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Continuous-stirring of a Granular Sludge Immersed Membrane Bioreactor for Treating Food Wastewater

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Abstract— **To reduce the membrane fouling in an immersed membrane bioreactor (MBR) during the treatment of food wastewater, an additional continuous stirring was introduced. The results showed that both a better membrane performance and a higher degradation efficiency for food wastewater were obtained. The membrane contributed mostly in concentrating the activated sludge. The bioreactor ensured the degradation of the organic matter. The resistance caused by concentration polarization decreased under continuous–stirring, and the membrane flux reached 14.68 L/h*m^2, which was 3 times the membrane flux under non-stirring condition. The COD removal and the ammonia nitrogen removal reached 93% and 90%, respectively.**

Index Terms— **food wastewater, granular sludge, membrane bio-reactor, stirring.**

I. Introduction

Food wastewater has high contents of bio-degradable organics (over 92% on a dry basis) and a large portion of water, hence of easy decay, leading to insect proliferation and malodorous emissions. Food waste also contains a variety of pathogenic bacteria, causing serious human hazards [1].

Flocculation and gravity separation are two traditional wastewater treatment technologies which have a limited potential to treat food wastewater. Flocculation is mature and of low cost, but requires a high dosage of flocculating agent. At high contents of colloidal matter, the treatment is insufficient and a large quantity of activated sludge will be generated. Flotation is suitable for oil and fat removal. Settling is imperfect for dealing with food wastewater which contains a high suspended solids content (5%) with possible coalescence and clogging effects [2]. Since these treatment methods are inefficient or not environmentally friendly, the use of a membrane bioreactor (MBR) to treat food wastewater is gaining interest.

Over the past two decades, the membrane bioreactor technology has been developed as a new way to treat wastewater efficiently. MBR is a modified activated sludge process, in which the activated sludge is concentrated by a microfiltration membrane unit in a bioreactor [3]-[5]. MBR` is now widely applied for domestic wastewater and has major advantages [6], and could be a potential treatment technique for the food wastewater.

Previous studies have reported that a MBR can reach a relatively high efficiency for high solid content wastewater treatment. Qiu [7] used sieve silk as membrane material to treat domestic wastewater. Under the condition of 6h hydraulic retention time (HRT) and membrane flux of 66.2L/(m^2·h), the average removal rates of COD, ammonia nitrogen, total nitrogen and total phosphorus were 94.0%, 97.6%, 49.2% and 83.7%, respectively. Voorthuizen [8] used an anaerobic MBR, an aerobic MBR and an UASB reactor which was followed by an effluent membrane filtration module to treat black (toilet) water and reported average COD removal rates of 86%, 91% and 91%, respectively. Scholz [9] treated oil contaminated wastewater in a MBR with sludge concentration up to 48 g/L. The largest biodegradation of fuel oil reached 0.82 g hydrocarbons degraded per day per gram MLVSS. The average biodegradation is 0.26-0.54 g hydrocarbons per day per gram MLVSS. The average COD removal and TOC removal was 94%, 96% for fuel oil, 97% and 98% for lubricating oil, respectively. It showed that wastewater containing oil even surfactants could be biodegraded in a MBR.

The MBR treatment of a high organic content wastewater is a challenge, mostly due to membrane fouling, which is mostly associated with the attached microbial growth on the membrane surface. Membrane fouling is the key factor that restricts the long-term operation of MBR [10], and around 60% of the MBR operating cost is attributed to membrane fouling [11]. Lots of efforts to avoid membrane fouling in MBR processes have been made through design, material, selection and fundamental research.

Many modifications were studied to control membrane fouling, such as seeding MBR with granular sludge [12] and coupling MBR with suspended carriers [13], [14]. Zhao et al. [15] and Liu et al. [16] demonstrated that membrane fouling was reduced with the addition of powder activated carbon, the reasons were mainly attributed to the decrease of the extracellular polymeric substances in microbial cells, the cake resistance reduction, the increase of floc size distribution and the decrease of viscosity.

Although the methods mentioned above can partly control membrane fouling, the decrease of membrane permeability remains inevitably due to pore clogging and membrane fouling. Few publications assess the effect of agitation which is equally important in the MBR process. Agitation can be regarded as a fundamental approach to avoid membrane fouling. Also an oxidation process can remove the ammonia nitrogen in food wastewater. The aeration of a traditional MBR can only partly remove the ammonia nitrogen, while the addition of stirring can further strengthen this removal.

In this study, the influence of agitation on the performance of food wastewater degradation and membrane fouling

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control in MBR was studied. The Influence on the removal of COD and TN by the addition of continuous-stirring was investigated. The kinetics and total resistance of the MBR process were also studied.

II. Materials and Methods

A. Experimental setup

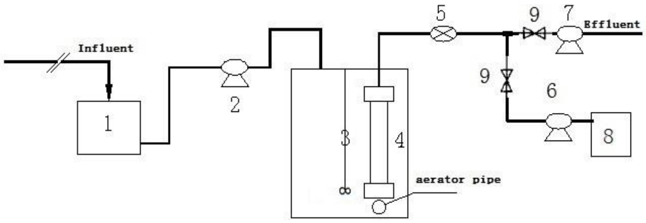

Fig.1 Schematic setup of the MBR

1. Influent tank; 2. Influent pump; 3. Stirring paddle; 4. Microfiltration membrane; 5. Vacuum meter; 6. Backwash pump; 7. Effluent pump; 8. Backwash tank; 9. Time relay

The system schematic (Fig.1) included a buffer tank, MBR, pumps and some accessories. The MBR was built in acrylic resin, of 100L volume (0.46m diameter; 0.78m height). A multi-orifice aeration tube was installed on the bottom to inject air into the liquid phase and enhance the agitation. A stirring paddle was installed 200mm above the bottom. The diameter of the paddle of 20 cm and the rotating speed of 130 rpm was chosen.

