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Experimental Study on Membrane Fouling and Cleaning in Hydrolytic Acidification -Membrane - Sequencing Batch Reactors Process

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Abstract: The process of hydrolytic acidification (HA) - membrane (M) - sequencing batch reactors (SBR) was used to deal with mixed municipal wastewater and landfill leachate. The system operated continuously for 90 days, and the changes of membrane filtration resistance and membrane cleaning were investigated. The results showed that the membrane fouling rate developed slowly during the continuous operation of the system, and the total membrane filtration resistance increased from the initial 2.82×10^{12} m^{-1} to 11.4×10^{12} m^{-1}. In addition, it was found that the reversible membrane fouling was the main part of the membrane-sequencing batch filter, which accounted for 47.35% of the total filtration resistance, by observing the surface of the membrane module before and after cleaning test. What's more, hydraulic cleaning could effectively remove the sludge and granular sediments deposited on the membrane surface, and the total resistance of membrane filtration decreased from 293.84% to 176.45%. However, by further soaking membrane components with 5‰ NaClO and 5‰ HCl solutions, the membrane filtration performance would be fully restored, and the total membrane filtration resistance would be restored to 104.35% of the initial state.

1. Introduction

Membrane separation technology is an efficient water treatment technology, but membrane fouling limits the application of membrane bioreactor [1]. Membrane fouling can be divided into physical fouling, chemical fouling and biological fouling according to its properties, which is mainly affected by membrane material properties, pollutants in water and membrane operation conditions [2]. In the actual research, different researchers have different understanding and classification of the fouling resistance outside the membrane inherent resistance, because of the different characteristics of the membrane and the filtrate and in order to establish the corresponding membrane fouling model [3-6]. However, in broad terms, membrane fouling is a general term for all the factors causing flux attenuation except the intrinsic resistance of the membrane.

In the membrane Bioreactor, an aerator can be installed under the membrane assembly to prevent the formation of a fouling layer on the membrane surface by turbulence of the membrane surface

caused by high intensity aeration, while keeping the membrane flux below the critical membrane flux, the development speed of membrane fouling was controlled by constant current operation and intermittent suction, so as to prolong the service life of membrane and reduce the cleaning frequency [7]. In addition, Fan et al. [8] studied the influence of fillers on the operation stability and membrane fouling characteristics of membrane bioreactor (MBR), it was found that the addition of fillers in the membrane area could improve the properties of the sludge mixture, increase the average particle size of the sludge, and prolong the operating time of the membrane. However, no matter what operation mode is adopted, the membrane flux will decrease with the increase of operation time due to membrane fouling. Therefore, certain methods should be adopted to clean the membrane module.

There are many cleaning methods of membrane, because of the different water quality of sewage treatment, the cleaning methods used are also different, which of membrane fouling include physical cleaning and chemical cleaning. Physical cleaning mainly for isobaric cleaning, backwashing, gas-liquid mixing oscillating cleaning, negative pressure cleaning and so on [9]. Commonly used chemical cleaning reagents include alkali, acid, metal chelating agent, surfactant, oxidant and enzymes, salts [10].

In recent years, hollow fiber membrane has been widely used in environmental protection fields such as sewage recycling, drinking water treatment and seawater desalination due to its high filling density and low market price [11-12]. Polyvinylidene fluoride (PVDF) membrane has been developed rapidly in many fields because of its advantages of acid and alkali resistance, easy cleaning, high water output and high rejection rate.

In this study, the characteristics of membrane fouling in the treatment of municipal mixed wastewater and landfill leachate by hydrolytic acidification- membrane- sequencing bioreactor (HA-M-SBR) were investigated using a PVDF hollow fiber membrane module, the membrane components were cleaned by hydraulic cleaning, acid cleaning and alkaline cleaning, and the changes of membrane filtration resistance were evaluated. According to the effect of hydraulic cleaning, membrane fouling was divided into reversible fouling and irreversible fouling [13-14], and the composition of membrane fouling was analysed, which would to provide reference for practical application of HA-M-SBR.

