaerobic membrane distillation bioreactors (Reproduced and modified from Maaz et al. (2019) with permission from the publisher) [2]

S1.2 Key stages of anaerobic digestion

Anaerobic digestion is a sequence of processes by which microorganisms breakdown the biodegradable materials under anoxic conditions. Anaerobic digestion comprises four key biochemical phases, including, hydrolysis, acidogenesis, acetogenesis and methanogenesis, as illustrated in Figure S4. The profile of these key phases determines the performance of anaerobic degradation process [17, 18].

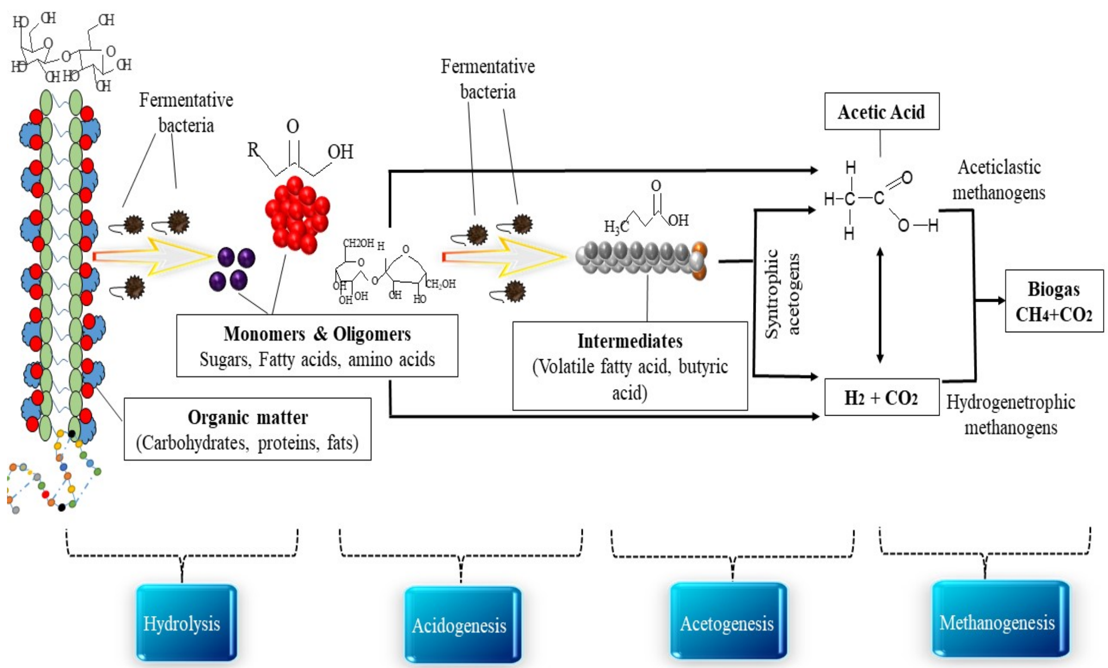

Figure S4. The simplified scheme of pathways in anaerobic digestion.

The anaerobic digestion starts with the bacterial hydrolysis of higher molecular weight compounds and insoluble organic substances like proteins, fats, and carbohydrates into soluble by-products such as sugars, fatty acids, and amino acids. In this phase, strict anaerobes such as facultative bacteria, bacteroides and clostridia are the dominating flora [19]. The hydrolysed monomers are further converted into alcohols, carbon dioxide (CO_2), volatile fatty acids VFAs and hydrogen in the following phase, acidogenesis [20]. The long-chain VFAs can be further transformed into CO_2, acetic acid and hydrogen in the third phase, acetogenesis [21]. These intermediate products are then finally converted into methane during the methanogenesis phase. Methanogenesis, the slowest phase of the anaerobic digestion process, is identified as the most critical one. On the other hand, the first three phases (hydrolysis, acidogenesis, acetogenesis) are fast-growing and responsible for biomass accumulation, decreased pH, and foaming [16]. In this whole process, methane production is a two-way process. Approximately 75% of methane is produced by aceticlastic methanogenesis and the remaining 25% is from hydrogenotrophic methanogenesis via converting CO_2 and hydrogen (H_2) into methane and CO_2. The methanogenic activity is the control step and is highly dependent on methane formers and substrate for methane yield. Meanwhile, the change in methanogenic activity also leads to less collection or blockage of organic load. The methanogenesis in this phase is derived from methanogens, which belongs to the archaea group [18]. The slow growing tendency of anaerobic sludge under low organic sludge feeding is the main hurdle in implementation of anaerobic digestion to sewage treatment.

