



# **MEMBRANE BIOREACTORs**

Week 6<sup>th</sup>: Membrane Fouling

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# CEV4362 MEMBRANE BIOREACTORS 2018-2019 Spring Semester

Time and Room: Wednesday 11:00 - 11:50 FZ-82 12:00 - 12:50 FZ-82

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# Week 6<sup>th</sup>: Membrane Fouling

- 4.4. Factors Affecting Membrane Fouling
- Membrane and Module
- Microbial Characteristics
- > Operation



#### 4.4. Factors Affecting Membran Fouling

The nature and extent of fouling are strongly influenced by three factors (Figure 4.17);

- > the characteristics of mixed liquor in membrane tank
- the membrane and module type
- the operating conditions

Individual fouling factors affect membrane fouling separately (HRT, SRT etc.) and/or mutually (EPS production, etc.).



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## 4.4. Factors Affecting Membrane Fouling (2/35)

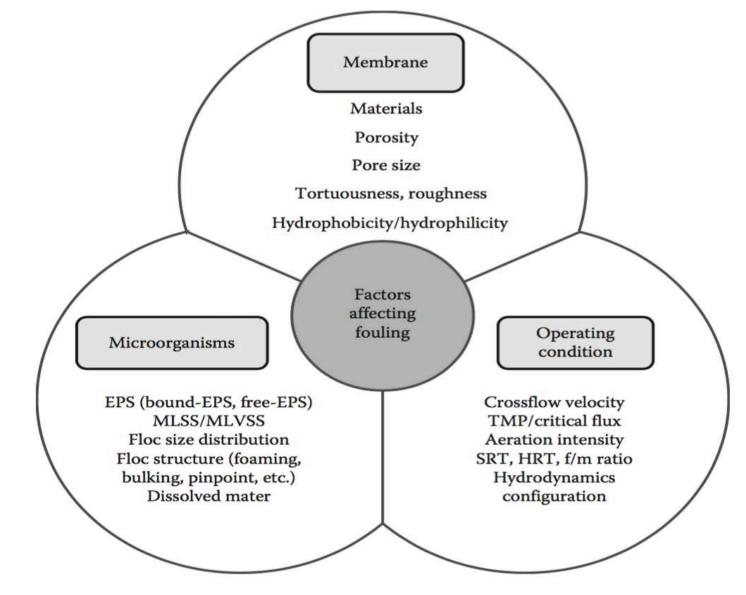


Figure 4.17. Factors affecting membrane fouling in MBRs



#### 4.4.1. Membrane and Module

Membrane characteristics that affect fouling in MBRs are;

- ✤ pore size
- porosity,
- surface energy
- charge,
- roughness,
- ✤ raw materials
- hydrophilicity/hydrophobicity



#### 4.4.1.1. Pore Size

The effect of pore size on fouling intercorrelates with the feed solution characteristics, particularly the particle size distribution of the activated sludge suspension. If the average pore size is similar to the size of the particles, pore plugging (or clogging) is likely to happen. So, larger pore sizes do not always lead to greater flux rates due to internal fouling.

The typical lower size range of activated sludge suspension particles is submicrometer (i.e., nearly close to the pore size of conventional micro ultrafiltration membranes). Therefore, ultrafiltration membranes that have smaller pore sizes than microfiltration membranes are often used in MBRs.



#### 4.4.1.2. Hydrophilicity/Hydrophobicity

Hydrophobic membranes interact more strongly with the feed solution's components than hydrophilic ones do (hydrophobic interaction). So, fouling is more likely to occur in hydrophobic membranes.

The most frequently used membrane materials in MBRs (polyethylene, polypropylene, and polyvinyledendifluoride) have a hydrophobic nature because they do not have polar groups in their molecular structures. Therefore, hydrophobic parts in the feed solution preferentially adsorb to the hydrophobic membrane surface.

