



Submerged membrane bioreactor for synthetic tomato processing wastewater treatment: membrane foulant and colloidal particles characterization

Tong Shu^a, Kuizu Su^{a,*}, Zhidong Wang^a, Weihong Wang^b

^aDepartment of Civil Engineering, Hefei University of Technology, Hefei 230009, China, Tel. +86 551 62905853; email: sukz@hfut.edu.cn (K. Su), Tel. +86 18225608327; email: Stong1328@163.com (T. Shu), Tel. +86 15156091673; email: 916299528@qq.com (Z. Wang)

^bCollege of Hydraulic and Civil Engineering, Xinjiang Agricultural University, Urumqi 830091, China, Tel. +86 13899910629; email: 2209319288@qq.com

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ABSTRACT

During the last several decades, membrane bioreactor technology has become increasingly popular for various industry treatments. In this study, a lab-scale submerged membrane bioreactor (SMBR) was operated to treat synthetic tomato processing wastewater under sub-critical flux. The membrane performance and the characteristics of membrane foulant were investigated. The SMBR demonstrated a high potential to handle the organic substances, ammonia and colloidal particles. The membrane foulant contained not only the extracellular polymeric substances but also some other organic matter and inorganic elements. The cake layer was identified as the main factor resulting in membrane fouling. The colloidal particles and solutes occupied a larger proportion in the membrane foulants compared with those of sludge suspension. Moreover, particle size in the washed liquid was considerably smaller than the sludge suspension in the SMBR. The gelation of colloidal and some organic foulants enhanced the formation of a gel layer, which caused fouled membrane that could not be totally recovered.

Keywords: Colloidal particles; Gel layer; Membrane bioreactor; Synthetic tomato processing wastewater

1. Introduction

Currently, the industry, especially the food industry is required to lower water consumption and improve the efficiency of water treatment [1]. According to surveys, for each ton of tomatoes produced, 15–50 m³ of wastewater is discharged. Tomato processing wastewater is generally characterized by a dark color, an offensive smell, and the inclusion of large numbers of organics, colloidal fractions and suspended solids (SS) [2]. Therefore, it exhibits a poor settling ability and biodegrades slowly, which causes significant issues in wastewater collection [3]. To date, the most common treatments of food processing wastewater are biological degradation or physicochemical process, which employs

precipitation and air flotation processes [4,5]. Though these methods remove a portion of chromaticity (color) and chemical oxygen demand (COD), they generally produce high chemical cost and large excess sludge. Hence, efficiencies are needed to meet the increasing requirement of water quality and reducing wastewater cost.

Submerged membrane bioreactor (SMBR) is a biological wastewater treatment process which combines physical separation and biological treatment by membrane filtration in one step only [6]. SMBRs offer several advantages over the conventional activated sludge process, including low sludge production, a higher biomass concentration, reduced footprint and better permeate quality [7–9]. However, the major obstruction of this process is membrane fouling, which causes either an increase in operation costs or a decrease in permeate flux [10,11].

* Corresponding author.

To mitigate membrane fouling, many researches have focused on the new design of MBR type [12], operating parameters optimization (such as operation flux, solid retention time (SRT), hydraulic retention time (HRT), loading rate and so on) [13] and addition of adsorbents and surfactant [14]. Field et al. [15] introduced the concept of critical flux. They defined critical flux as the point below which a decline of flux with time does not occur. Afterwards, some have put forward that an increase of transmembrane pressure (TMP) occurred even under critical flux. Therefore, it has led to the introduction of sub-critical flux, in which the fouling rate can be accepted during the operation of SMBR. It is widely accepted that sub-critical fouling is mainly caused by the released of extracellular polymeric substances (EPS), soluble microbial products (SMP) and other organic macro-molecules. Previous works [16,17] have set out to clarify the effect of different fractions to membrane fouling under sub-critical flux, which included SS, colloids and solutes. However, in many studies, the contribution of colloidal particles to membrane fouling remains a serious problem. Bouhabila et al. [18] estimated the contribution of major components to membrane fouling and reported that colloid particles account for 50% of total measured fouling. Sim et al. [19] demonstrated that colloidal fouling is a persistent problem due to the inherent size range of colloids. Until now, we have been confused about the behavior of colloid particles on membrane fouling and in need of a comprehensive study on properties of the membrane foulant during the operation of tomato wastewater treatment.