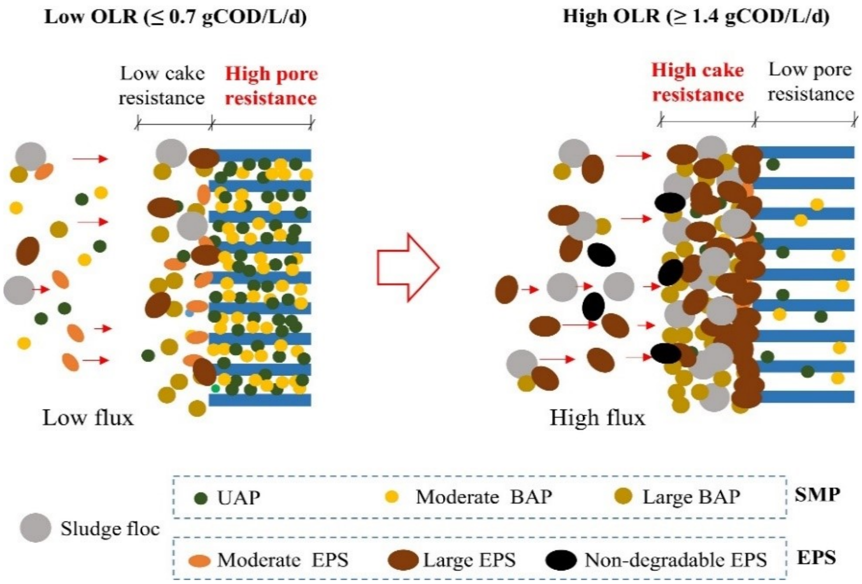

Figure S5. Fouling behaviour of SMP and EPS at low and high OLRs. (copied from Chen et al. (2017), with permission from the publisher) [22].

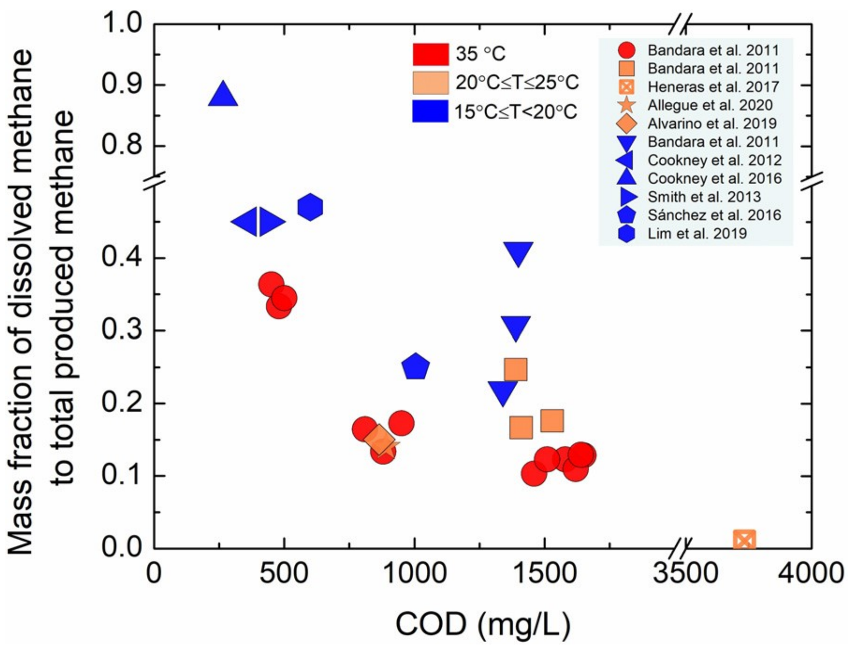

Figure S6. Mass fractions of dissolved methane to the total produced methane. (Copied from Li et al. (2021), with permission from the publisher) [23].

