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Abstract: Membrane bioreactor (MBR) has been deemed to be a promising technology for wastewater treatment and reclamation; however, the MBR filtration performance inevitably decreases with filtration time attributed to the deposition of soluble and particulate materials onto and into the membrane under the interactions between activated sludge components and the membrane. Cake layer formation on membrane surfaces has been a major challenge in the operation of MBRs under supra-critical flux operation, and/or caused by uneven distribution of aeration intensities, etc.; however, it was argued that a thin cake layer might improve filtration operation by some researchers. This paper provides a critical review on the formation mechanisms, properties, the role of sludge cake in membrane filtration, and the corresponding strategies of controlling cake fouling in MBRs. Drawbacks and benefits of the formation of sludge cake were also discussed in order to better understand the characteristics and role of sludge cake formation in MBRs.

Keywords: Cake layer, membrane bioreactor (MBR), membrane fouling, sludge cake characteristics

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INTRODUCTION

Membrane bioreactor (MBR) technology, which combines activated sludge process with a direct solid-liquid separation by membrane filtration, has been deemed to be a promising technology for wastewater treatment and reclamation (1). Compared to conventional activated sludge (CAS) process, MBR technology features advantages such as a small footprint, high quality effluent, a low sludge production rate, and easy manipulation of the sludge retention time (SRT), etc. (2–4). However, the MBR filtration performance inevitably decreases with filtration time. This is attributed to the deposition of soluble and particulate materials onto and into the membrane under the interactions between activated sludge components and the membrane. This major drawback and process limitation has been under investigation since the early MBRs, and remains one of the most challenging issue facing further MBR development (5,6).

It is reported that the main cause of fouling problems is sludge cake formation on the membrane surfaces in MBRs (5,7–9). The sludge cake layer attached to membrane surfaces appears to have a great filtration resistance. A sludge cake formed on the membrane with a thickness of less than 1 mm might have a filtration resistance of $1.7 \times 10^{13}\,m^{-1}$, which could cause a pressure drop of about 0.5×10^{5} Pa for a low effluent flux of $0.25\,m^3/(m^2\,d)$ or less in an MBR (10). Lee et al. (9) observed that the contribution of cake resistance, membrane intrinsic resistance, irreversible fouling resistance to the total resistance of a submerged MBR was 80%, 12%, and 8%, respectively. A consensus reached by this group of researchers is that sludge cake resistance is the major contributor to membrane fouling, and the formation of sludge cake on membrane surfaces is definitely detrimental to the filtration performance (11). Another group of researchers (9,12,13), however, argued that the formation of sludge cake on membrane surfaces was beneficial to the filtration operation of MBRs. It was verified that sludge cake layer acted as a secondary or dynamic membrane which screened the primary membrane from the more strongly fouling species, e.g., particles with smaller size (9,12). It was also reported by Giraldo and LeChevallier (13) that a thin cake layer on the membrane could improve MBR performance. In their study, they adopted a controlled aeration rate which ensured that a thin cake layer occurred but headlosses were minimized. This cake could prevent the membrane from soluble microbial product (SMP)-related fouling and protect against long-term fouling (13). It is essential to generate an overall understanding of sludge characteristics and its role in membrane filtration by summarizing and analyzing the recent literature and research progress on sludge cake fouling in MBRs.

The work presents an overview on the formation mechanisms, properties, role of sludge cake in membrane filtration, and the corresponding strategies of controlling cake fouling in MBRs. Drawbacks and benefits of the formation of sludge cake were also discussed based on the recent research on sludge cake in the literature. This critical review is expected to provide a comprehensive understanding of sludge cake fouling and to facilitate MBR design and operation.

MECHANISMS OF CAKE LAYER FORMATION

Mechanisms of Particle Transport

In cross-flow membrane filtration, particle transport depends on two main actions: one action is to move the particles toward the membrane surface (negative direction) and the other one is to shift them away from the membrane surface (positive direction) (14). The negative direction forces (vector) include gravity force ($\vec{F}_g$), van der Waals attraction forces including attractions between atoms, molecules and surfaces ($\vec{F}_A$), and permeation drag force induced by filtration ($\vec{F}_J$), while the positive direction ones (vector) include buoyancy force which is the upward force on a particle produced by the surrounding fluid due to the pressure ($\vec{F}_b$), electrical double layer repulsion force between the diffusive double layers surrounding each particle ($\vec{F}_R$), Brownian diffusion force as a result of thermal molecular motions ($\vec{F}_B$), shear-induced diffusion force caused by cross-flow velocity (CFV) ($\vec{F}_s$), and lateral inertial lift force produced by the velocity gradient near a plane wall ($\vec{F}_l$). The rate of momentum of a particle was determined by the sum of all the forces ($\vec{F}$) imposed on the particle, as schematically shown in Fig. 1, in a fluid stream along the membrane surface. The Newton's second law states that the net force exerted on a particle is the sum of all the forces mentioned above, which could be expressed as follows (14).

$$\vec{F} = \frac{\pi}{6}\rho_p d_p^3 \frac{d\vec{v}_p}{dt} = \left(\vec{F}_b - \vec{F}_g\right) + \left(\vec{F}_R - \vec{F}_A\right) + \left(\vec{F}_B + \vec{F}_s + \vec{F}_l\right) - \vec{F}_J \tag{1}$$

The particle transport equation can be transformed into the form composed of corresponding velocities as given in Table 1 (15–18) through dividing Eq. (1) by $3d_p\eta^{-1}$ (14).