The objective of this study is to estimate the capacity of a membrane bioreactor for treatment of synthetic tomato wastewater. Further research was implemented to investigate the characteristics of the fouling layer under sub-critical flux operation. The role of colloid particles and inorganic elements on the membrane foulant were also studied. These findings will aid in optimizing operating conditions and finding the most effective methods for mitigating membrane fouling in the MBR for treating synthetic tomato wastewater.

2. Materials and methods

2.1. Influent source

Tomato processing wastewater discharged from plants is generally characterized as having a high organic concentration which requires pretreatment before disposal. The typical composition is listed in Table 1. The synthetic influent was made by adding tomato juice and deionized water, in the ratio equal to 1:70, which showed a COD in the range

Table 1
Typical composition of real wastewater stream

| Items | Range |
|---|-----------|
| COD (mg L ⁻¹) | 600–1,500 |
| TOC (mg L ⁻¹) | 200–500 |
| NH ₄ ⁺ -N (mg L ⁻¹) | 6–16 |
| SS (mg L ⁻¹) | 50–80 |
| Color (times) | 200–600 |
| pH | 4.0–5.0 |

600–1,500 mg L⁻¹. The quality of synthetic wastewater was listed in Table 2. The real tomato processing wastewater include lots of organic acids, resulting in an acidic nature. Therefore, the Na₂CO₃ solution was used to adjust the pH in the synthetic wastewater.

2.2. Experimental setup and operational conditions

Fig. 1 presents the schematic of the lab-scale SMBR in the experiment. The aerobic reactor had a working volume of 2 L. The membrane module was made of hydrophilized polyvinylidene fluoride (PVDF) with a pore size of 0.3 μm and a filtering surface area of 0.3 m² (Yuanxiang Incorporated, Zhejiang, China). Compressed air was supplied at 10 L min⁻¹ through the air diffuser below the membrane model in order to maintain the desired dissolved oxygen and induce a cross-flow velocity along the membrane surface. The influent synthetic wastewater was transported into the reactor through the peristaltic pump. Seed sludge was taken from a sedimentation tank in the Zhuzhuanjing wastewater treatment plant in Hefei. Next, the mixed liquor suspended solid concentration was adjusted to approximately 3.5 g L⁻¹ before adding to the SMBR. The HRT was approximately 8 h. The SMBR was operated as SBR and each running cycle was 4 h, including 10 min feeding, 180 min aeration, 30 min settling and 20 min effluent discharging. The SMBR was operated under

Table 2
Synthetic influent characteristics of MBR

| Items | Range | Mean value ^a | N ^b |
|---|-----------|-------------------------|----------------|
| COD (mg L ⁻¹) | 700–1,000 | 826 ± 82 | 30 |
| TOC (mg L ⁻¹) | 350–450 | 412 ± 18 | 30 |
| NH ₄ ⁺ -N (mg L ⁻¹) | 10–15 | 13 ± 1 | 30 |
| SS (mg L ⁻¹) | 70–95 | 84 ± 9 | 20 |
| Color (times) | 250–400 | 346 ± 32 | 20 |
| pH | 7.5–8.5 | 8.1 ± 0.3 | 30 |

^aMean ± standard deviation.

^bNumber of measurements.

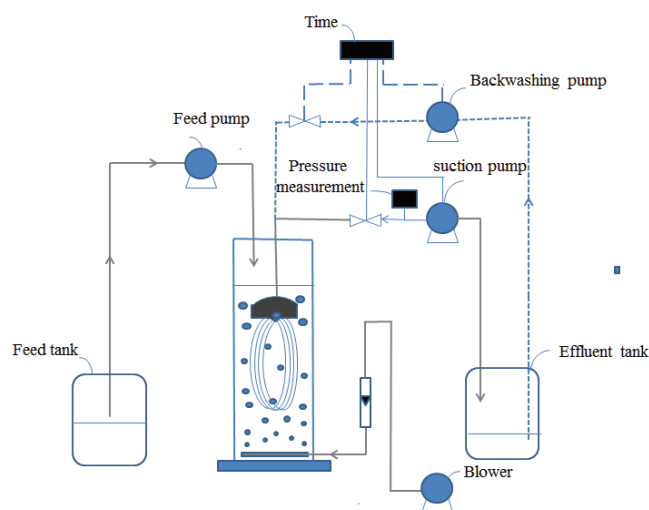


Fig. 1. SMBR schematic.

temperatures in the range of 16°C–20°C, which is nearly same as that in the tomato-processing wastewater treatment plant. The TMP was monitored by pressure gauge, and after the TMP reached approximately 40 kPa, the fouled membrane was taken out of the reactor and physically cleaned.