2 Materials and methods

2.1 Experimental device

In this experiment, an integrated membrane bioreactor was used. The experimental apparatus was shown in Figure 1, which was divided into three parts: regulating tank, hydrolysis tank and membrane tank. The mixed municipal wastewater was treated with the fresh leachate from the garbage incineration power plant at a ratio of 400:1. In order to slow down the pressure rise caused by membrane pollution, constant current operation and intermittent pumping were adopted for the system [7], with the pumping set at 8 min and the pumping stopped at 2 min. The test membrane module was made of PVDF hollow fiber microfiltration membrane, the membrane aperture was 0.1 m, and the surface area of the single membrane module was 0.50 m^2.
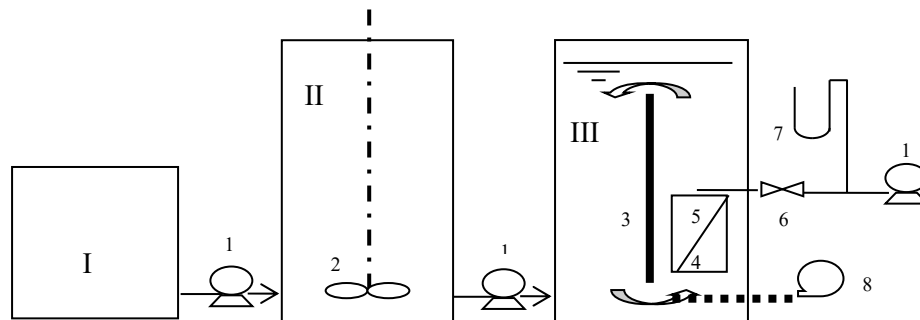
I: Raw water regulation basin; II: Hydrolytic tank; III: M-SBR tank. 1-Peristaltic pump; 2-Stirring device; 3-Partitioning plate; 4-Microporous aerator; 5-Membrane module; 6- Solenoid valve; 7- Differential pressure gauge; 8-Air compressor

Figure 1. System flow chart

2.2 Experimental method

2.2.1 Cleaning of fouling film. After 90 days of continuous operation, the membrane filtration resistance increased obviously, and the fouling membrane module was removed for cleaning experiment. In this experiment, the membrane module was washed by water for 5 minutes, then soaked in 5‰ NaClO solution for 18 hours, and finally soaked in 5‰ HCl solution for 18 hours.

2.2.2 Membrane filtration resistance analysis method. Membrane fouling is usually characterized by pollution resistance, which is proportional to membrane filtration pressure difference and inversely proportional to membrane flux. The substances that cause membrane fouling in a particular reactor vary greatly with the water quality of the test, so the characteristics of membrane fouling in different reactors are also different. In this experiment, membrane fouling resistance is divided into three parts: one is reversible membrane fouling resistance R_r, the other is inherent membrane resistance R_m, the last part of it is irreversible membrane pollution resistance R_{ir}, the total filtration resistance R_t of membrane components in a bioreactor at a certain time is as follow:

$$R_t = R_m + R_r + R_{ir} \tag{1}$$

According to Darcy Equation [6], the membrane flux and membrane filtration pressure were measured, and the resistance and proportion of each part were calculated by Resistance-In-Series (RIS) resistance model [15]. The expression is as follows:

$$R_t = \triangle P / (\mu_t J_t) = R_m + R_r + R_{ir} \tag{2}$$

$$R_m = \triangle P / (\mu_0 J_0) \tag{3}$$

$$R_{ir} = \triangle P / (\mu_t J_t) - R_m \tag{4}$$

$$R_r = \triangle P / (\mu_t J_t) - R_m - R_{ir} \tag{5}$$

Where, J_t is the membrane flux of the mixture during operation, $m^3/(m^2 \cdot s)$; J_0 is the membrane flux of clean water filtration test, $m^3/(m^2 \cdot s)$; μ_0 is viscosity of water, $Pa \cdot s$; μ_t is the viscosity of membrane filtrate, $Pa \cdot s$, approximately equal to the viscosity of clear water at the same temperature.

