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Performance of MBR in treating tomato paste processing wastewater: a comparison of aerobic granules with activated sludge

Hui Zhou[a], Kuizu Su[a,b,*], Tong Shu[a], Tianwei Hao[c,*], Weihong Wang[d], Wei Wang[a], Li Liu[e]

[a]*Department of Civil Engineering, Hefei University of Technology, Hefei 230009, China, Tel. +86 551 62905853;*
emails: sukz@hfut.edu.cn (K. Su), 13966763012@163.com (H. Zhou), 673983099@qq.com (T. Shu), wang_wei@hfut.edu.cn (W. Wang)
[b]*Anhui Provincial Engineering Laboratory for Rural Water Environment and Resources, Hefei 230009, China*
[c]*Department of Civil & Environmental Engineering, University of Macau, Macau, China, Tel. +853 88224945;*
email: twhao@umac.mo (T. Hao)
[d]*College of Hydraulic and Civil Engineering, Xinjiang Agricultural University, Urumqi 830091, China, Tel. +86 13899910629;*
email: 2209319288@qq.com (W. Wang)
[e]*School of Chemistry, Beihang University, Beijing 100191, China, Tel. +86 15210265304; email: lililiu@buaa.edu.cn (L. Liu)*

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ABSTRACT

Membrane bioreactor technology has become increasingly popular for treating various industry wastewater because of its high treatment efficiency, small footprint and excellent effluent quality. Performance of aerobic granules (AG) and activated sludge (AS)-based submerged membrane bioreactors implemented for tomato paste processing wastewater treatment was investigated and compared in this work, particularly in terms of membrane fouling and microbial community structure at different conditions. The results revealed that the chemical oxygen demand in the aerobic granular system showed slightly better removal efficiencies (92%) than those in the activated sludge system (90%) while the performance of activated sludge membrane bioreactor is more stable over the experimental test time. According to the total filtration resistance, the aerobic granular system demonstrated superiority initially, while the activated sludge system performed better with disintegrated aerobic granules. Indeed, the membrane fouling resulted from disintegrated aerobic granules was most serious, which was further confirmed by atomic force microscopy. In addition, the cleaning efficiency was 96% in the aerobic granular system, which was nearly 10% higher than that of the activated sludge system. The difference between the bacterial community structures in both systems was analyzed, and the dominant bacteria were transformed into *Rhodocyclaceae* and ASSO-13 at the family level indicating that consortia gradually became the predominant bacteria in the tomato paste processing wastewater.

Keywords: Tomato paste processing wastewater; Membrane bioreactors; Aerobic granules; Activated sludge; Membrane fouling; Microbial community structure

1. Introduction

Tomato paste processing wastewater is characterized by a dark colour, a malodourous smell, and the inclusion of large numbers of organics, colloidal fractions and suspended solids (SS), which exhibit a poor settling ability and slow biodegradation [1–3]. The concentration of pollutants in the effluent varies considerably with time and space due to the changes in the harvested fruit composition and season [4]. Hence, efficient treatment means are needed to satisfy the increasing requirement of water quality and to reduce wastewater cost.

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* Corresponding authors.

The submerged membrane bioreactor (MBR) is a combination of the membrane and activated sludge process [5,6]. Degradation of organic pollution is contributed to the adapted microorganisms, while the separation of microorganisms from the treated wastewater is performed by the membrane [7,8]. MBR offers several advantages, including the complete removal of solids from an effluent, superior nutrient and organic removals and low sludge production rate, which have been considered to be one of the most promising processes for wastewater treatment [9]. Previous studies reported that MBR showed good performance in treating various high strength wastewater, such as municipal wastewater [10], textile wastewater [11] and food wastewater [12] for organics and color removal, which can be justified by the elevated biodegradability and the high biomass. However, the major obstruction of the traditional activated sludge membrane bioreactor (ASMBR) is membrane fouling, which causes either an increase in the operation costs or a decline in the permeate flux [13].