2.3. Analytical methods

2.3.1. Membrane foulant collection and sample pretreatment

At the end of the operation cycle, the fouled membrane module was taken out and washed with pure water. After that, approximately 500 mL washed liquor was collected from it and mixed well. A separate sample of approximately 500 mL was collected from the sludge suspension on the membrane bioreactor. Then the two samples were prepared for further specific analysis.

2.3.2. Soluble COD and colloidal COD analysis

In order to evaluate which components were mainly responsible for the membrane fouling. The mixed liquor in SMBR was fractioned by size into three parts: soluble, colloidal and supernatant fractions and then their COD concentrations were measured. The supernatant was centrifuged twice for 2 min at $3,000 \times g$, then the soluble COD (COD_s) was obtained after filtering the supernatant through a $0.22 \mu\text{m}$ membrane filter. The colloidal COD (COD_c) was obtained by subtracting the COD_s from the supernatant COD [20].

2.3.3. Particle size distribution analysis

The two sample collections for particle size distribution measurement as described in section 2.3.1 were determined by laser scattering particle analyzer (MS-2000, United States).

2.3.4. SEM and EDX measurement

At the end of the operation cycle, the membrane covered with cake layer was taken out from the MBR and a piece of membrane was cut from the middle of the fouled membrane module. Immediately the sample was fixed with 2.5% glutaraldehyde overnight at 4°C, and then washed three times with 0.1 M phosphate buffer solution for 30 min. After that, the fixed membrane, dehydrated by the concentration gradient of ethanol and silver-coated by a sputter, was imaged on the scanning electron microscope (JEOL Ltd, Japan). The other sample was washed with clean water, also in accordance with the method as mentioned above. The scanning electron microscopy (SEM) coupled with an energy-dispersive X-ray (EDX) analyzer (Phoenix, EDAX Incorporated, USA) was employed to determine the inorganic components of membrane foulants.

2.3.5. FTIR spectroscopy

To characterize the major functional groups of biopolymers in membrane foulants, approximately 200 mL washed liquid, as described in section 2.3.1, was freeze-dried for 48 h according to Ma et al. [21]. The prepared sample was examined by a Fourier transform infrared (FTIR) spectrophotometer (Thermo Electron Corporation, USA).

2.3.6. Analysis of the fouling behavior

In this study, the analysis of the fouling behavior was evaluated by filtration resistance, which was calculated by Darcy's law as show in Eq. (1):

$$J = \frac{\Delta P}{\mu R_t} \quad (1)$$

$$R_t = R_m + R_f + R_c \quad (2)$$

In Eq. (1), J is membrane permeate flux ($\text{m}^3 \text{m}^{-2} \text{s}^{-1}$), ΔP is the membrane pressure (Pa), μ is the permeate water viscosity ($\text{Pa}\cdot\text{s}$), and R_t is membrane total resistance. As presented in Eq. (2), the R_t can be expressed as the sum of intrinsic membrane resistance (R_m), fouling resistance (R_f) and cake resistance (R_c). R_t was determined by measuring the flux of the last day. R_m was measured through the flux of new membrane with pure water. R_f is fouling resistance caused by irreversible adsorption and pore blockage, which was measured by filtration of pure water after removing the cake layer from the membrane surface. R_c is cake resistance by the cake layer formed on the membrane surface, which was obtained by subtracting the R_m and R_f from the R_t .

2.3.7. Other item analysis

Measurements of COD, ammonia and SS were performed according to the standard methods (APHA, 2005) [22]. The total organic carbon (TOC) was measured by a TOC analyzer (TOC-V_{CNP} Shimadzu, Japan). SMP were represented by the dissolved organic carbon (DOC). The sample was acquired by filtering the sludge through the $0.45 \mu\text{m}$ filter, and then the DOC was measured by a TOC analyzer (TOC-V_{CNP} Shimadzu, Japan).

3. Results and discussion

3.1. SMBR performance

After running for some period, the SMBR was close to the steady state. There was no sludge excreted from the bioreactor except the amount pulled for sludge sampling. The following data are obtained from this process. As mentioned previously, a large amount of organic matter was contained in synthetic wastewater. In this study, it was monitored by TOC, because organic matter often could not be easily oxidized by potassium dichromate. The influent and effluent of TOC are presented in Fig. 2(a). Regardless of the fluctuation of influent TOC from 350 to 450 mg L^{-1} , the concentration of TOC in membrane effluent was kept lower than 50 mg L^{-1} . Above 90% TOC removal was achieved in SMBR during the experiment, which demonstrated the great potential of SMBR in tomato processing wastewater treatment. Moreover, the SMBR reactor exhibited excellent nitrogen removal, which was maintained at more than 94% as presented in Fig. 2(b). This could be attributed to the effective membrane retention of slow-growing nitrifying microorganisms [23]. The variation of influent and effluent COD_c concentration with the operation time is presented in Fig. 2(c). The efficiency of COD_c removal