$$\frac{\pi}{18}\rho_p \eta d_p^2 \frac{d\vec{\nu}_p}{dt} = (\vec{\nu}_b - \vec{\nu}_g) + (\vec{\nu}_R - \vec{\nu}_A) + (\vec{\nu}_B + \vec{\nu}_s + \vec{\nu}_l) - \vec{J} \tag{2}$$

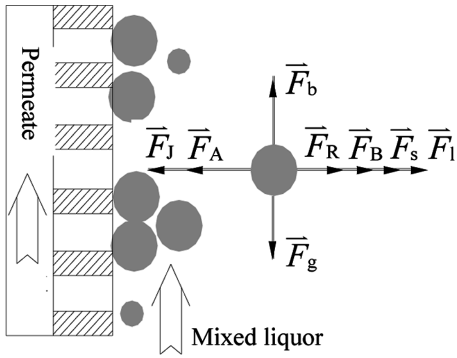


Figure 1. Schematic diagram of the forces exerted on the particle.

Assuming that the density of the biosolids suspended in a flowing stream is the same as that of the liquid, the above equation at a certain state ($\frac{d\vec{\nu}_p}{dt} = 0$) can be simplified to Eq. (3) (14).

$$\vec{J}_c = (\vec{\nu}_R - \vec{\nu}_A) + (\vec{\nu}_B + \vec{\nu}_s + \vec{\nu}_l) \tag{3}$$

where $\vec{J}_c$ could be considered as the critical flux below which particle at diameter d_p would not deposit onto the membrane surface.

Table 1. Factors affecting particle transport in cross-flow membrane filtration (15–18)

Factor	Expression[a]
Toward the membrane	
$\overline{F}_g$	$\overline{\nu}_g = \pi\eta d_p^2 \rho_p g/18$
$\overline{F}_A$	$\overline{\nu}_A = A\pi\eta s^2/36$
$\overline{F}_j$	J
Away from the membrane	
$\overline{F}_b$	$\overline{\nu}_b = \pi\eta d_p^2 \rho_l g/18$
$\overline{F}_R$	$\overline{\nu}_R = 2k\varepsilon\zeta^2\eta\exp(-ks)/3$
$\overline{F}_B$	$\overline{\nu}_B = kT\pi\eta d_p\delta/3$
$\overline{F}_s$	$\overline{\nu}_s = 0.0225u_0 d_p^2\delta/h$
$\overline{F}_l$	$\overline{\nu}_L = 0.108\rho_p u_0^2 d_p^3 h^2/\eta$

[a] d_p, particle diameter (m); ρ_p, particle density (kg/m^3); η, dynamic viscosity (Pa s); A, Hamaker constant; s, separation distance (m); ρ_l, liquid density (kg/m^3); k, Debye-Hückel parameter; ε, fluid permittivity; δ, boundary layer thickness calculated by the Lévéque equation (m); ζ, zeta potential (mV); u_0, average fluid velocity (m/s); h, half-channel height (m); T, Kelvin temperature (K).

According to expressions listed in Table 1, it could be known that $(\vec{\nu}_R - \vec{\nu}_A)$ is a constant vector. The vectors $\vec{\nu}_B$, $\vec{\nu}_s$, and $\vec{\nu}_l$ are all related to the particle diameter d_p, and thus the following equation could be deduced based on Eq. (3) (19).

$$d_p = f(J_c) \tag{4}$$

From Eq. (4), it could be concluded that at a certain permeation flux there is a corresponding particle size below which the kinds of particles will move toward the membrane to cause fouling and above whose attachment/deposition of those kinds of particles onto membranes could be avoided due to hydrodynamic effects (19).

Cake Layer Formation

Most researchers have attributed the initial flux decline when operating at imposed transmembrane pressure (TMP) and the steady TMP increase when operating at an imposed flux above the critical to the deposition of particles on the membrane surface and the formation of a cake layer (20,21). A detailed three-phase mechanism of sludge cake layer formation for the constant TMP operation was proposed by Bae and Tak (22). In Phase I, all sludge particles including large sludge flocs, colloids, and solutes are deposited onto the membrane surface and rapid flux decline occurs in this stage. Large sludge flocs attached to the membrane surface can be removed, to some extent, by cross-flow due to their size, while small particles like colloids and solutes are deposited onto the membrane surface and the pore walls to cause irreversible fouling. During Phase II, sludge constituents are accumulated not only on the membrane surface but also on previously deposited layers and the rate of flux decline is alleviated compared to that of Phase I. The permeation drag of this stage has significantly decreased because of the initial flux decline in Phase I, and thus the amount of particle deposition declines from the rates of Phase I. The permeation drag and the back transport of solutes reach equilibrium state in Phase II, and the colloids and the suspended solids contribute to an increase in the filtration resistance. Phase III is the stage during which the deposited cake layer is gradually pressurized and compacted. The compaction of the cake layer also increases the filtration resistance and decreases the permeability. When the permeation drag of the suspended solids decreases until it becomes equal to the back transport velocity, flux reaches steady state. It is worth pointing out that under constant flux operation mode, a sludge cake layer is formed at the imposed membrane flux above the critical one, i.e., supra-critical flux operation (23). There is no obvious staged fouling phenomenon as constant flux operation (supra-critical flux) mode and the cake layer will be gradually thickened and compacted as TMP increases during the filtration operation (23). The uneven distribution of aeration intensities on the membrane for surface scouring is another contributor to a partial sludge cake coverage, which initiates a progressive sludge cake growth on the membrane (7).