3. Results and discussion

3.1 Surface characteristics of fouling membrane
When the membrane module was removed from the Bioreactor, a thick layer of mud cake was formed on the surface of the membrane filament, and a large amount of sludge was deposited in the root of the

membrane module, and the sludge color turned black. This is due to the fact that the suspended matter and other solid matter in the Bioreactor can easily block the pores of the membrane and adsorb and deposit in the pores. As the Gel layer and the solid deposit layer on the surface of the membrane become thicker and denser, the bottom sludge becomes anaerobic and the sludge is black in colour.

(a) Water washing
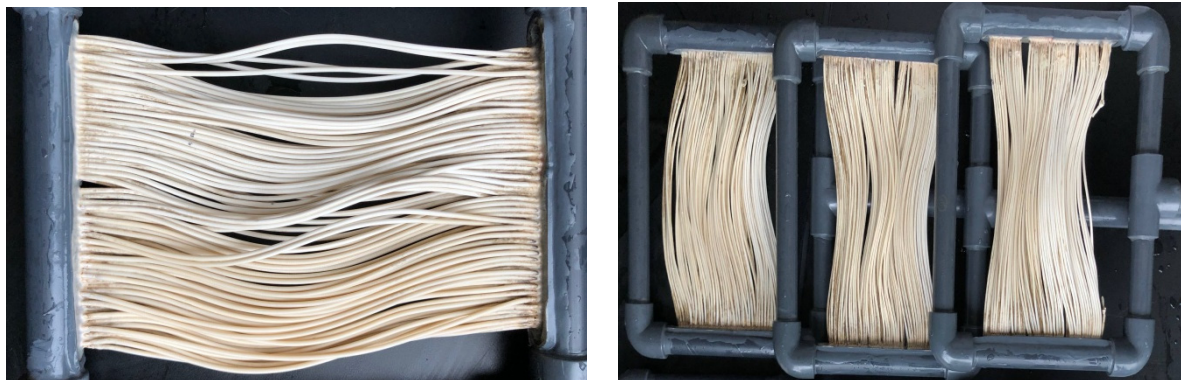
(b) 5‰ NaClO
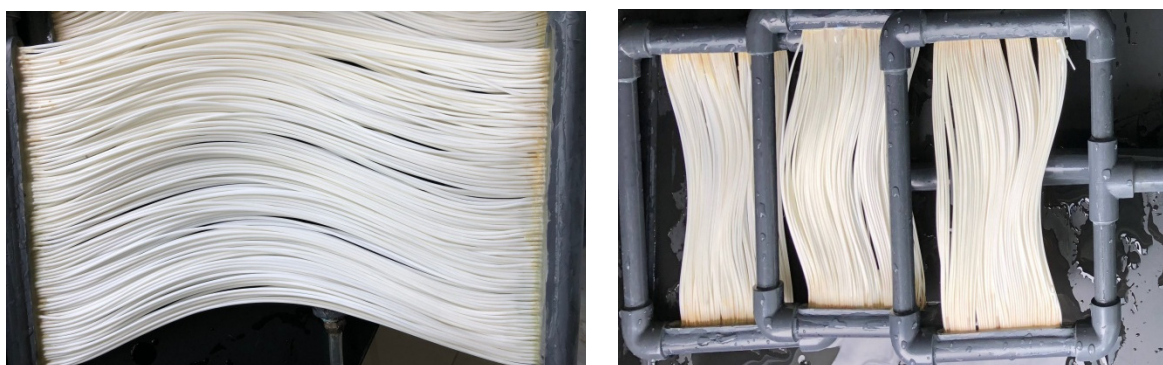
(c) 5‰ HCl