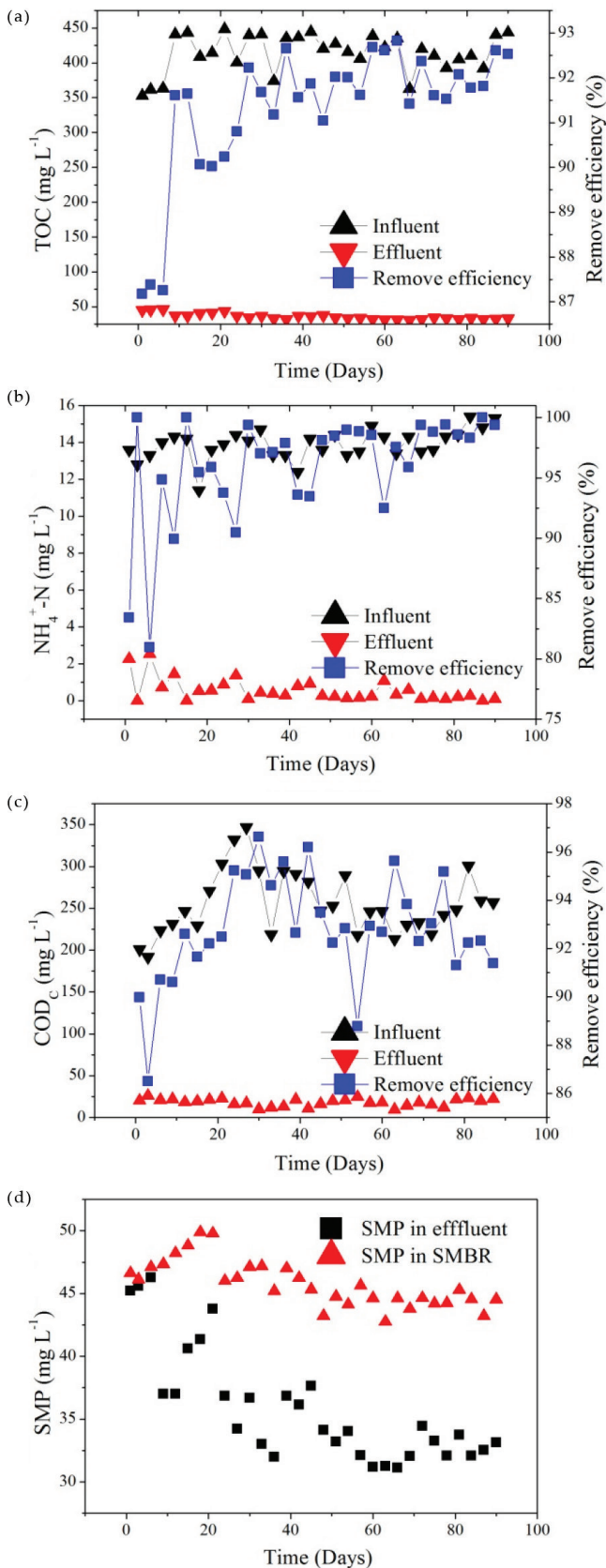


Fig. 2. Evaluation of different concentrations of (a) TOC, (b) $\text{NH}_4^+\text{-N}$, (c) COD_c and (d) SMP in SMBR process.

was remarkably stable at rates of 90%, while the influent concentration ranged from 200 to 350 mg L^{-1} , indicating that SMBR could effectively reduce colloid particles during tomato processing wastewater treatment.

SMP are a group of organic substances mainly within the colloidal substances. They have been found to adversely affect the kinetic activity and the flocculating properties of activated sludge, which have a significant impact on the membrane [24]. Fig. 2(d) shows the concentration of SMP in effluent and in SMBR, respectively. It can be seen that the concentration of SMP in SMBR increased slightly, then decreased and reached a steady value. The results demonstrated that aeration shear stress caused the floc breakage and led to the increase of SMP in sludge suspension after a short period. Moreover, during the operation, the concentration of SMP in SMBR supernatant was always higher than those in effluent. It could be concluded that the membrane acted as a barrier which caused the accumulation of SMP in the reactor. In return, SMP are thought to block membrane pores and form a gel structure on the membrane surface. The findings were similar to those previously reported [25].