In sub-critical flux operation, membrane fouling was reported by various authors to have a two-step fouling phenomenon, i.e., a slow increase of TMP followed by a rapid increase (24–26). In the first period, fouling gradually occurs on the porous surface and appears, after long runs, to be hydraulically irreversible (24) and always a gel-layer is formed on the membrane surfaces. In the second period, the hydraulic response of the environment is close to that observed in supra-critical conditions and a cake layer (reversible) could be formed if the filtration is not stopped. Ognier et al. (24) proposed a local flux concept to explain the fouling phenomenon and to illustrate the different layers deposited/adhered onto membrane surfaces. During the first period, the interactions of membranes and mixed liquors induce a reduction in the number of pores open to the filtrate flow. As the permeate flow is kept constant during the filtration operation, this reduction of the area open to the flow is expressed as a gradual increase in circulation rate, or local flux, in the pores remaining open. The increase slowly intensifies as the pores close, and may lead to the local flux reaching a level equal to the critical flux value. A deposition of sludge flocs on the membrane then occurs, which translates to very high hydraulic resistance marking the onset of the second filtration period. The relationship between the formation of the cake layer or the gel layer and different membrane flux operation is demonstrated in Fig. 2. It shows that if operational flux is higher than the critical flux (J_c), sludge cake is formed and there is no obvious staged fouling phenomenon; however, at operational flux less than critical flux, the two-step fouling phenomenon occurs, and the gel layer is formed in the first period while the cake layer is deposited in the second period. The gel layer formation is mainly attributed to the deposition of macromolecules, colloids, and SMP, etc. while the cake layer is always comprised of a large quantity of sludge particulates (flocs) (27). Another difference between the cake layer and the gel layer is the irreversible/reversible characteristics, i.e., the cake layer is reversible while the gel layer is more irreversible which could be removed by dramatic forward flushing or chemical cleaning.

In the process of sludge cake formation, it is reported that biopolymers play an important role (8). Nagaoka et al. (28) found that the specific resistance of extracellular polymeric substances (EPS), which was

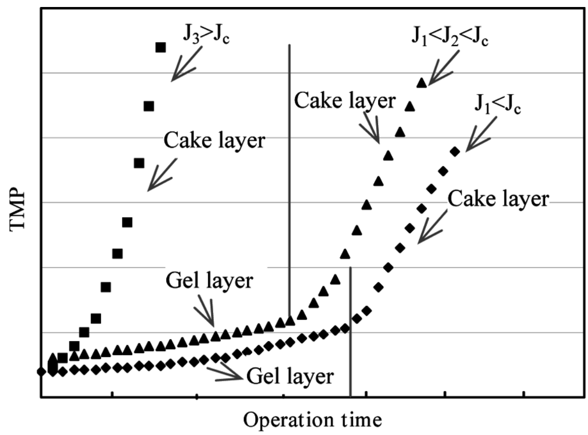


Figure 2. Relationship between fouling layer formation and membrane flux operation (J_c, critical flux).

defined as the filtration resistance (m^{-1}) divided by the EPS density on the membrane (kg-TOC/m^2), was on the order of 10^{16} to 10^{17} m/kg. The deposition of biopolymers such as polysaccharides, proteins, etc. increases the stickiness of the membrane surface, which allows easier and faster bacterial adhesion. In addition, the sludge flocs could be embedded in EPS which increases the difficulty of sludge removal by aeration turbulence (29). Some researchers even called this unique fouling layer as "biofilm" because of its special architecture (30,31). Many researchers used confocal laser scanning microscopy (CLSM) to verify the structural features and the spatial distribution of cellular and polymeric cake constituents (30,32). They found that polysaccharides and proteins were distributed in the cake layer and the spatial distribution of these biopolymers contributed to the low porosity and the unique cake architecture.

SLUDGE CAKE CHARACTERISTICS

Components of Sludge Cake

Table 2 reports results obtained in 8 studies related to the characterization of components in sludge cake of MBRs (7,29,30,32–36). It could be seen that sludge cake is comprised of not only sludge particles determined as suspended solids (SS), volatile suspended solids (VSS), biovolume, biopolymers including EPS, proteins, and carbohydrates, but also inorganic matters. Those complicated mixtures result in the unique

Table 2. Components of sludge cake formed on membrane surfaces

MBR type[a]	Membrane type[b]	Wastewater	Operational flux	Cake components[c]	Ref.
SMBR	Hollow fiber, PE, 0.4 μm	Domestic wastewater	12.5 L/(m^2 h)	EPS 20.6 mg TOC/g SS	(7)
SMBR	Hollow fiber, PE, 0.4 μm	Synthetic wastewater	6–27 L/(m^2 h)	Bound EPS 90–160 mg EPS/g SS	(29)
SMBR	Hollow fiber, PE, 0.4 μm	Synthetic dye wastewater	5 L/(m^2 h)	Polysaccharide 22–55 mg/g MLVSS	(30)
SMBR	Hollow fiber, PE, 0.4 μm	Synthetic wastewater	25 L/(m^2 h)	Biovolume 1.32–2.39 $\times 10^{-4}$ m^3/m^2	(32)
SMBR	Hollow fiber, PE, 0.1 μm	Synthetic wastewater	10 L/(m^2 h)	VSS 19.85 g/m^2; COD_c 1.98 g/m^2; COD_s 2.64 g/m^2; inorganic matter 7.34 g/m^2	(33)
SMBR	Plate frame, PE, 0.4 μm	Synthetic wastewater	10 L/(m^2 h)	TOC 222–261 mg/L; protein 325.6–402.7 mg/L; carbohydrate 64.9–109.6 g/L	(34)
SMBR	Flat-sheet, PTFE, PCTE, and PETE, 0.1 μm	Municipal wastewater	9.1 L/(m^2 h)	DOC 24.6–67.3 mg/m^2; carbohydrate 8.0–13.0 mg/m^2; protein 9.6–14.8 mg/m^2	(35)
SMBR	Hollow fiber, PE, 0.1 μm	Synthetic wastewater	Constant TMP mode 55 (initial) – 10 (end) L/(m^2 h)	SS 7.9–34.6 g/m^2; COD_c 1.11–3.19 g/m^2; COD_s 1.67–2.33 g/m^2	(36)