Table S1. Methane loss estimation.

Reactor type	Scale	Degree of supersaturation	Average CH_4 lost in effluent (%)	Temp (°C)	Ref.
AnMBR	Bench	1.5	40–50	15	[24]
AnMBR	Pilot	Not reported	19	22	[25]
SAnMBR	Pilot	1.009	43	33	[26]
SAnMBR	Pilot	1.007	46	21	[26]
AnMBR	Bench	1.0	88	25	[27]
UASB	Pilot	6.9	85	28	[28]
Fixed Film	Pilot	4.3	81	29	[29]

*AnMBR, Anaerobic membrane bioreactor; SAnMBR, Submerged anaerobic membrane bioreactor; UASB, Upflow Anaerobic Sludge Blanket.

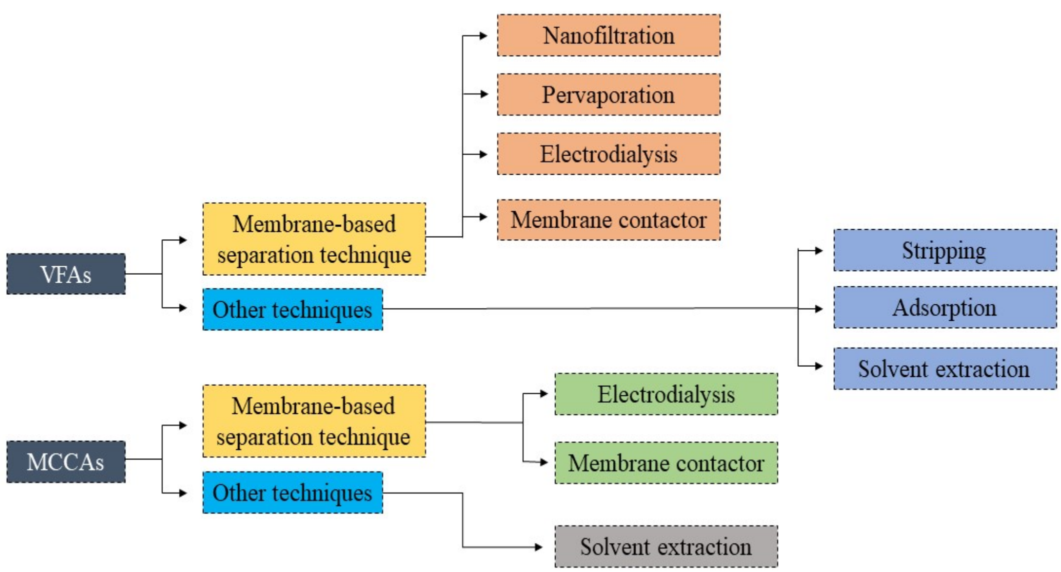

Figure S7. Diagram of various methods to extract VFAs.