Membrane hydrophobicity is quantified by measuring the contact angle between a water droplet and the membrane surface. On the other hand, the hydrophobicity of the floc particles in the activated sludge suspension is quantified by measuring the "relative hydrophobicity,"



#### 4.4.1.3. Membrane Raw Materials

Polymeric materials have inherent limitations to cope with extreme conditions. Particularly, polymeric membranes are very vulnerable to wide ranges of pH values and oxidizing agents when chemical cleaning is carried out like Cleaning In Place (CIP).

**Inorganic membranes** (such as ceramic, alumina  $(Al_2O_3)$ , zirconia  $(ZrO_2)$ , silicon carbide (SiC), and titanium oxide (TiO<sub>2</sub>) have superior hydraulic, thermal, and chemical resistance compared to polymeric materials have received attention lately. But, the application of inorganic membranes to MBRs has been limited due to their cost and module manipulation limitations. Most inorganic membrane modules have the geometry of tubular monoliths, resulting in much lower packing densities than hollow fiber bundles with the same volume. If this difficulty is overcome, applications of inorganic membranes to MBR would be widespread because simple and powerful cleaning options using chemicals under extreme conditions.



#### 4.4.1.4. Charge

Membrane charge is considered an important parameter in determining the permeability of charged ions in nanofiltration or reverse osmosis processes because the rejection mechanism is strongly correlated with the static charge interaction between the membrane and transported solutes. Even though the flocs in MBRs are slightly negatively charged particles, the charge interaction between the membrane and flocs is not good enough to overcome the pressurized convection to the membrane.

#### 4.4.1.5. Module

Packing density is an important design parameter of hollow fiber membrane modules in MBRs. It is defined by the membrane surface area per unit cross-sectional area of the module header (m<sup>2</sup>/m<sup>2</sup>) or by the membrane surface area per module volume (m<sup>2</sup>/m<sup>3</sup>). High packing densities reduce the number of membrane modules and/or the footprint of the module in the aeration tank of the MBR.



#### 4.4.1.5. Module (Cont.)

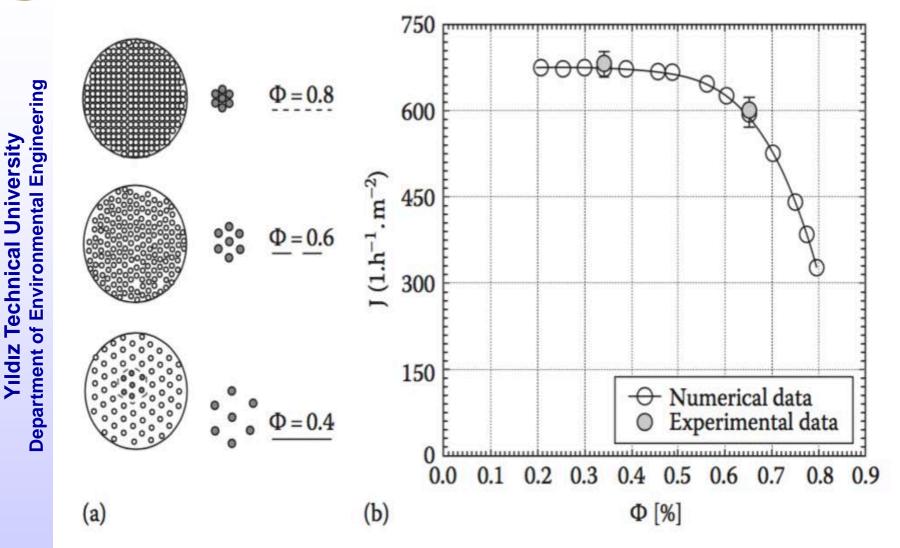
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However, overpacked modules can badly influence the mass transfer efficiency within the fiber bundles, resulting in a decreased design flux. Proper packing density design is important to keep the flux high and prevent clogging within the module in MBRs.

Recent developments in computational fluid dynamics (CFD) provide more insight to this packing density. Figure 4.18 is one example showing the effect of packing density on the water flux although the experiments were not for MBR. In this gure, a packing density between 0.5 and 0.6 would provide a good compromise.