[a]SMBR, submerged MBR; [b]PE, Polyethylene; PTFE, Polytetrafluoroethylene; PCTE, Polycarbonate; PETE, Polyester; [c]TOC, total organic carbon; COD_c, colloidal COD; COD_s, soluble COD; DOC, dissolved organic carbon.

structure and generally high filtration resistance of sludge cake during filtration operation.

Cake Layer Morphology

A variety of instruments could be employed for characterizing the morphology of sludge cake. A very commonly used technology is the scanning electron microscopy (SEM) and various researchers reported their findings on cake morphology (8,9,33,37,38). Figure 3 illustrates the SEM photographs of membrane surface of cleaned and fouled membranes obtained by Chu and Li (9), and other researchers' findings are similar to the images. Membrane pores of the fresh membrane or cleaned membrane could obviously be seen in the SEM photographs; however, a cake layer built up on the membrane surface seems to be dense and non-porous. The adsorption of biopolymers like EPS on the membrane surface modified the cake surface property and led to easier biomass attachment and tighter sludge cake deposition which resulted in a progressive sludge cake growth and serious membrane fouling (9). Another useful tool for identifying sludge cake morphology is the atomic force microscope (AFM), which could quantify the surface roughness. Based on the root-mean square (RMS) of AFM images, the roughness of the membrane surface and sludge cake could be obtained (38). It was reported by Zhao et al. (39) that the mean square roughness of the fouled membrane was in the range of 75–132 nm and the thickness of the cake layer was ranging from 463 nm to 1200 nm in a submerged MBR. Figure 4 illustrates the AFM images of a new membrane and a fouled membrane in the study of Zhang et al. (40). Compared with the surface

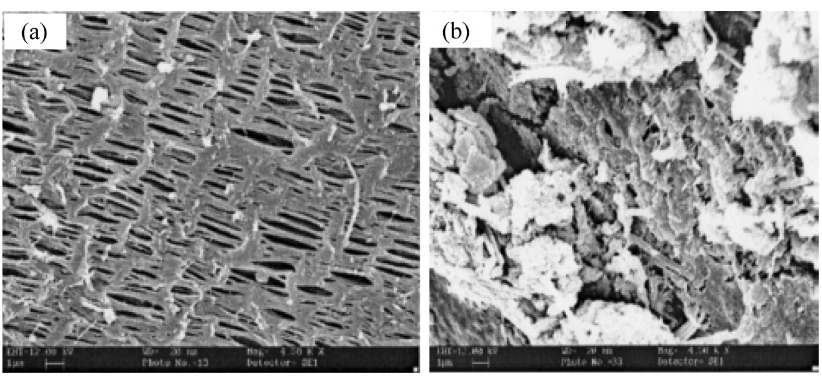


Figure 3. SEM photographs of (a) cleaned membrane and (b) membrane surface with sludge cake coverage (from reference (9)).

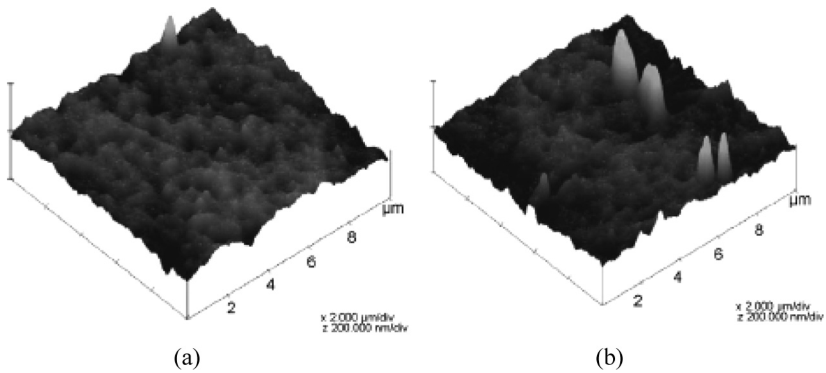


Figure 4. AFM images of fresh and fouled membrane surfaces, (a) fresh PES membrane (roughness = 12.989 nm) and (b) fouled PES membrane (roughness = 20.792 nm) (from reference (40)).