To mitigate membrane fouling, many studies have focused on the new design of an MBR type [14]. Aerobic granular sludge is a new form of microbial self-aggregation [15], possessing advantages of good settling capacity, ability to withstand impact load rates and complex environmental conditions over activated sludge [16]. Replacing the activated sludge by granular sludge in the MBR system has been proposed to mitigate membrane fouling [17]. Tay et al. [18] investigated pollutant removal and membrane fouling in granular sludge-based membrane bioreactor (AGMBR) and ASMBR and found that AGMBR demonstrated a better performance. The large size and rigid structure of the aerobic granules expectedly reduced pore blocking and cake layer formation in AGMBR [19]. However, there are few studies that focus on systematic investigation of the performance and membrane fouling of AGMBR in purifying tomato paste processing wastewater. Whether the membrane fouling mitigation can still be inherited; can AGMBR achieve a long-term stable condition? These unknowns are about to be addressed in this work. Aerobic granules were introduced into a bench-scale MBR, and AGMBR and ASMBR were simultaneously operated in intermittent mode to treat synthetic tomato paste processing wastewater.

The objectives of our study were (i) to compare the performance of nutrient removal efficiency and membrane fouling in AGMBR and ASMBR for treating synthetic tomato paste processing wastewater, (ii) to investigate the difference of the total filtration resistance and the cleaning efficiency of the two systems, and (iii) to study the difference in the membrane fouling morphology and the functional microbial community structure in the two MBRs.

2. Materials and methods

2.1. Reactor set up and operating conditions

Fig. 1 presents the schematic of the lab-scale SMBR in this experiment. The reactors are made of polymethylmethacrylate with a diameter of 9 cm and an effective volume of 2.5 L. The membranes used in both reactors were polyvinylidene fluoride hollow fiber membranes with a length of 13 cm, a pore size of 0.2 μm and a membrane area of 0.15 cm^2. Compressed air was supplied at 10 L min^{-1} (the superficial air velocity was 2.62 cm s^{-1}) through the air diffuser below the membrane model in order to maintain the desired dissolved oxygen. The hydraulic retention time was approximately 8 h. The MBRs were operated sequentially with a cycle time of 4 h, which included 2 min influent feeding, 218–223 min aeration, 5–10 min settling and 10 min effluent withdrawal, with a volumetric exchange ratio of 50%. All experiments were performed under ambient temperature (20°C ± 5°C) which is nearly the same as that in the tomato-processing wastewater treatment plant [20].

2.2. Seed sludge and wastewater

The seed sludge acquired from the wastewater aeration tank (Zhuzhuanjing wastewater treatment plant in Hefei, China). Aerobic granules were cultivated in a sequencing batch reactor (SBR) which had a working volume of 6 L with an internal diameter of 11.5 cm. The SBR was fed with synthetic wastewater consists of 500–1,500 COD mg L^{-1} at 25°C ± 1°C and pH 7.0 ± 0.1. The settling time was shortened from 5 to 1 min gradually. The sludge volume index decreased continuously for a few days and then stabilized at 22 ml g^{-1}. Granules started to appear after 60 d operation. After the granules formed and stabilized, the sludge was seeded to the MBR. The activated sludge with the same operating conditions was transferred to another membrane bioreactor. The mixed liquid suspended solids (MLSS) of the two reactors were 2.5 g L^{-1} in the commissioning period. The production of tomato paste leads to the generation of wastewater from cleaning processes [1] and the typical composition is listed in Table 1 [20]. Table 2 presents the main characteristics of the synthetic tomato paste processing wastewater, which was made by diluting concentrated synthetic wastewater, in the ratio equal to 1:70.