3.2. Membrane fouling behavior

The membrane fouling during the operation of SMBR was demonstrated through the monitoring of TMP (Fig. 3). It can be observed that the slow, gradual increase in TMP was followed by a rapid, abrupt increase. During the first period, the rise of TMP was slow. The membrane module was taken out of the bioreactor. The cake layer on the membrane surface could not be observed clearly, which demonstrated that the adsorption of dissolved matter and colloidal blocking were responsible for the fouling resistance [26] in this period. To a great extent, the fouling behaviors demonstrated that the level of membrane fouling was effectively slowed by adopting sub-critical flux operation. After that, a sudden increase of TMP occurred, which attributed to successive closure of pores and resulted in local fluxes exceeding critical value. The growth of the second curve can be expressed as Eqs. (3) and (4):

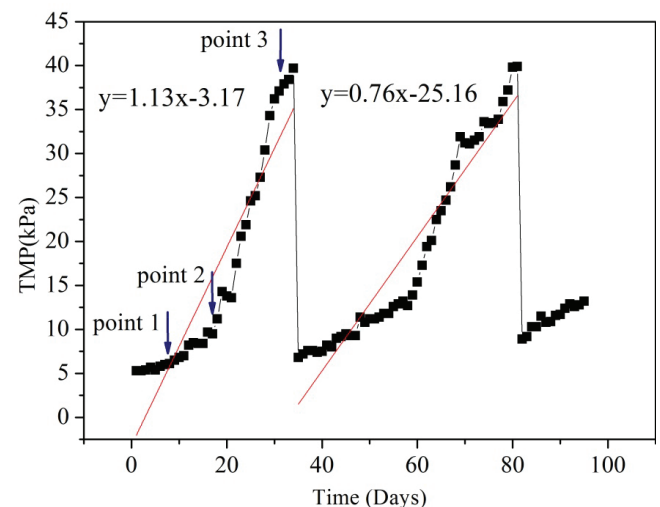


Fig. 3. Variation of TMP during MBR operation.

$$y = 1.13x - 3.17 \tag{3}$$

$$y = 0.76x - 25.16 \tag{4}$$

On days 32 and 80, the membrane module was taken out of the system and then was washed to remove the sludge cake. After the first wash, the TMP dropped dramatically, however, the membrane module was not totally recovered since the gel layer was not completely removed after physical cleaning. The gel layer mainly results from some colloidal substances retained by the membrane during filtration [27]. It could be deduced that the rate rise of TMP with time was slower during the second operation. Combined with the results of section 3.4, after physical washing, the existence of the gel layer enhanced the filtration of organic matter and prevented the membrane pore from blocking some particles.

The TMP under sub-critical flux has been described as a three-stage process [28]: initial “conditioning” fouling, TMP jump and rapid TMP increase. To identify the main contributor to the total resistance at each point, this study analyzed different hydraulic resistance of the membrane. It can be seen from Fig. 4 that intrinsic membrane resistance (R_m) and cake resistance (R_c) appeared to be the minor portion of the total resistance compared with fouling resistance (R_f) at point 1. However, after that, cake resistance steadily increased with the rising of TMP. At point 3, the R_c was nearly fourfold higher than the R_f , indicating that R_c became predominant in the SMBR. The formation of cake layer was dependent on two opposite forces which were suction drag and aeration shear stress. The aeration intensity strongly impacts the formation of sludge cake, therefore, it is an effective way to mitigate the membrane fouling.

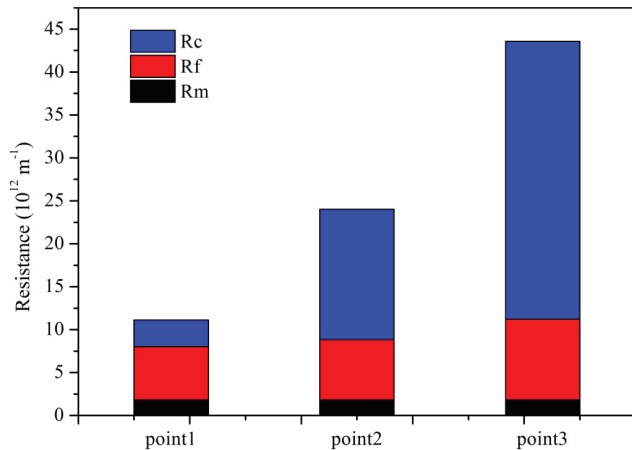


Fig. 4. Resistance through the membrane and biocake at the corresponding points in Fig. 3.