of the new membrane, the surface with sludge cake coverage on the fouled membrane shows larger roughness. A direct observation (DO) technique, which allows real-time, in situ direct observation through membrane (DOTM) of the particle formation process, was also developed and reported by Li et al. (41) and Le-Clech et al. (42). This technology employs a microscope, a camera, and a VCR to continuously record the formation phenomenon of sludge layer and the change of the membrane surface. Li et al. (41) reported that rolling of the particles was found during the filtration of 6.4 µm latex near the critical flux whereas a flowing cake layer was observed during the filtration of 3 µm latex. Le-Clech et al. (42) adopted the DO technology and revealed the creation of a well-structured dual fouling system (bentonite-concentrated layer of 50 µm embedded and covered by a concentration polarization of alginate greater than 240 µm) on the surface of a hollow fiber membrane filtrated by 500 mg/L of alginate mixed with 100 mg/L of bentonite under membrane flux 53 L/(m^2 h). Besides those technologies as mentioned above, small-angle neutron scattering (SANS) and static light scattering (SLS), etc. can also be used for characterizing cake layer surface morphology (43–45). It was found that steeper decay of scattering intensity profiles indicated a more dramatic deposition of proteins within membrane pores during cross-flow filtration of bovine serum albumin (BSA) by using SANS technology (44). In the application of SLS technique, the build-up of a layer on the membrane surface could be expressed as the absorption of light, and this variation in signal intensity corresponded directly to the cake layer thickness through calibration curve recorded at the beginning of an experiment (45).

Cake Layer Structure

Among the various techniques to evaluate cake layer structure, confocal laser scanning microscopy (CLSM) allows the non-destructive in situ examination of the inner architecture and can be effectively used for visualization and quantification when combined with the application of fluorescent probes (29,32,46–48). A series of fluorescent dyes that are specific for each component are needed to stain the sludge cake before CLSM analysis. The list of fluorescent dyes commonly used in the cake staining is summarized in Table 3. By using the fluorescent dyes and corresponding molecular probes to stain cake layer, CLSM could identify and visualize the spatial and temporal distribution of bacterial cells, polysaccharides, proteins, EPS, and the live/dead distribution of bacterial cells (31,49–52).

After CLSM analysis, IMARIS (v4.1.3, Bitplane AG, Zurich, Switzerland) could be used to reconstruct the three-dimension (3D) views of sludge cake (53). The CLSM images of sludge cake, which were reported by Hwang et al. (29,32), are shown in Fig. 5. The cake images could be used to analyze cake structure in terms of cake porosity, biovolume (e.g., total volume of microbial flocs in a given cake), and the average run length by using the software of Image Structure Analyzer ISA-2 in three-dimensions (54). The highest bacterial cell density near the substratum (i.e., lower layer) was reported by some investigators (32,55), while others have observed higher cell density in the outer region of the cake (biofilm) formed in river water at some distance from the substratum (56). In a submerged MBR system, because the top layer of the cake is always exposed to shear force induced by air scouring, bacterial cells at the upper part of the cake would have greater opportunity for sloughing and back-transport to the bulk solution than those inside the cake. Consequently, the bacterial cell density at the top region should be less than at any other parts of the cake layer. It was reported that the live-to-dead ratio changed continuously along the depth of the cake layer, i.e., the ratio at the lower layer of the cake became increasingly smaller, whereas the ratio at the upper layer became increasingly larger (29). Due to the poor transfer of oxygen and nutrients from the bulk solution, the lower layer near the membrane surface was becoming a stressful environment for the microorganisms (e.g., resulting in low live-to-dead ratio) as the bio-cake accumulated on the membrane surface.

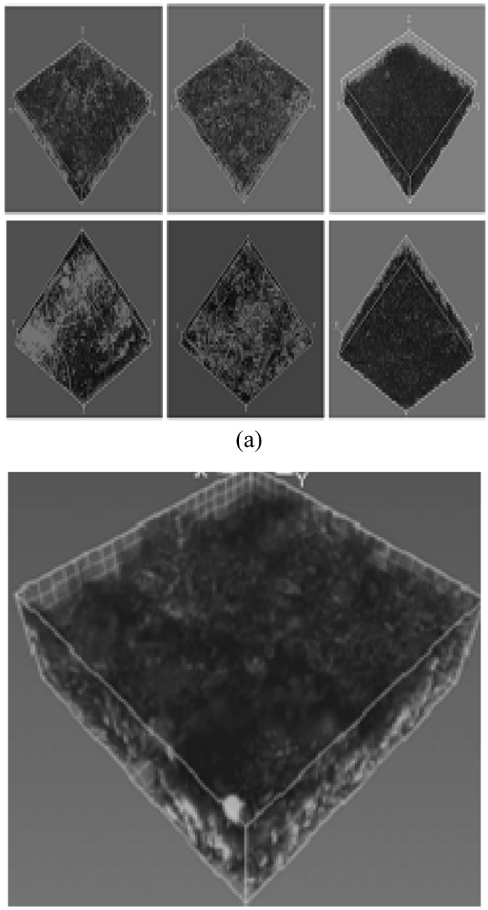
Cake Resistance

In order to analyze the resistance of sludge cake in MBRs, resistance-in-series model was commonly used for determining cake resistance (R_c),

Table 3. Fluorescent dyes used in staining the cakes and their spectral characteristics