2.3. Membrane resistance analysis

The analysis of membrane resistance was calculated by Darcy law as shown in Eq. (1) [20]:

$$R_m = \frac{\Delta P}{\mu_w J_w} \tag{1}$$

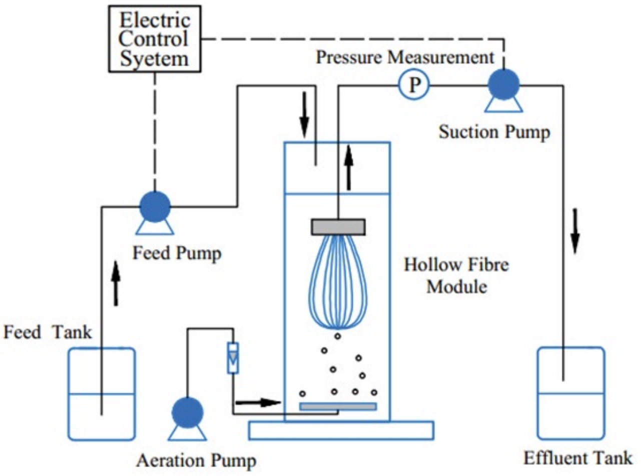


Fig. 1. Schematic diagram of the experimental system.

Table 1
Typical composition of real tomato paste processing wastewater

Component	Concentration
COD (mg L^{-1})	600–1,500
NH$_4^+$–N (mg L^{-1})	6–16
SS (mg L^{-1})	50–80
Color (CU)	200–600
pH	4.0–5.0

Table 2
Components of synthetic tomato paste processing wastewater

Component	Concentration
COD (mg L^{-1})	700–1,000
NH$_4^+$–N (mg L^{-1})	10–20
SS (mg L^{-1})	60–90
Color (CU)	300–600
pH	7–8

$$R_t = R_m + R_r + R_{\mathrm{ir}} \tag{2}$$

μ_w is the permeate water viscosity (Pa s), J_w is membrane permeate flux (m^3 m^{-2} s^{-1}), ΔP is the membrane pressure (Pa), R_t is membrane total resistance which can be expressed as the sum intrinsic membrane resistance (R_m) reversible fouling (R_r), and irreversible fouling (R_{ir}) resistances. R_m was measured through the flux of new membrane with pure water, R_{ir} and R_r can be calculated at the end of filtration after washing with water, alkaline, and acid detergents and after washing with water, respectively.

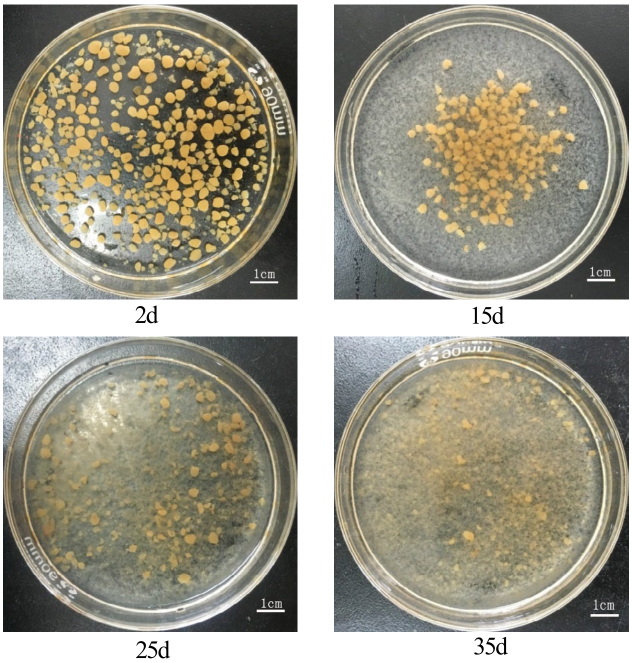
2.4. Other analytical methods

Chemical oxygen demand (COD) and color in the liquid samples were measured following the standard methods [21]. The particle size distribution of the sludge samples was measured by a laser particle size analysis system (MS-2000, United States). The analysis of membrane resistance was made according to Darcy's law, and the experimental procedure to determine each resistance value followed Wang et al. [22]. In addition, the fouled membrane was cleaned by physical and chemical methods in turn, which followed the procedure of Li et al. [23]. The membrane surface morphology and topography were taken using a digital instrument atomic force microscopy (AFM) [24]. With NanoScope Analysis software, the roughness analysis of the layer was conducted using AFM. Both systems for microbial community investigation were detected using the high throughput microbial analysis method [25]. AG1, AG2, and AG3 represent the seed sludge, suspended sludge, and cake layer sludge of AGMBR, respectively; AS1, AS2, and AS3 also symbolize the same meaning for ASMBR.