Table 3
Analysis results of membrane foulants

| Components of membrane foulants (%) | | | | Components of sludge suspension (%) | | | |
|-------------------------------------|--|--|--------------------------------------|-------------------------------------|--|--|--------------------------------------|
| SS (g m^{-2}) | COD _s (g m^{-2}) | COD _c (g m^{-2}) | Total foulants (g m^{-2}) | SS (g L^{-1}) | COD _s (g L^{-1}) | COD _c (g L^{-1}) | Total foulants (g L^{-1}) |
| 44.98 (83.2) | 5.22 (9.66) | 3.85 (7.14) | 54.05 (100) | 8.12 (98.4) | 0.097 (1.2) | 0.032 (0.4) | 8.249 (100) |

Note: All the data are transformed to the COD values.

Since the cake layer was important for membrane fouling, further study was conducted to evaluate the cake properties. Different fractions of cake layer and sludge suspension are shown in Table 3. The cake layer and sludge suspension consists of SS, colloidal particles and solutes. It can be seen that SS accounted for the largest proportion, both in membrane foulants and the sludge suspension, which contributed 86.2% and 99.2%, respectively. Although the proportion of colloidal particles and solutes was small, they played an important role in membrane fouling as they caused severe pore blocking. Moreover, compared with the COD_c and COD_s in the sludge suspension, they occupied a larger proportion in the membrane foulants. It is believed that the colloidal particle and solutes had a tendency to deposit on the membrane.

3.3. Particle size of sludge suspension and washed liquid

To understand the effect of microbial characteristics on membrane fouling, the particle size distribution for washed liquid and sludge suspension was determined. From Fig. 5, it can be observed that the sludge suspension had a narrower range profile of size distribution, and the mean particle size was larger than that of washed liquid. The data on the size distribution of two kinds of liquid are summarized in Table 4. More than 75% of the sludge suspension fell in a size range from 60 to 400 μm , with a sharp peak at 150 μm . On the contrary, in the washed liquid only 34% of the particles are distributed from 60 to 400 μm . These figures indicate that particle size in the washed liquid was much smaller than in the sludge suspension in the SMBR.

Previous research [29] has shown that membrane fouling was mainly attributed to cake formation; however, this study indicates that the particle size acts as the major role in this process. The small particle size was due to the presence of colloids and the substances which originate from the release of EPS. During the process of pressure-driven membrane separation, a portion of these particles enter the membrane pore and remain deposited on the membrane, forming the cake layer. Moreover, small particles often are difficult to remove under aeration force.

3.4. SEM and EDX analysis

SEM analysis revealed sludge cake deposits on the membrane surface and the structure of the fouling layer [30]. The fouled membrane and cleaned membrane were presented in Figs. 6(a) and (b), respectively. In Fig. 6(a), it shows that the membrane fouling is extremely serious, as the cake layer formed on the membrane surface was made up of bacteria clusters covered with biopolymers. In Fig. 6(b), after physical washing, the fouling layer was much smoother

than before, the thick sludge layer was reduced and some membrane holes were clearly distinguishable. However, the membrane surface was still covered by a slime gel layer, which is called irreversible fouling. Once the gel layer is formed, it is believed to be more resistant to removal by shear due to its cross-linked structure [31]. Therefore, a more effective method to alleviate the fouling layer needs to be developed.

The study sought to identify the chemical components on the fouling layer by element analysis. The elements of C, F, O, K, Al, S, Si and Ca were revealed, as presented in Fig. 7. The sharp peak of F mainly resulted from the film itself, which was made up by PVDF. Although the relative contents of K, Al, Si and Ca were small, these elements coupled with the organic foulant as mentioned above was a major contributor to the formation of the gel layer [32]. Moreover, it shows that the MBR with internal membrane system had more serious inorganic fouling compared with the external. Previous studies also emphasized the important role of inorganic fouling in membrane system [33]. The membrane fouling caused by inorganic matter could not be eliminated easily. Therefore, the biopolymers, such as colloids and solute, together with inorganic matter in the SMBR under sub-critical flux operation should be controlled. Based on the characteristics of inorganic elements and colloidal particles, on the one hand, it is possible to control it by pretreating the influent. On the other hand, it could be controlled through the operation parameters, such as HRT and SRT [34].