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## 4.4. Factors Affecting Membrane Fouling (9/35)



**Figure 4.18.** (a) Conceptual diagram of packing density and (b) the effect of packing density on flux.



## 4.4. Factors Affecting Membrane Fouling (10/35)

#### 4.4.2. Microbial Characteristics

Mixed liquor of activated sludge is a complex and variable heterogeneous suspension containing;

- $\checkmark\,$  unmetabolized feed components
- ✓ metabolites produced during biological reactions
- ✓ biomass

Many individual components of the mixed liquor (such as biomass solids, dissolved polymers (EPSs) etc.) can contribute to membrane fouling.

Each microbial factor affecting membrane fouling is strongly influenced by operating conditions (if operating conditions change, microbial characteristics such as MLSS or EPS concentrations change).



#### 4.4.2.1. MLSS

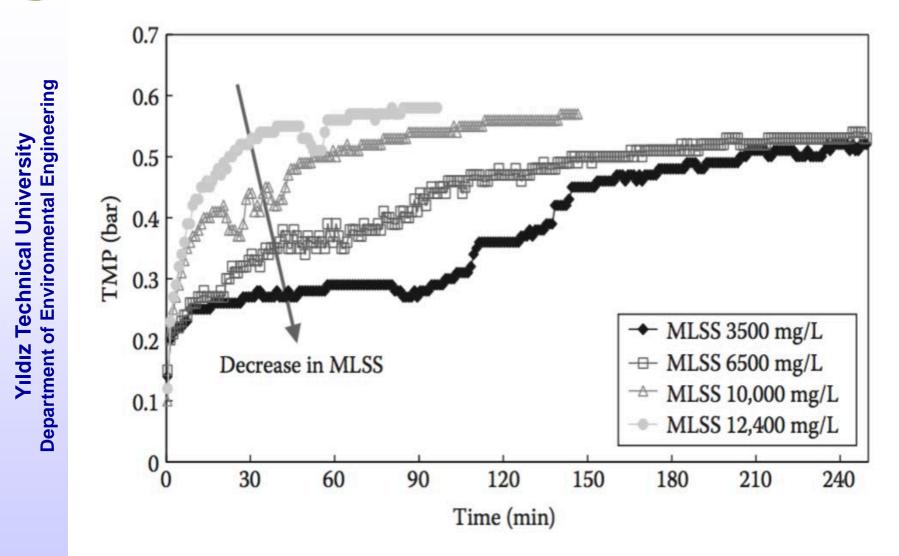
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MLSS (biomass) concentration in MBR tanks is directly proportional to membrane fouling if other important microbiological factors are kept constant. This is because the cake layer is thicker (or denser) when the biomass concentration becomes greater. However, this hypothesis is only true under very limited conditions as the MLSS concentration could be responsible for membrane fouling.

Figure 4.19 shows high normalized flux  $(J/J_{iw})$  values for batch-type filtrations as the MLSS concentration decreases. As shown in Figure 4.20, the TMP buildup for a submerged continuous-type bioreactor increases rapidly as the MLSS concentration decreases.



### 4.4. Factors Affecting Membrane Fouling (13/35)



**Figure 4.20.** Effect of MLSS concentration on the TMP in a continuous filtration system as a function of time.



#### 4.4.2.2. Floc Size

Among the various factors affecting membrane fouling in MBRs, the most dominant one is presumably floc size. In general, particle sizes of activated sludge flocs range from submicron to several hundred micrometers. However, the shearing force arising from either pumping in sidestream MBR or coarse aeration in submerged MBRs results in floc breakups, generating fine colloids and cells that then form a denser cake layer.

Two important factors determine the cake layer resistance;

- ➤ the particle size
- the porosity of the cake layer

and their mutual influence should be noted. The general consensus is that porosity decreases if the particle size increases. However, porosity remains constant as the particle size increases or decreases; otherwise, cake layer volume is equal. The size of the particles does not make any changes in the porosity as shown in Figure 4.22.