3. Results and discussion

3.1. Morphology evolution of aerobic granules in AGMBR

As shown in Fig. 2, in the initial period, the faint yellow aerobic granules were ellipsoidal with an average diameter of 1.50 mm. On day 15, the obvious disintegration of granules took place in the reactor, and the floc sludge increased. In addition, a large number of flocs were retained in the reactor due to the blockage of membrane modules. To a certain degree, the disintegration of granular sludge possibly due to the influent COD/N ratio and a major microbial community shift [26], which is consistent with the result of this study. The aerobic granules further disintegrated on day 25. Nevertheless, large-sized particles of aerobic granules can still be observed in the reactor. On day 35, almost all of the aerobic granules disintegrated into flocs.



2d 15d
25d 35d

Fig. 2. Variation of aerobic granule morphology in the AGMBR.

Granular analyses, which are provided in Fig. 3, reveal the size distribution of the two systems. The median particle size of aerobic granules was reduced from 681.68 to 125.85 μm, which indicated that the aerobic granules in the membrane bioreactor had disintegrated completely, which is consistent with Fig. 2. In addition, the particle size of activated sludge in ASMBR was between both mentioned above.

3.2. Comparison of performance of the MBR systems

Compared to other wastewater, tomato paste processing wastewater has more colloidal and suspended solids and higher color. Fig. 4 shows the removal rate during continuous treatment, which was similar to the results of other studies when the granular sludge and activated sludge were used in tomato paste processing wastewater treatment [27,28]. Adverse impacts imposed by the tomato paste processing wastewater were not observed in these MBRs. The removal rate of AGMBR can reach more than 90% in the first 15 d. After 15 d, the removal rate of COD by AGMBR decreased

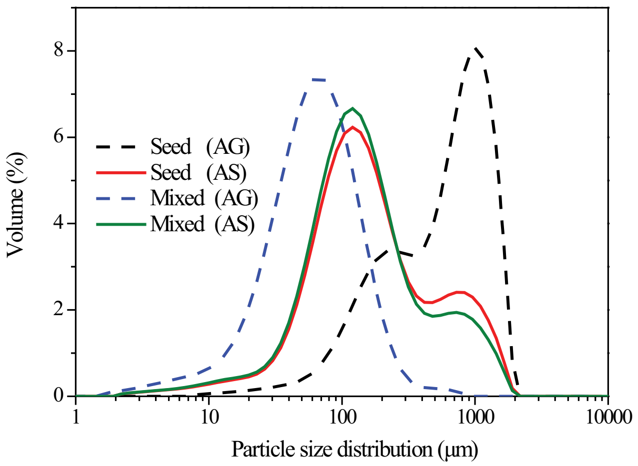


Fig. 3. Particle size distribution of the sludge at different stages.

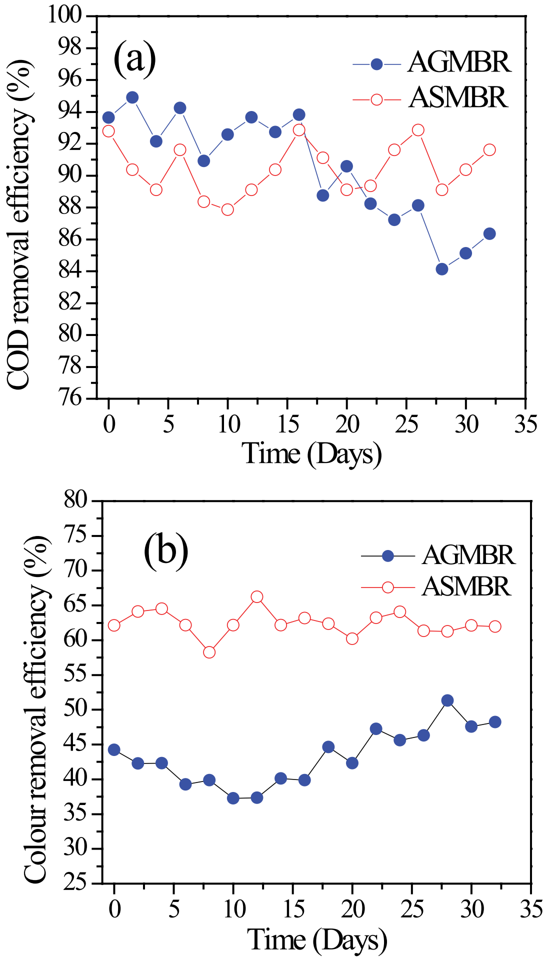


Fig. 4. Comparison of the removal performance in the MBR systems: (a) COD and (b) Color.