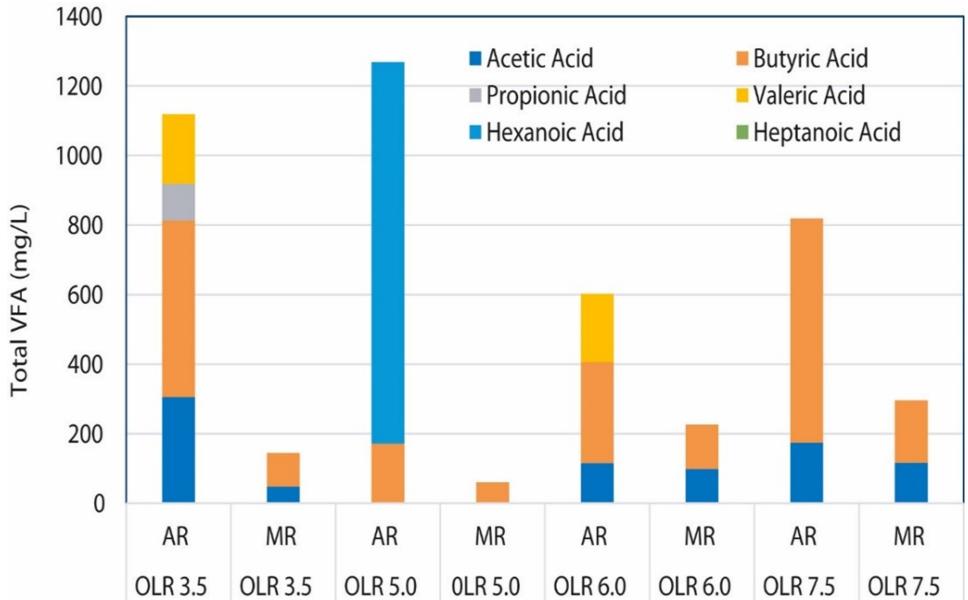

Figure S8. Effect of various OLR concentrations on the VFA accumulation within the methanogenic reactor (MR) and acidogenic reactor (AR). (copied from Mahat et al. (2021), with permission from the publisher) [30].

S2. Pathway to calculate the energy demand of AnMBRs

The equations to calculate the aforementioned seven parameters are given below. The heating, $E_{H,rec.}$, is estimated according to Eq. (4.4).

$$E_{H,\ rec.} = \frac{\rho \cdot Q \cdot \gamma \cdot (T_r - T_{air})}{Q \cdot VS} \quad (4.4)$$

where ρ (g/L): influent density (1000), Q (L/M^3): influent flowrate, γ (J/g-VS/°C): specific heat of the influent (4.18), T_r (°C): reactor temperature (37), T_{air} (°C): air temperature (10), VS: concentration of volatile solid.

The energy demand in fouling control strategies (E_B), can be further classified based on scouring methods, including gas and particle sparging, and the rotating membrane. The biogas sparging AnMBR requires highest energy demand followed by particle sparging AnMBR and rotating membrane AnMBR, respectively [31].

In biogas sparging AnMBRs, the biogas and sludge circulation mainly consume energy, denoted by E_B and can be estimated according to Eq. (4.5).

$$E(KJ/g - vs) = \frac{P_p}{J} \times \frac{10^3 L}{m^3} \times \frac{1}{VS\ (g - VS/m^3)} \times 3600KJ/KWhs \quad (4.5)$$

Where P_p: the power of blower per membrane surface area, J: flux (L/m^2/h).

Secondly, the energy demand in biogas sparging AnMBRs is providing cross-flow velocity (CFV), E_{CFV}, is estimated according to Eq. (4.6)

$$E_{CFV}(KJ/g - vs) = \frac{P_{CFV,m}}{\xi_p J A_m} \times \frac{10^3 L}{m^3} \times \frac{1}{VS\ (g - VS/m^3)} \times 3600KJ/KWhs \quad (4.6)$$

Where $P_{CFV,m}$: required power to pump the sludge, ξ_p: recirculation pump efficiency. Thus, total energy demand for biogas sparging is calculated with the sum of Eq. (4.5) and (4.6).

Energy demand for particle sparging is associated with GAC fluidization and is estimated according to Eq. (4.7).

$$E_{GAC}(KJ/g - vs) = \frac{P_p}{\xi_p J A_m} \times \frac{10^3 L}{m^3} \times \frac{1}{VS\ (g - VS/m^3)} \times 3600KJ/KWhs \quad (4.7)$$

Where P_p: power requirement of the recirculation pump.

Energy demand for the rotating membrane AnMBR is assessed according to Eq. (4.8).

$$E_{RM}(KJ/g - vs) = \frac{P_{r,m}}{\xi_p J A_m} \times \frac{10^3 L}{m^3} \times \frac{1}{VS\ (g - VS/m^3)} \times 3600KJ/KWhs \quad (4.8)$$

Where: $P_{r,m}$: motor power for ignition, A_m: membrane surface area, J: flux.