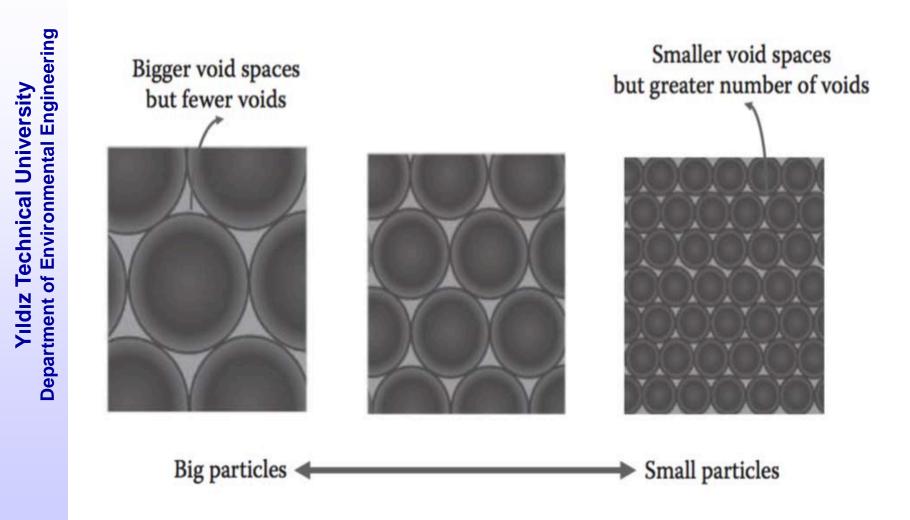


Figure 4.22. Relationship between the particle size and porosity.



#### 4.4.2.3. Compressibility of the Cake Layer

In conventional filtration using filter media such as sand or anthracite rather than membranes, specific cake resistance has been used to express the filterability of media.

If high pressure is applied for membrane filtration, the cake layer on the membrane surface will be compressed, and pore plugging happen.



## 4.4. Factors Affecting Membrane Fouling (18/35)

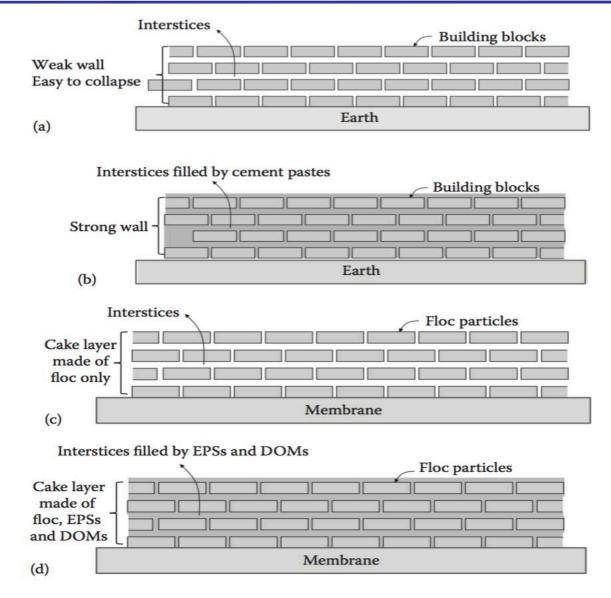
#### 4.4.2.4. Dissolved Matter

DOM present in the aeration basin of MBR plants include unmetabolized feed components and metabolites produced during biological reactions (such as SMPs and free EPSs). In terms of membrane fouling, they cannot be distinguished from each other based on the chemical structure.

DOM impact both internal and external fouling. They can be adsorbed to the pores' surfaces and walls, and cause inner membrane fouling. This happens mostly at the initial stage of filtration.

As shown in Figure 4.24, the sludge flocs are the main building blocks of the cake layer on the membrane surface. Soluble matter including DOM can fill the interstices of the building blocks present inside the cake layer, resulting in the formation of dense cake layers. DOM act as glue, just like the role of cement in the walls made of blocks, consolidating the cake layer.

## 4.4. Factors Affecting Membrane Fouling (19/35)



**Figure 4.24.** Conceptual illustration of the analogy of blocks–wall construction (a, b) and cake layer formation (c, d).