obviously. However, the removal efficiency of ASMBR was kept steady in the same operation.

It was noticed that the treatment of color removal includes biological methods employing different fungi, bacteria and algae and physico-chemical methods such as adsorption coagulation/precipitation, oxidation and membrane filtration [29]. Color removal of ASMBR (60%) was higher than that of AGMBR (40%) (Fig. 4b). Pala and Tokat [30] studied the low biodegradability of many dyes and textile chemicals and indicated that biological treatment is not always successful in the treatment of cotton textile wastewater, in terms of color removal. This is due to the smaller specific surface area of the aerobic granules compared to that of the activated sludge, which has weaker adsorption of the color. Therefore, the removal rate of color improved when the aerobic granules disintegrated.

3.3. Comparison of membrane resistance

To compare the membrane fouling rate of AGMBR and ASMBR, the total filtration resistance of the two systems was investigated (Fig. 5). The resistances increased with the proceeding of the experiment in both systems. The fouling development was affected by the biomass characteristics

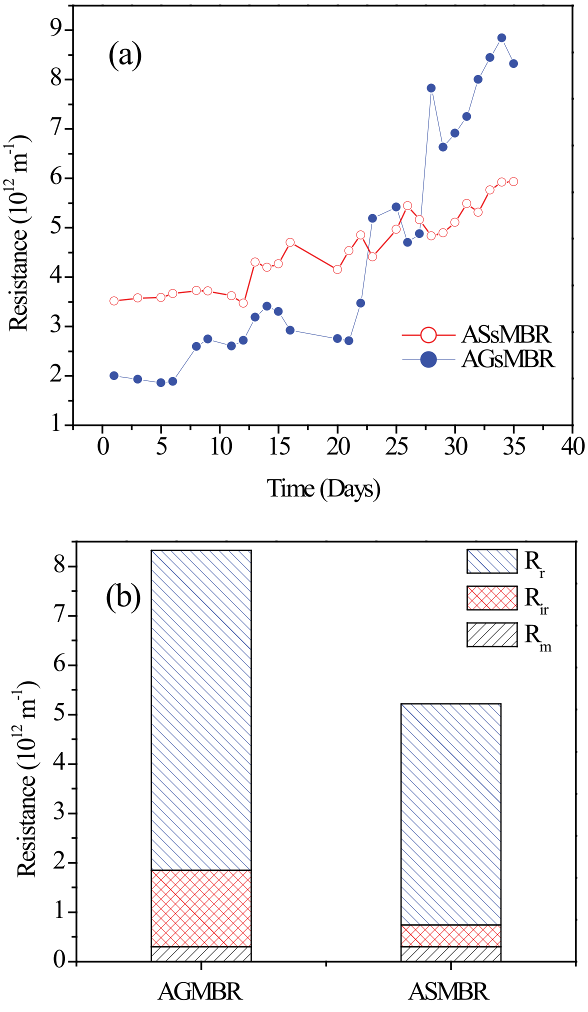


Fig. 5. Comparison of membrane resistance in MBR: (a) total filtration of resistant and (b) hydraulic cleaning reversibility of filtration resistance (R_m: membrane resistance, R_r: membrane reversible resistance, R_{ir}: membrane irreversible resistance).

Table 3
Roughness analysis of the membrane fouling layer

Roughness	New membrane	Fouled membrane (AGMBR)	Fouled membrane (ASMBR)
Ra (nm)	103	79	86
Rq (nm)	121	93	102

and the process variables, such as the filtration flux, sludge concentration and aeration intensity [31]. The ASMBR system did not present a sharp resistance increase within the given operating period.