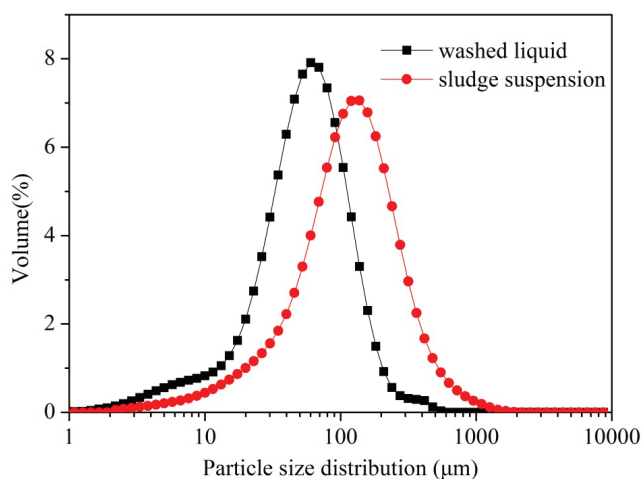


Fig. 5. Comparison of particle size distributions between the washed liquid and the sludge suspension.

Table 4

Statistical result of sludge particle size distributions in washed liquid and sludge suspension

| | Particle size (μm) | | Particle size distribution (%) | | | |
|-------------------|---------------------------------|------|--------------------------------|---------------------|----------------------|--------------------|
| | Mean | Peak | <10 μm | 10–60 μm | 60–400 μm | >400 μm |
| Washed liquid | 73 | 70 | 5.52 | 60.63 | 33.42 | 0.43 |
| Sludge suspension | 167 | 150 | 2.07 | 18.34 | 75.25 | 4.34 |

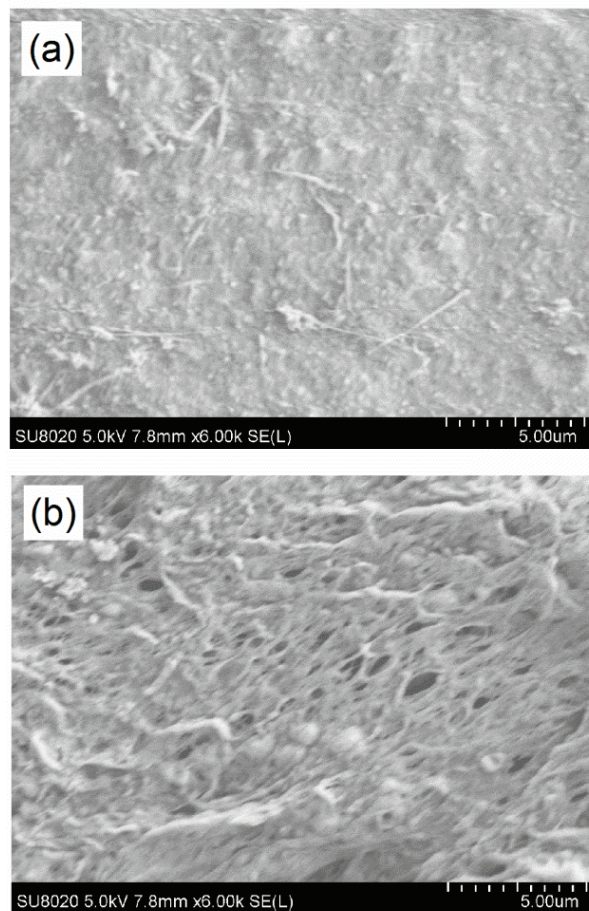


Fig. 6. SEM image showing (a) fouled membrane and (b) cleaned membrane.

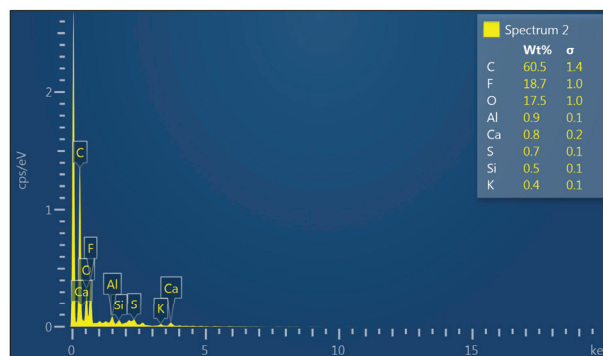


Fig. 7. EDX analysis of fouled membrane surface.