#### 4.4.2.5. Floc Structure (Foaming, Pinpoint Floc, and Bulking)

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The structure of activated sludge flocs depends mostly on the physicochemical characteristics of biomass, nutrient balance, and feed characteristics. The floc structure of activated sludge is classified into three types according to the balance of floc-forming and filamentous bacteria (Figure 4.25);

- ✓ Bulking flocs
- ✓ Pinpoint flocs
- ✓ Ideal normal flocs

A proliferation of filamentous microorganisms leads to bulking sludge, while the filamentous bacteria are not observed in pinpoint sludge flocs. On the other hand, normal activated sludge shows a good balance of filamentous and floc-forming bacteria. Activated sludge with the three previously mentioned kinds of floc structures can be obtained by controlling the HRT, SRT, and F/M ratio.



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(a)



(c)

Figure 4.25. Photos of three structure types of sludge flocs: (a) normal sludge, (b) pinpoint flocs, and (c) bulking sludge.

(b)



#### 4.4.2.5. Floc Structure (Foaming, Pinpoint Floc, and Bulking) (Cont.)

Typical SVI values for pinpoint floc, normal sludge, and bulking sludge range from <50, 100–180, and >200 mL/g, respectively. According to the studies of Chang et al. (1999), the order of fouling tendency was found to be normal sludge < pinpoint sludge < bulking sludge. They explained that the key factors controlling cake resistance were the shape, the size of the activated sludge flocs, and the porosity of the cake layer accumulating on the membrane surface. However, Wu and Huang (2009) reported that the zeta potential and SVI have no effect on membrane filterability.

Since it is almost impossible to cultivate activated sludge suspensions exhibiting different SVI values but with all other parameters affecting membrane fouling kept constant such as MLSS, F/M ratio, and HRT, the impact of SVI on fouling cannot be explained by itself, making it difficult to study membrane fouling in MBRs.



#### 4.4.2.6. Influent Characteristics

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The composition of influents to an MBR directly influences microbial metabolism. There are no major differences in sewage compositions worldwide; however, the composition of industrial wastewater differs from site to site. Domestic wastewater has a typical composition that does not vary by location. Typical nutrient ratios for the growth of microorganisms are known to be 1:5:100 for P/N/C, which represents the phosphorus, nitrogen, and carbon, respectively. More practically, a simple C/N ratio is also recommended to evaluate the balance of influent composition. Obviously, unbalanced nutrient conditions negatively affect microbial activity, leading to partial failures in WWTPs.



## 4.4. Factors Affecting Membrane Fouling (24/35)

#### 4.4.2.7. Sludge Hydrophobicity

The main components of EPSs (proteins and polysaccharides) are exposed on the surface of sludge flocs. The more hydrophobic the flocs are, the more prone they are to be adsorbed to the membrane surface. Foaming sludge is strongly hydrophobic. Foaming sludge is known to have troubles in settling as well as scum formation in secondary sedimentation tanks, leading to poor filterability.

Although the hydrophobicity of membranes can be determined by measuring the contact angle of the water droplet on a membrane surface, it is not easy to measure the sludge flocs' hydrophobicity directly. Instead, relative hydrophobicity tests using an organic solvent are recommended for measuring sludge hydrophobicity. The principle of the determination of relative hydrophobicity is based on solvent extraction from aqueous solution using organic solvents, that is, when an aqueous solution is intimately mixed with an immiscible organic solvent, the solutes (flocs in this case) in the aqueous phase distribute themselves according to its solubility in the two solvents. Figure 4.26 shows the basic procedure for 27 determining the relative hydrophobicity of a sludge sample.



#### 4.4.3. Operation

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As indicated several times in the previous section, many operating conditions directly affect microbiological characteristics such as MLSS, EPS (or SMP) productions, and floc structures. The most dominant parameters determining membrane fouling in MBRs are microbial characteristics; however, microbial characteristics are strongly influenced by operating conditions.