Figure 2. Apparent appearance change after cleaning membrane module

The polluted membrane assembly can be rinsed with clean water and rubbed slightly to remove most of the sludge deposited by the root of the membrane assembly and the membrane filaments. The surface of the membrane filaments that have been deposited by the sludge is black. This is because during operation, due to the alternations of hypoxia/aerobic conditions, the inner sludge anaerobic respiration attached to the mud cake layer on the outer surface of the membrane filament [16], as

shown in Figure 2 (a). After soaking the membrane module with 5‰ NaClO solution for 18h, it can be found that the black substance on the outer surface of the membrane silk has been basically removed, but there are still a lot of brown substances, as shown in Figure 2(b). After being cleaned with 5‰ NaClO solution and then soaked with 5‰ HCl solution for 18h, most of the membrane silk surface turned white and the brown substance was basically removed, as shown in Figure 2 (c).

3.2 Change of total membrane filtration resistance

The system has been running continuously for 90 days without any cleaning during this period, and Figure 3 is the curve of total resistance of membrane filtration with running time. As can be seen, the membrane filtration resistance has been rising steadily. At the end of the experiment, the total membrane filtration resistance increased from the initial 2.82×10^{12} m^{-1} to 11.4×10^{12} m^{-1}, with an average rising rate of 1.0×10^9 (1/m·d^{-1}). The results showed that membrane fouling developed slowly in the process of the experiment, mainly because the supernatant was filtered by hollow fiber microfiltration membrane in the settling stage of the SBR process, the adsorption and deposition of suspended matter and sludge particles on the surface of the membrane during the suction process were reduced, the membrane fouling in the separation process was effectively reduced, and the stability of the membrane filtration performance was well maintained.
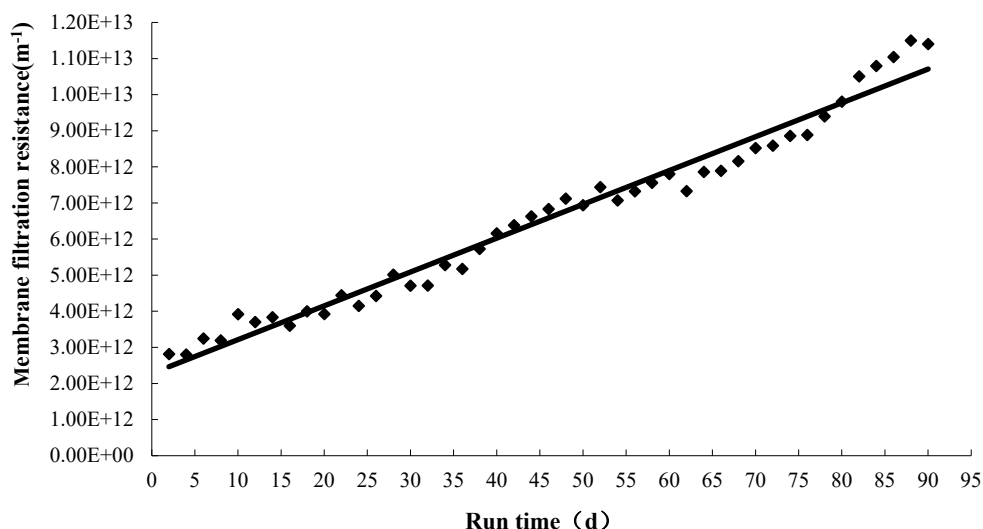

Figure 3. Change of membrane filtration resistance

3.3 Effect of fouling Membrane cleaning on total resistance of membrane filtration.

The membrane filtration resistance was calculated by water flux test after cleaning, and compared with the resistance of the new membrane itself to characterize the recovery of the membrane filtration performance. The total membrane filtration resistance after each step of cleaning is compared in Figure 4.