Fluorescent dyes and probes	Label	Excitation wavelength (nm)	Emission wavelength (nm)	Specificity (target)	Channel	Reference
SYTO 9	Eugene	488	515–530	Nucleic acids (bacterial cell)	Green	(29,48)
SYBR Green I	Eugene	488–497	515–530	Nucleic acids (bacterial cell)	Green	(30–32,49)
SYTO 63	Carlsbad	633	650–700	Nucleic acids	Red	(50)
Fluorescein isothiocyanate (FITC)	Eugene	488	500–540	Amine-reactive compound like proteins and amino sugars	Green	(50)
Concanavalin A (Con A)[a]	Eugene	555–568	600–650	α-Mannose, α-glucose (polysaccharide)	Red	(29,30,321)
Con A[a]	Eugene	543	550–600	α-Mannose, α-glucose (polysaccharide)	–	(50)
Wheat germ agglutinin	Eugene	568	600–650	$(\beta\text{-GlcNAc})_2$, NeuNAc (polysaccharide)	Red	(30)
Calcofluor white	Sigma	400	410–480	β-linked D-glucopyranose polysaccharides	Blue	(50)
Benzoxanthene yellow (Hoechst 2495)	Sigma	395	450–480	Protein	Blue	(32,51)
A-9771	Sigma	494	520	BSA–fluorescein conjugate	Green	(47)
O-23021	–	596	615	Ovalbumin–Texas red conjugate	Red	(47)
BacLight Live-Dead staining kit	Eugene	–	–	Live/dead distribution of bacterial cells	–	(29,52)

[a]conjugated with tetramethyl rothamine isothiocyanate (TRITC).



(a)

(b)

Figure 5. Volumetric 3D reconstructed cake images as (a) reported by B.K. Hwang et al. (29); (b) by reference (32). (For identification of color in this figure legend, readers could refer to the web version of articles of the references listed above. Green color: bacterial cells; red color: polysaccharides.).

pore-blocking resistance (R_p), and intrinsic membrane resistance (R_m) (57). Specific cake resistance (α) is a cake resistance normalized to the mass of biosolids deposited on the unit area of cake layer, which is also can be used to characterize the cake layer (58,59). Table 4 shows various resistances and specific cake resistance in MBRs reported in the literature. Due to the impacts of sludge concentration on cake resistance, we also list the mixed liquor suspended solids (MLSS) concentration in those studies in this table.

Table 4. Analysis results of α, R_m, R_p, R_c, and total membrane (R_t) in MBRs

		Resistance ($10^{12}\,m^{-1}$)					
MLSS (g/L)	α (m/kg)	R_m	R_p	R_c	R_t	Cake resistance ratio (%) R_c/R_t	Reference
8.0–12.0	4.9E + 13	–	4.0	9.0	–	–	(7)
2.0–3.0	3–60E + 12	0.49–0.50	0.35–0.81	2.94–3.39	4.24	69.0–80.0	(9)
3.1–3.4	–	1.3–1.9	0.6–1.1	32.2–38.0	35.2–39.9	92.0–95.0	(22)
1.8–4.6	–	1.1	0.8–0.9	8.8–8.9	10.8	81.5–82.4	(30)
4.7 ± 0.25	–	0.20	0.80	15.40	16.40	93.9	(32)
6.0	–	0.48	0.40	4.66	5.54	84.1	(33)
6.0	2.3–5.2E + 13	0.11	0.25–0.42	0.59–1.38	0.98–1.74	53.0–79.4	(36)
1.0–2.0, 7.0–12.0	1–100E + 12	1.16–1.17	0.38–0.46	4.14–4.23	5.77[a]	72.0–73.0	(31)
0.09–3.7	0.7–18.5E + 8	0.89–1.56	1.16–1.51	2.06–5.39	4.49–7.44	40.2–72.4	(58)
5.2–10.0	0.6–1.2E + 12	–	–	–	–	–	(59)

[a]resistance values calculated at 30 kPa.

It could be observed from Table 4 that the resistance caused by sludge cake layer is the main contributor to the total resistance. The cake resistance (R_c) which is closely associated to the membrane permeability, is a function of specific cake resistance (α) and the weight of biomass deposited on a certain area of membrane surface as expressed by the following equation (60).

$$R_c = \frac{\alpha M}{A_m} \tag{5}$$

where α is the specific resistance of sludge cake (m/kg), M is the mass of biomass (kg), and A_m is the membrane area (m^2).

In Table 4, the sludge cake attached to the membrane surface in the literature generally has a high specific resistance of the order of 10^{12}–10^{14} m/kg except the value reported by Chang and Kim (58). Much difference in terms of filterability could be found between the cake sludge that was removed from the fouled membrane and the bulk sludge of the MBR suspension. It was reported by Wang et al. (7) that the cake sludge had an average filtration resistance of 4.9×10^{13} m/kg, whereas the bulk sludge in MBR has an average filtration resistance of only 1.9×10^{11} m/kg which was more than two orders of magnitude lower than the resistance of the sludge cake layer. They argued that a pool of biopolymer clusters (BPC) trapped within the sludge cake on the membrane surface resulted in the higher specific filtration resistance of the sludge cake layer and the interaction of the biomass flocs and the BPC built up a sludge cake with an unusually high filtration resistance. According to the Carman-Kozeny equation as given below, both the size of the particles (e.g., microbial flocs) and the porosity of the cake layer are key parameters determining the specific cake resistance of the cake (31,61).

$$\alpha = \frac{180(1 - \varepsilon_p)}{\rho_p d_p^2 \varepsilon_p^3} \tag{6}$$

where ε_p is the porosity of the cake, ρ_p is the density of particle (kg/m^3), and d_p is particle diameter (m).

Based on the Carman-Kozeny equation, it is obvious that smaller particles increase the specific cake resistance, which will correspondingly deteriorate the membrane permeability. Specific cake resistance is inversely proportional to the porosity, and higher porosity leads to lower α value. It is worth noting that the size of the particles in the sludge cake might be interrelated to the porosity. Smaller average particle size may result in lower porosity of sludge cake (31) and thus high specific cake resistance.