For the AGMBR, on the other hand, during the initial 20 d operation, the rising rate of total membrane fouling in AGMBR was lower than that in ASMBR. The introduction of aerobic granules into the MBR system was beneficial for mitigating membrane fouling through controlling the dominating foulant [23]. However, the filtration resistance of AGMBR increased sharply as the particles gradually disintegrated and even reached 8.32 × 10^{12} m^{-1} at the end of the operation, which can be seen from the evolution of particle size distribution in Fig. 3. Wang et al. [31] studied the sludge cake that was attached to the membrane with an unusually high filtration resistance, which resulted in serious membrane fouling in the SMBR treatment process. Hence, the AG did not easily flatten to the membrane surface or enter into the interior of the membrane through pores to form membrane fouling. According to the results in Fig. 3, the compressibility of sludge increased as the amount of small- and medium-sized sludge increased in the AGMBR. Therefore, these disintegrated aerobic granules trended to easily deposit onto the membrane surface by the suction pressure, which led to pore blockage, thus resulting in the filtration resistance of AGMBR being larger than that of ASMBR during the later period of the operation.

The hydraulic cleaning reversibility of the filtration resistance for the two systems in 35 d is revealed in Fig. 5. The final reversible resistance accounted for 77.83% and 81.6% in the AGMBR and ASMBR systems, respectively, during the examined time. To summarize, physical cleaning could effectively decrease the membrane resistance, which indicated that the fouling was caused mainly by the deposition of floc sludge for the MBR system. However, chemical cleaning could achieve a recovery of 96% in the AGMBR, which was nearly 10% higher than that of the ASMBR system. This finding is in agreement with the results of Park et al. [32].

3.4. Comparison of the membrane surface morphologies between AGMBR and ASMBR

AFM has been proven to be a rapid method for assessing membrane-solute interaction (fouling) of membranes [33]. The three-dimensional images (Fig. 6) of the membrane surface morphologies of the two systems were taken by AFM, which illustrate the figures of the foulants that accumulated on the membrane surfaces [34]. The images show some differences in the morphologies of the two fouled layers (day 35): the fouling layer peaks of membrane AGMBR were low, smooth and dense, while those of ASMBR presented deeper, rougher peaks and more holes. Previous research [35] showed that better filtration performance was mainly attributed to the cake layer that formed on the membrane surface. Therefore, the fouling layer that formed on the membrane of the AGMBR surface may have poor permeability with pore blockage and lead to a faster fouling rate than that of ASMBR. With the computation results (Table 3), the surface roughness analyses of the image that were carried out with NanoScope Analysis software further demonstrated the prediction. The values of Ra (79 nm) and Rq (93 nm) which are square root roughness and root-mean-square roughness of the fouled membrane in AGMBR were significantly greater than those of ASMBR, and there was a positive correlation between the numerical value and the roughness of the membrane surface. Combined with Fig. 5, the membrane roughness of AGMBR in this experiment was larger, and the total filtration resistance was also larger. This was likely due to the filling of the membrane valleys and pore structures with foulant materials [36]. So the fouling layer formed on the membrane of the AGMBR surface may have poor permeability and lead a faster fouling rate than that of ASMBR. Vrijenhoek et al [37] studied the effect of the roughness of the membrane surface on the permeate flux. The results showed that at the initial stages of fouling, AFM images clearly showed that more particles deposited onto the rough membranes than onto the smooth membranes. Particles preferentially accumulate in the "valleys" of rough membranes, thus resulting in "valley clogging", which causes a more severe flux decline than that of smooth membranes.

3.5. Analysis of the microbial community structure

Fig. 7 illustrates the dominant composition of bacterial communities of the sludge samples in different phases. In both systems, *Proteobacteria* showed the highest relative abundance at the phylum level accounting for over 45%, and the dominated bacteria at the class level was also similar and mainly consisted of *Betaproteobacteria* in any samples. However, the obvious difference of the dominant bacterial communities was displayed at the family level, which was consistent with previous research on bacterial communities from sewage [38], membrane bioreactors [39] and tomato paste processing wastewater treatment [1].