It can be seen from Figure 4 that after 90 days of continuous operation, most of the sludge attached to the membrane surface could be removed through hydraulic external washing and slight rubbing of the membrane wire surface. The total membrane filtration resistance decreases from 293.84% to 176.45% of the initial membrane resistance, and the membrane filtration performance is greatly recovered. After soaking for 18h in 5‰ NaClO solution, the total membrane filtration resistance decreased to 121.74% as at the beginning. After further immersion in 5‰HCl solution for 18h, the total membrane filtration resistance further decreases to the initial 104.35%, which performance basically returns to the new membrane state. Generally, alkali solution can effectively remove protein contamination, destroy gel layer, and make it peel off from the membrane surface. Acid cleaning agent can dissolve and remove inorganic minerals, so that the Ca^{2+}, Mg^{2+} and other inorganic ions in the gel layer and scale layer are dissolved out, and the remaining gel layer and scale layer are thoroughly

eluted from the film surface to restore its permeability ability. The results show that the contribution of organic and biological pollutants to membrane fouling is greater than that of inorganic.
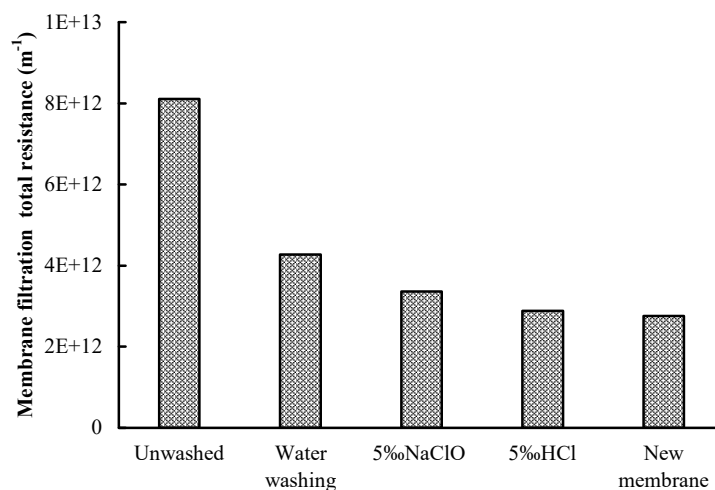

Figure 4. Cleaning effect of hollow fiber membrane

3.4 Composition of membrane fouling

On the 90th day of continuous operation under the experimental conditions, the membrane filtration resistance caused by reversible fouling was 47.35% of the total membrane filtration resistance, and the membrane filtration resistance caused by irreversible fouling was 18.62%. This shows that membrane filtration resistance caused by reversible membrane fouling is dominant in the process of HA-M-SBR treatment of municipal wastewater and landfill leachate, the operating parameters of the system can be optimized by controlling the aeration intensity and operating under the critical membrane flux to reduce the deposition rate of contaminants to the membrane surface, thus reducing the development rate of reversible membrane fouling and prolonging the membrane filtration cycle, reduce the frequency of membrane cleaning.

4. Conclusions

(1) In the experiment, the hollow fiber microfiltration membrane was used to filter the supernatant in the settling stage of SBR process, which reduced the adsorption and deposition of suspended matter and sludge particles to the surface of the membrane during the process of suction, the average rising rate of membrane filtration resistance was 1.0×10^9 (1/m·d^{-1}), which could keep the stability of membrane filtration performance in the system, and provided a reference for the project of coordination treatment of municipal mixed sewage and landfill leachate by HA - M - SBR process.

(2) It was found that the membrane resistance caused by reversible membrane fouling accounted for 47.35% of the total membrane filtration resistance by observing the surface of the fouling hollow fiber membrane and comparing the cleaning effect on the membrane module, the rising speed of membrane filtration resistance could be reduced by controlling the operating process parameters. Besides, Hydraulic cleaning would effectively remove the sludge and sediment deposited on the surface of the membrane, and the filtration performance of the membrane could be recovered to a great extent. Moreover, by using 5‰ NaClO and 5‰ HCl solution to further membrane assembly, the total resistance of membrane filtration can be restored to the initial state.