The energy demand for extracting the permeate, $E_{E, per}$, is estimated according to Eq. (4.9).

$$E_{E, per}\,(KJ/g - vs) = \frac{m * g * h_{MBR} \times mgh_{CSTR} \times mgh_{TMP}}{m \times \eta} \times \frac{1kJ}{1000J} \times \frac{1000/L}{VS\,(g - VS/L)}$$

(4.9)

Where h_{MBR}: liquid height of membrane unit, h_{CSTR}: liquid height of CSTR, TMP: is trans-membrane pressure, η: pump efficiency.

The sludge cycle, E_{Slu}, is estimated according to Eq. (4.10)

$$E_{slu, per}\,(KJ/g - vs) = \frac{x(m * g * h_{MBR} - m * g * h_{CSTR})(J)}{m \times \eta} \times \frac{1kJ}{1000J} \times \frac{1000/L}{VS\,(g - VS/L)}$$

(4.10)

Where x: sludge recycle time.

The energy consumed in terms of the heat loss, $E_{H, loss}$, is estimated according to Eq. (4.11)

$$E_{H, loss} = \vartheta - E_H = \frac{T_{water, b} - T_r}{T_{water, b}} \cdot E_H \qquad (4.11)$$

Where ϑ: percentage of heat recovered and $T_{water, b}$: temperature of water bath.

The energy for mixing, E_{Mixing}, is estimated according to Eq. (4.12)

$$E_{mixing} = \frac{V \cdot \omega}{Q \cdot VS} \qquad (4.12)$$

Where ω: energy consumption for mixing.

The energy for feeding, $E_{Feeding}$, is estimated according to Eq. (4.13)

$$E_{Feeding}(KJ/g - vs) = \frac{(m * g * h_{CSTR} - \frac{1}{2} m * g * h_{sub})(J)}{m \times \eta} \times \frac{1kJ}{1000J} \times \frac{1000/L}{VS\,(g - VS/L)}$$

(4.13)

Where h_{sub}: liquid height of substrate tank.

The following equation is widely used to estimate the individual energy demand of blowers and pumps in submerged AnMBRs [32].

$$P_B\left(\frac{J}{S}\right) = \frac{M.R.T_{gas}}{(\alpha - 1).\ \eta_{blower}} [(\frac{p_2}{p_1})^{\frac{\alpha - 1}{\alpha}} - 1] \qquad (4.14)$$

Where P_B (J/S): blower power required, M (mol s^{-1}): flow rate of biogas, R ($Jmol^{-1}K^{-1}$): constant of gas, P_1 (kPa): input pressure, P_2 (kPa): output pressure, T_{gas} (K): reactor temperature, α: adiabatic index, η_{blower}: blower efficiency.

$$P_g\left(\frac{J}{S}\right) = q_{imp} \cdot \rho_{liquor} \cdot g \cdot \frac{\{[(\frac{L + L_{eq} \cdot f \cdot V^2}{D \cdot 2 \cdot g})asp. + (\frac{L + L_{eq} \cdot f \cdot V^2}{D \cdot 2 \cdot g})imp.] + [Z_1 - Z_2]\}}{\eta_{blower}} \qquad (4.15)$$

Where P_g (J/S): power demand by pump, q_{imp} (m^3s^{-1}): flow rate, ρ_{liquor} (Kgm^{-3}): liquid density, g (ms^{-1}): gravity, L (m): length of pipe, L*eq* (m): equivalent length of pipe with pressure drop, V ($Kgm^{-1}s^{-1}$): viscosity, f: friction factor, d (m): diameter, Z_1-Z_2 (m): height difference and η_{pump}: pump efficiency.

In summary, total energy consumption is estimated by combining these parameters, according to Eq. (4.16).

$$E_C = E_{mixing} + E_H + E_{H,\,loss} + E_B + E_{feeding} + E_{slu,cycle} + E_{E,\ per} \qquad (4.16)$$

The net energy balance, E_{Net}, is calculated according to Eq. (4.17)

$$E_{Net} = E_R - E_C \qquad (4.17)$$

Where E_R: Energy output and E_C: energy input.

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