Modeling of Sludge Cake Fouling

For a sludge particle that approaches the membrane, two opposite forces act on the particle to affect its tendency of attachment to the membrane as mentioned in Section titled "Mechanisms of Particle Transport". If the negative direction force exerted on a particle is larger than the positive direction force, the particle tends to deposit onto the membrane surface which could result in pore blockage and/or cake layer. A series of mathematical models have been developed to characterize and simulate the sludge cake fouling, and to explain the sludge cake fouling mechanisms. Based on shear-induced hydrodynamic diffusion, Romero and Davis (62,63) developed a theoretical model of sludge cake fouling in cross-flow microfiltration process, which includes the time-dependent decline of permeate flux. They suggested that the initial flux decline due to cake buildup during cross-flow microfiltration can be approximated by modeling it as dead-end filtration. The following model, as shown in Eq. (7), could be obtained according to Darcy's law and cake resistances determined by mass balance. This model was successfully used for cake fouling simulation in a submerged membrane anaerobic bioreactor for high-strength alcoholic wastewater treatment (64).

$$J(t) = J_0\left[1 + \frac{2\alpha\phi_b\Delta P}{(\phi_c - \phi_b)\eta R_m^2}t\right]^{-0.5} \tag{7}$$

where J_0 is the initial flux (m^3/m^2), ϕ_b the solid volume fraction in the suspension, ϕ_c the solid volume fraction in the cake, ΔP the TMP (Pa), and t filtration time (s).

Silva et al. (65) modified the model developed by Romero and Davis (62,63) and proposed a semi-empirical model that could be used to predict permeate fluxes in high-shear microfiltration systems. It assumed that non-diffusive transport phenomena are the main mechanisms for the back-transport of particles from the membrane surface to the bulk solution and incorporated an equation for the transient flux based on particle mass balance at the membrane surface. This modified model was also validated in a laboratory-scale MBR using two different suspensions (65).

Ho and Zydney (66,67) proposed a combined pore blockage and cake filtration model to describe the fouling behavior. In this model, foulants first deposit on the bare membrane, reducing the area available for unhindered filtration. As the membrane surface becomes more heavily fouled, the foulant will also begin to deposit directly on the fouling layer, causing an increase in the hydraulic resistance to flow associated with the growing deposit. This is exactly what occurs in the classical cake filtration model though this cake growth is now assumed to occur simultaneously with the coverage of the remaining open area of the membrane (67). The filtrate flow rate given by the combined blockage and cake filtration model can be expressed by Eq. (8).

$$Q = Q_0 \left[\exp\left(-\frac{\alpha \Delta P C_b}{\eta R_m} t \right) + \int_0^t \frac{\alpha \Delta P C_b}{\eta (R_m + R_p)} \exp\left(-\frac{\alpha \Delta P C_b}{\eta R_m} t_p \right) dt_p \right] \quad (8)$$

where Q is the filtrate flow rate (m^3/s), Q_0 is the initial filtrate flow rate through clean membrane (m^3/s), C_b is the bulk foulant concentration (kg/m^3), t is the filtration duration (s), and t_p is the time at which the deposit begins to grow (s).

A much simpler analytical solution can be developed for the filtrate flow rate by assuming a uniform resistance of the layer over the fouled membrane surface. Under these conditions, the coefficient multiplying the exponential in the convolution integral in Eq. (8) is a constant and can be pulled outside of the integral to give Eq. (9) (67).

$$Q = Q_0 \left[\exp\left(-\frac{\alpha \Delta P C_b}{\eta R_m} t \right) + \frac{R_m}{R_m + R_p} \left(1 - \exp\left(-\frac{\alpha \Delta P C_b}{\eta R_m} t \right) \right) \right] \quad (9)$$

The combined pore blockage-cake filtration model was applied to analyze flux decline data and to evaluate the underlying fouling mechanisms for the filtration of natural organic matter (NOM) through a series of surface modified polyethersulfone (PES) membranes by Kilduff et al. (68). Ye et al. (69) simulated flux data during dead-end filtration of sodium alginate using both classical fouling models and the combined pore blockage and cake filtration model, and found that the cake filtration model (as shown in Eq. (7)) could effectively describe the flux decline obtained by ultrafilatration membranes while the combined pore blockage and cake filtration model was applicable for analyzing the experimental results obtained with 0.2 μm track-etched and 0.22 μm poly(vinylidene fluoride) (PVDF) membranes.

BENEFITS AND DRAWBACKS OF CAKE FOULING

Benefits of Cake Layer

The formation of sludge cake layer on the membrane surface could serve as a secondary dynamic membrane to filter SMP and colloids (12,13,70). The conceptual illustration of the dynamic membrane is shown in Fig. 6.

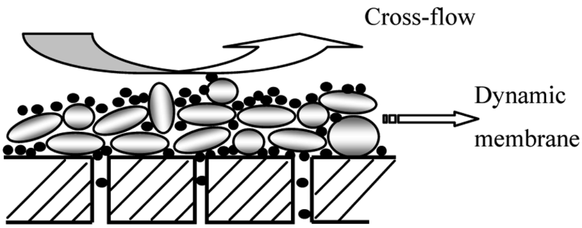


Figure 6. Schematic of the secondary dynamic membrane formed on primary membrane surfaces.

The dynamic membrane could prevent small particles, to some extent, from adsorbing onto the surface or inside the membrane pores (13).