References

[1] Pulido, J.M.O. (2016) A review on the use of membrane technology and fouling control for olive mill wastewater treatment. Science of the Total Environment, 563/564: 664-675.

[2] Li, W.G., Wang, L.Y., Zhang, W.Z., Wang, B.H., Wang, X,Y. (2014) PVDF membrane fouling control and cleaning method discussion. Technology of water treatment, 2014, 40 (3): 91-93.

[3] Cui, X.Q., Lin, J.F., Shen, H., Jin, Q. (2012) Dominated pollutants and fouling resistance of membrane surface in an aerobic membrane bioreactor treatmenting landfill leachate. Chinese Journal of Environmental Engineering, 2012, 6 (6): 1980-1984.

[4] Melin, T., Jefferson, B., Bixio, D., Thoeye, C., Wilde, W.D., Koning,J.D., Graaf, J.V.D., Wintgens, T. (2006) Membrane bioreactor technology for wastewater treatment and reuse. Desalination, 187 (1-3): 271-282.

[5] Chen, Y.Z. (2016) Research on membrane fouling control by process of pre-oxidation enhanced coagulation-membrane to remove algae. Membrane Science and Technology, 36 (1): 104-108.

[6] Bai, L., Qu, F., Liang, H. (2013) Membrane fouling during ultrafiltration (UF) of surface water: Effects of sludge discharge interval(SDI). Desalination, 319:18-24.

[7] Bae, T. H., Han, S. S., Tak, T. M. (2003) Membrane sequencing batch reactor system for treatment of dairy industry wastewater. Process Biochemistry, 9 (2): 221-231.

[8] Fan, J.W., Yang, Y.Z., Yang, H.B., Li, Y.G (2020) Influence of the filler addition method on characteristics of operation stability and membrane fouling in MBR. Chinese Journal of Environmental   Engineering. http://kns.cnki.net/kcms/detail/11.5591.X.20200212.0750.002.html

[9] Li, Q.G. (2017) Analysis and cleaning process optimization for column membrane bioreactor membrane fouling. Technology of water treatment, 43 (10): 65-66.

[10] Lee, H., Amy, G., Cho, J., Yoon,Y., Moon, S.H., Kim, I.S. (2001) Cleaning strategies for flux recovery of an ultrafiltration membrane fouled by natural organic matter. Water Research, 35 (14): 3301-3308.

[11] Xiao, K., Xu, Y., Liang, S., Lei, T., Sun, J.Y., Wen, X.H., Zhang, H.X., Chen, C.S., Huang, X. (2014) Engineering application of membrane bioreactor for wastewater treatment in China: Current state and future prospect. Front Environ Sci Eng., 8 (6): 805-819.

[12] Mu, S.T, Fan, H.J., Han, B.J., Xiao, K., Liu, C., Huang, X. (2018) Review of membrane fouling stages and mathematical models for hollow fiber membrane. Membrane Science and Technology, 38 (1): 114-121.

[13] Zhou, C., Wu, D.H., Lu, G.H., Wang, Z.L., Wei, L., Han, F. (2019) Research progress in the chemical control over membrane fouling in membrane water treatment. Industrial Water Treatment, 39 (2): 6-10.

[14] Jelemensky, M., Sharma, A., Paulen, R., Fikar, M. (2016) Time-optimal control of diafiltration processes in the presence of membrane fouling. Computers & Chemical Engineering, 91: 343-351.

[15] Gao, Y.L., Wang, L.Y., Wang, W.F., Song, X.W. (2012) Filtration Performance Study on Membrane-Sequencing Bioreactor. Technology of Water Treatment, 38 (5): 28-30.

[16] Fu, W.X., Li, L. (2004) Mechanism and method for membrane washing on a membrane bioreactor. Techniques and Equipment for Environmental Pollution Control, 2004, 5 (8): 43-46.