#### 4.4.3.1. HRT

HRT is determined by the reactor volume (m<sup>3</sup>) divided by the influent flow rate (m<sup>3</sup>/h). This parameter used for determination of reactor performances of both continuous stirred tank reactors (CSTRs) and plug flow reactors (PFRs). Generally speaking, the biodegradation of organics in the influent becomes more stable as the HRT increases.



#### 4.4.3.1. HRT (Cont.)

HRT is directly related to the F/M ratio, which is defined as the ratio of food per microorganisms:

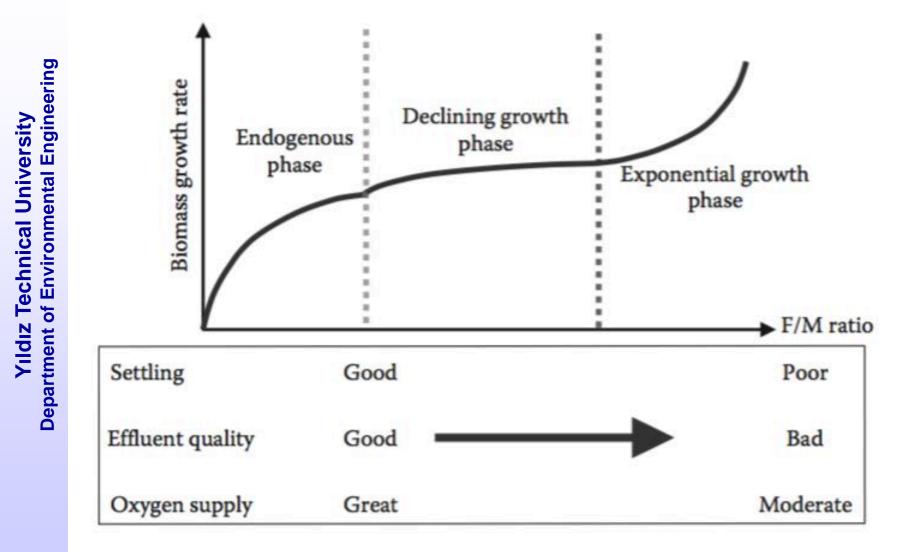
F F/M(kg BOD/kg MLSS · d) = 
$$\frac{QS_o}{VX} = \frac{S_o}{\theta X}$$

where Q is the flow rate, m<sup>3</sup>/d S<sub>o</sub> is the influent substrate concentration, kg BOD/m<sup>3</sup> V is the bioreactor volume, m<sup>3</sup> θ is the HRT, day X is the biomass concentration, kg MLSS/m<sup>3</sup>

The F/M ratio decreases as the HRT increases according to the earlier equation, which results in direct changes in microbial characteristics because the biomass growth rate strongly depends on the F/M ratio, as shown in Figure 4.27.



## 4.4. Factors Affecting Membrane Fouling (28/35)



**Figure 4.27.** Relationship of the F/M ratio and microbial characteristics and performances.



#### 4.4.3.1. HRT (Cont.)

HRT is determined by the reactor volume (m<sup>3</sup>) divided by the influent flow rate (m<sup>3</sup>/h). This parameter used for determines reactor performances of both continuous stirred tank reactors (CSTRs) and plug flow reactors (PFRs). Generally speaking, the biodegradation of organics in the influent becomes more stable as the HRT increases.

The effluent water quality such as biochemical oxygen demands (BOD) and SS deteriorates and the settling ability becomes poor as the F/M ratio increases. On the other hand, oxygen consumption becomes increased at low F/M ratios because the microorganisms in the endogenous phase require huge amounts of oxygen for the auto-oxidation of biomass.) Membrane fouling of microorganisms in the endogenous phase. Therefore, HRT affects membrane fouling indirectly via the change in microbial characteristics.



#### 4.4.3.2. SRT

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SRT is directly related to the MLSS concentration in a bioreactor. Extended SRT operation leads to an increase in cell residence times, resulting in increased MLSS concentrations (i.e., MLSS tends to increase in accordance with the SRT). So, SRT affects membrane fouling indirectly via changing the microbial characteristics.