It is well known that a thick sludge cake layer would cause severe fouling resistance which is detrimental to MBR operation (7–9). Operators commonly relax the membrane flux periodically to limit cake accumulation on the membrane surface. Cake accumulates on the membrane surface during a filtration cycle, and aeration scours this cake off when the permeate flux is stopped to relax the membrane. If the aeration intensity is not enough to remove all the accumulated cake, the cake will thicken, eventually creating more headlosses than the system could handle. If the aeration does completely scour off the cake, it will increase internal pore fouling because there is not enough cake to filter SMP and protect the membrane. Ideally, MBR operators need a strategy that uses the cake's filtration effect to minimize the internal membrane fouling while allowing them to control headlosses during relatively high permeate fluxes. Giraldo and LeChevallier (13) adopted an aeration control strategy by matching the aeration rate to the permeate flux rate which not only helped control the cake layer but also minimized the related headlosses. This measure allowed that a thin cake with relatively low filtration resistance could be formed on the membrane surface. The strategy which takes advantage of the thin cake layer, facilitates the long-term and stable operation of MBRs.

Drawbacks of Cake Layer Formation

As discussed in Section titled "Cake Resistance", sludge cake has a high specific resistance, and thus the formation of the cake layer in particular the thick layer on the membrane surface could result in high filtration resistance. That could decrease the effective filtration area and finally enable the coverage of sludge cake on the total membrane surface. It generally leads to a high TMP during the filtration performance, rapid membrane cleaning and replacement, and to high operation and maintenance costs. Due to the high membrane resistance caused by sludge cake, much research (5,19,23,35,71–73) has dedicated to the prevention and control of sludge cake fouling during filtration operation which will be discussed in detail in the following section.

STRATEGIES FOR CONTROLLING SLUDGE CAKE FOULING

Operational conditions play a key role for mitigation of fouling in the MBR process. It has been well recognized that critical flux is an important concept in MBRs, below which the increase of TMP or the decline of flux with time does not occur and above that level the fouling is observed (71,73). Based on critical flux concept, sub-critical flux operation was proposed and proven suitable for MBR operation (3). Under sub-critical flux operation, sludge cake fouling could be, to a great extent, controlled during filtration in MBRs. Instead of sludge cake fouling, a gel layer attached to the membrane surface during the first fouling period was reported by lots of researchers when they operated MBRs under the sub-critical flux mode (27,74–77). This operation mode could enable MBR to achieve a long-term, stable operation without frequent membrane cleaning. There are many other measures which could mitigate sludge cake fouling, such as the improvement of hydrodynamic conditions of the reactor, the optimization of aeration intensity, the implementation of suction and non-suction protocol (intermittent filtration), the selection of proper sludge retention time (SRT), and hydraulic retention time (HRT), etc (3,5,78). The addition of membrane fouling reducer (MFR) including polymer or inorganic materials (coagulant/flocculant), suspended carrier, and so on, to MBRs is also conducive to controlling of sludge cake fouling by the decrease of cake thickness, the increase of cake porosity, and the change of cake architecture (3,32,79).

Although the measures mentioned above can control sludge cake fouling to some extent, the decrease of membrane permeability is inevitable. Once TMP increases dramatically to a certain value, the membrane cleaning procedure is needed to recover the membrane permeability. Back-flushing is an effective way for high-pressure-resistance membranes (80), such as hollow fiber and ceramic membranes, to remove the sludge cake layer and biofouling substances adhered onto the membrane surface or clogged in the membrane pores. In general, back-flushing consists of reversing the filtration direction for 5–30 s every 30–60 min. Other mechanical cleaning methods were also developed by Xu and Fan (81) by using a hollow fiber membrane module with enhanced self-mechanical-cleaning function which was suitable for high sludge concentration and flux operation. Sun et al. (82) adopted sponge scouring to remove the sludge cake layer in a submerged flat-sheet MBR. Chemical cleaning has been extensively studied and extensively used for removing membrane fouling and recovering membrane permeability. A variety of chemical agents were applied in membrane cleaning, such as acids (hydrochloric acid, sulfuric acid, critic acid, etc.), alkali (sodium hydroxide), oxidants (sodium hypochlorite, perhydrol, etc.), and their effects on foulant removal were identified (83–88). A general conclusion that could be drawn from those studies is that acids can effectively remove inorganic foulants while alkaline solution and oxidants perform well in removing organic substances and biofouling. Much higher cleaning efficiency could be achieved by employing multi-step chemical cleaning, for instance, sodium hypochlorite cleaning followed by acid cleaning and/or alkaline cleaning.

CONCLUDING REMARKS

Sludge cake formation plays an important role in the MBR filtration operation. The formation mechanisms were reported in recent literature based on particle transport in cross-flow filtration. In order to understand the sludge cake characteristics, intensive efforts were dedicated to the identification of the components in cake layer, the analysis of cake morphology, the evaluation of cake layer structure, the measurement of cake resistance and specific cake resistance, and the modeling of sludge cake fouling behavior. Many researchers reported that sludge cake resistance was the major contributor to membrane fouling, and the formation of sludge cake on the membrane surface was detrimental to the filtration performance; however, another group of researchers argued that the formation of sludge cake on membrane surfaces was beneficial to the filtration operation of MBRs. The sludge cake could act as a secondary dynamic membrane, and even a thin cake layer controlled by aeration strategy was found to be helpful for improving MBR performance. Several measures and protocols could mitigate sludge cake fouling. The adoption of a sub-critical operation could lengthen the operational period of MBR, and the optimization of other operational parameters is also a positive method. Besides those measures, physical/chemical cleaning for the recovery of membrane permeability is also needed when the TMP reaches a certain value.

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