Previous studies [40,41] showed that the *Proteobacteria* and *Bacteroidetes* phyla were the predominant bacteria in most MBR treatment processes and were also the two predominant consortia that could adapt best in response to changes in the external environment. It can be seen from Fig. 7 that the relative abundance of *Proteobacteria* and *Bacteroidetes* gradually decreased during the tomato paste

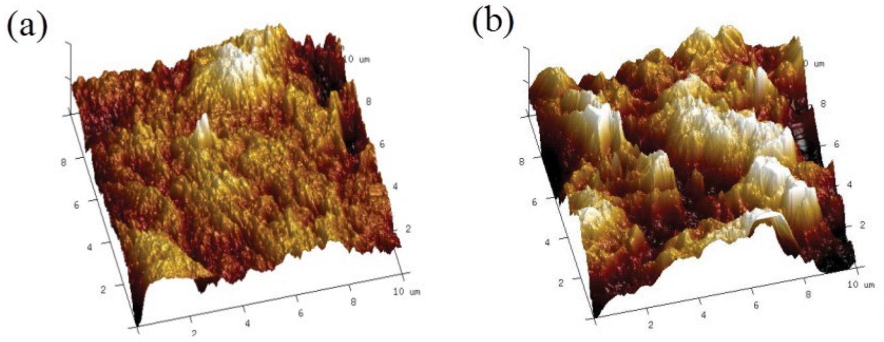


Fig. 6. Membrane surface morphology obtained by AFM: (a) fouled membrane surface in AGMBR and (b) fouled membrane surface in ASMBR.