Typical SRT values for conventional activated sludge systems are around 10 days, but common MBR plants have SRTs that are over 30 days. This prolonged SRT obviously leads to a large MLSS concentration of over 10,000 mg/L, which in turn lowers F/M ratio, which makes the microorganisms in the bioreactor endogenous. Therefore, membrane fouling becomes less severe under this extended SRT situation.

High SRT leads to low membrane fouling and better activated sludge bioflocculation. <sup>33</sup>



#### 4.4.3.2. SRT (Cont.)

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The SRT goes to infinity because most of the substrates are consumed for the maintenance needs of microorganisms. However, the MLSS concentration increases as the SRT increases, leading to an increase in the viscosity of the mixed liquor. This situation deteriorates membrane fouling; moreover, it requires excessive aeration. Therefore, optimum SRT selection is needed to control membrane fouling properly.



#### 4.4.3.3. Shear Stress

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In a sidestream MBR, a recirculation loop from the bioreactor to the membrane module is formed by a pressurized pump (or recirculation pump) in order to give shearing forces to the membrane surface to scour the cake layer. The flocs and microorganisms in the loop as well as in the bioreactor are exposed to steady shear stresses arising from the recirculation flow. The shear forces directly influence the microbial characteristics (such as altering the microbial morphology and size) releasing intracellular or extracellular compounds, and affecting microbial viability. Obviously, these microbial changes afect membrane filterability.

The most dramatic effect of shear stress is floc breakage. The fragile microbial flocs are easily broken into smaller flocs producing colloidal and ne particulates. Due to shear stresses, floc structures can collapse and the size of the flocs will therefore decrease (it affect filtration performance negatively), particularly during the  $_{35}$  initial stages of pump operation .



#### 4.4.3.3. Shear Stress (Cont.)

Another impact of floc breakup is the release of EPSs from the flocs to the bulk solution. Lots of EPSs interconnecting the microbial cells can be damaged by floc breakage and can be released to the bulk solution (membrane fouling increase).

Kim et al. (2001) also found that the shear stresses imposed on microbial flocs by a rotary-type pump would be certainly more severe than those by a centrifugal pump. So, selection of pumping device is also important to control membrane fouling in MBRs.



#### 4.4.3.4. Aeration

In submerged MBRs, coarse aeration is widely practiced aiming at controlling membrane fouling as well as supplying air to the microorganisms in the aeration basin. The coarse air supply can effciently remove or at least reduce the cake layer on membrane <u>surfaces.</u> To provide shearing forces (or scouring) to a membrane surface, coarse aeration should be extensive and excessive. But, it has unbenefical impacts on microorganisms (such as deflocculation, and floc size reduction) due to high air flow rates. The success of submerged MBR plants depends on how the fouling control strategy is implemented. If coarse aeration is too extensive, the operating costs will increase and de flocculation would be anticipated. On the other hand, if coarse aeration is not sufficient, membrane fouling will also become severe. Therefore, optimization of aeration intensity allowing sufficient shear force to reduce the cake layer is needed.

The aeration intensity (m/h), refers to the ratio of the supplied air flow rate (m<sup>3</sup>/h) to the membrane area (m<sup>2</sup>).  $^{37}$ 



#### 4.4.3.5. Flux (Critical Flux)

If the initial operating flux in an MBR starts as low as possible, the rate of fouling would be retarded. There have been many controversies about what the exact meaning of critical flux is. However, the concept of critical flux in MBRs is simple—the highest initial flux for which TMP remains stable during operation of the MBR.

Several methods for determining the critical flux have been suggested but no single protocol has been accepted. A common practice to determine the critical flux is the flux step method—to incrementally increase the flux for a fixed duration as long as each flux step leads to a stable TMP. Critical flux is determined by the flux when the TMP increases with time. The TMP increase indicates a greater resistance to permeation provided by a growing cake formation and internal fouling. TMP is dependent on fouling parameters described previously such as MLSS, membrane materials, and system hydrodynamics.



Thank you...