3.5. FTIR analysis

EPS are reported as playing a major role in membrane fouling. Since the EPS accumulated both on membrane module and mixed liquor. The spectra of membrane foulants and EPS are presented in Fig. 8. In Fig. 8(a), the spectrum shows a broad region of absorption at $3,421\text{ cm}^{-1}$, due to the stretching of the O–H bond in hydroxyl functional groups, and a sharper peak at $2,919\text{ cm}^{-1}$, which is attributed to stretching of C–H bonds [35]. It is observed that there are three sharp peaks ($1,650$; $1,542$ and $1,261\text{ cm}^{-1}$) in the spectrum, which are unique to the proteins' secondary structure, namely, amides I, II and III [36]. It follows that the proteins are components present on the fouling layer. Based on the broad peak at $1,052\text{ cm}^{-1}$, the membrane foulant consisted mainly of polysaccharides or polysaccharides-like substances.

Compared with Fig. 8(a), the spectrum of EPS was presented in Fig. 8(b), and shows peaks of $3,415$, $2,917$ and $1,654\text{ cm}^{-1}$. This demonstrates the existence of O–H, polysaccharides and proteins in the EPS. Although these results are similar to those mentioned above, the number of peaks between $1,000$ and $1,600\text{ cm}^{-1}$ is considerably less than the membrane foulant spectra, indicating that the organic matter in EPS is much simpler than that in membrane foulant. This analysis shows that EPS are only part of the membrane fouling layer, and further study is needed on other complicated

organic substances. The results compared with FTIR spectra of membrane foulant obtained by [37] show that a significant membrane fouling mainly stemmed from EPS and inorganic materials.

4. Conclusions

In this study, the performance of the laboratory-scale SMBR treating tomato synthetic wastewater was investigated. The study demonstrated that the average removal efficiencies of TOC, ammonia, colloidal particles achieved approximately 90%, 94% and 90%, respectively. The quantity of effluent has reached the standard. During the operation, cake resistance was seen as the main cause of severe membrane fouling. The components of cake layer and sludge suspension were divided into SS, colloidal particle and solutes. It was proved that pore blocking resulted from the colloidal particles and solutes, which are more likely deposited on the membrane surface than those of sludge suspension. The fine particle tended to be deposited on the membrane surface, which acts as a major role in cake formation. The FTIR spectra demonstrated that the membrane foulants had more peaks than that of EPS. It was concluded that the membrane foulant consists not only of the EPS but also other organic matter. SEM and EDX demonstrated that a dense cake and gel layer was deposited on the fouled membrane surface, and consisted not only of organic substances but also inorganic elements, such as Ca, K, S and Si. Due to the existence of colloidal and its cross-linked structure, the gel layer caused the irreversible fouling on the membrane surface.

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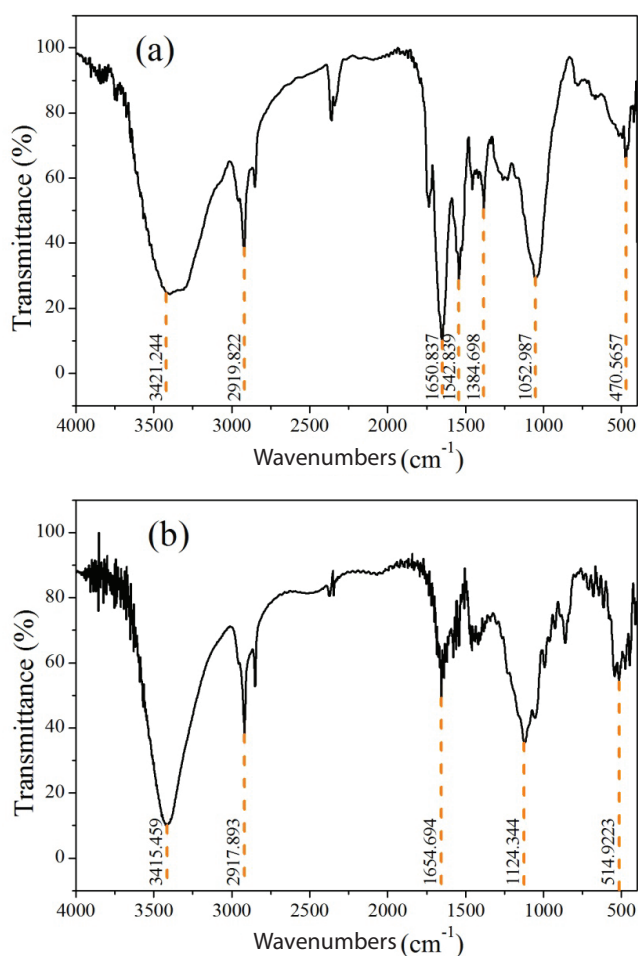


Fig. 8. FTIR spectra of (a) membrane foulants and (b) EPS.

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