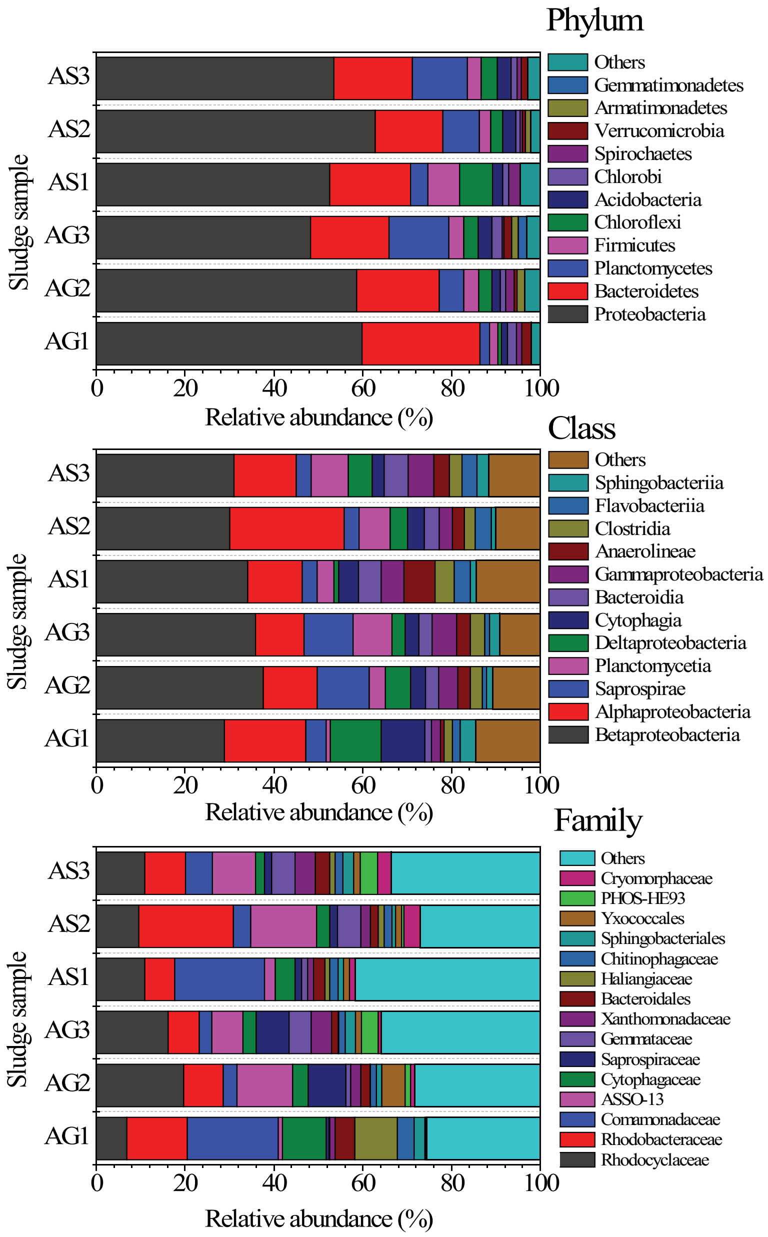


Fig. 7. Abundance of bacterial community structures in both ASMBR and ASMBR systems ("Others" represents all classified taxa that were below 2% in all samples).

processing wastewater treatment process in our AGMBR system. Conversely, *Planctomycetes* accounted for 2.1% and 3.9% in the inoculated sludge of AGMBR and ASMBR at the phylum level, respectively, while the proportion increased to 13.5% and 12.3% in the suspended sludge during the later stage of the systems, which suggested that the bacteria gradually became predominant in the tomato paste processing wastewater treatment. It is noted that *Firmicutes* represented 3.4% and 3.1% of the AG3 and AS3, respectively. The *Firmicutes* were also mentioned in other studies [42], although it was almost non-existent in late operation. Similar results can be observed at the class level. However, at the family level, the dominant populations included *Comamonadaceae* and *Cytophagaceae* in seed sludge samples at the end of the experiment. The dominant bacteria were transformed into *Rhodocyclaceae* and ASSO-13, which further revealed that *Rhodcyclaceae* and ASSO-13 were mainly responsible for the effectiveness of the tomato paste processing wastewater treatment. The interaction between different bacterial populations may play an important role in the degradation of tomato paste processing wastewater and the stability of the membrane bioreactors and deserves continued investigation.

3.6. Discussion of mechanism

This preliminary study of the ASMBR and AGMBR pointed out undeniable difficulties for both the granules formation and their maintenance. In SBR, the aerobic granules with small size and poor settling capability can be discharged easily by controlling the hydraulic selection pressure such as decreasing sedimentation time. However, the effluent in MBR was pumped out by a suction pump connected to the membrane module and the smaller particles can not be discharged, which leads to the absence of the hydraulic selection pressure. As the experiment goes on, it is difficult to achieve the maintenance of aerobic granular sludge in MBR. In this case, periodically manually selection pressure would be recommended in the MBR system, for example, periodically manually sludge discharge is available for the maintenance of reactors. Besides, the control of sludge retention time results in reduce the extracellular polymeric substances (EPS) production by bacteria, which could mitigate the membrane fouling as well.

4. Conclusion

Based on the obtained of this study, we draw the following conclusions:

- Compared to activated sludge, aerobic granules in MBR showed a better removal performance, but aerobic granules disintegrated gradually with the experiment process, which could be a concern for this process.
- For the filtration resistance, although lower than the aerobic granules, activated sludge is better than the disintegrated aerobic granules. Furthermore, the cleaning efficiency could achieve 96% in the AGMBR, which was nearly 10% higher than that of ASMBR.
- The roughness of the cake layer by AFM indicated that the introduction of aerobic granules into the MBR system is benefited for mitigating membrane fouling, but disintegrated granular sludge presents a more serious membrane fouling.
- There was a high similarity of bacterial community structures between the suspension and the microorganisms on the cake layer, and the microbial community richness on the cake layer was higher than that of the suspension. Although the two systems differ in the microorganism community structure, the dominant bacteria were transformed into *Rhodocyclaceae* and ASSO-13 on tomato paste processing wastewater treatment.
- New experiments are necessary to focus on biological and management issues for the aerobic granular sludge stability in MBR. For example, the selective pressure should be created with decreasing sedimentation time and the organic loading rate should be increased gradually. Besides, sludge retention time can be controlled to reduce the EPS production by bacteria which could reduce the membrane fouling as well.

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