A membrane module which contained 50 polyvinylidene Fluoride (PVDF) hollow fibers was used in this study. The module was disinfected with 0.11% (w.t.) NaClO solution for 2 hours before the experiments. Each hollow fiber was 900mm long with an outside diameter of 2.1 mm and inside diameter of 0.9mm. The diameter of the micro-pores was 0.04μm. The effective area of the membrane module was 0.297 m^2. After sterilization, the membrane module was immersed in the liquid during the experiments while the inner space of each hollow fiber was connected by manifold to a vacuum pump to maintain a negative pressure. The membrane flux of the MBR was measured under a given pressure of 51.5 kPa. The operation parameters were listed in Table 1.

Table 1 Operating parameters

Item	*Parameter*
Organic loading rate	21.6 g COD/d
Temperature	30℃
Hydraulic retention time	100 hr
aeration flow rate	10L/min
Stirring speed	130r/min
Cycle time	15min
Influent/backwash time	13:2
Backwash	Deionized water
Transmembrane Pressure	51.5 kPa

B. Characteristics of inoculum and feedstock

The microorganisms were inoculated in the MBR prior to the experiment. The MBR was stably operated for 30 days with the acclimatized membrane. The TS of the sludge was kept at 8,000 mg/L in the MBR during the stable operation.

Activated sludge (about 20000 mg SS/L) was obtained as inoculum from a wastewater treatment plant in Shunyi District, Beijing, China. The sludge was thickened using a frame filter to reach a moisture content of 75-80%. The concentrated sludge was air dried at ambient temperature for 20 days and stored at -20 ℃ for later experiments.

The food wastewater was collected as substrate from a campus dining hall of the Beijing University of Chemical Technology. The MLSS, SV_{30}, pH, COD and ammonia nitrogen (NH_4^+-N) of the inoculum and feedstock were analyzed according to the Standard Method [17].

The COD, NH_4^+-N and pH of original food wastewater were 80 000-100 000 mg/L, 800-1200 mg /L, 6.8-7.5, respectively. To achieve an acceptable organic loading in the MBR, the leachate was diluted with tap water to a COD of 800-1000 mg/L, ammonia nitrogen concentration of 8-12 mg/L and pH of 6.8-7.5.

C. Fundamental parameters

The total resistance of the membrane was calculated (based on the Darcy law) by Eq. (1) and the resistance contributions can be described by Eq. (2).

$$R = \frac{\Delta p}{\mu J} \qquad (1)$$

$$R = R_m + R_c + R_i + R_p \qquad (2)$$

where R is the total resistance of filtration (m^{-1}); J is the permeation flux (L/ (m^2 ·h)); Δp is the transmembrane pressure (Pa); μ is the absolute viscosity of the leachate (Pa.s).

R_m (membrane resistance) is the hydraulic resistance of the clean membrane (m^{-1}): the membrane was firstly immersed with 0.11 wt. % of NaClO solution. The flux of NaClO solution and the operation pressure were determined to calculate R_m.

R_p (polarization resistance) is the resistance due to the polarization (m^{-1}): after the operation, the membrane was directly immersed in the water, the flux of water and the operation pressure were determined to calculate R_0, R_p was the difference between R and R_0.

R_c (cake resistance) is the resistance arising from the cake formation (m^{-1}): after the determination of R_0, the sludge was removed from the membrane, then the membrane was immersed in the water, the flux of water and the operation pressure were determined to calculate R_1, R_c was the difference between R_0 and R_1.

R_i (inner resistance) is related to the absorption and blockage of the membrane pores, hence the resistance of inside fouling (m^{-1}): R_i was the difference between R_1 and R_m.

Lim et al. [18] reported that the principle of membrane fouling can be described by the combination of a membrane resistance control model (Eq. (3)), a pore blocking resistance model (Eq. (4)) and a cake resistance model (Eq. (5)) based on the different membrane fouling degrees of the whole MBR process.

$$\frac{1}{J(t)} = \frac{1}{J(0)} + k_m t \qquad (3)$$

$$\ln J(t) = \ln J(0) - k_p t \qquad (4)$$

$$\frac{1}{J^2(t)} = \frac{1}{J^2(0)} + k_c t \qquad (5)$$

where, J (t) is the membrane flux at the time of t (m^3/m^2), J(0) is the membrane flux at the beginning, t is the period of filtration, k_m, k_p and k_c are system parameters related to the membrane resistance, pore blocking resistance, and cake resistance, respectively.

The variation of flux with operation periods could be obtained from the slope of the appropriate regression analysis.

III. Results and Discussion

A. Influence of agitation on the membrane performance

In MBR process, the membrane performance is mainly reflected by the membrane flux and total resistance. Total resistance, the key factor to determine the membrane flux, is made up of membrane resistance, polarization resistance, cake resistance and inner resistance. The membrane flux is influenced by different resistance in different periods. Since the agitation has a direct influence on the membrane performance, the influence of additional continuous-stirring was investigated.

a. Influence of agitation on the membrane flux

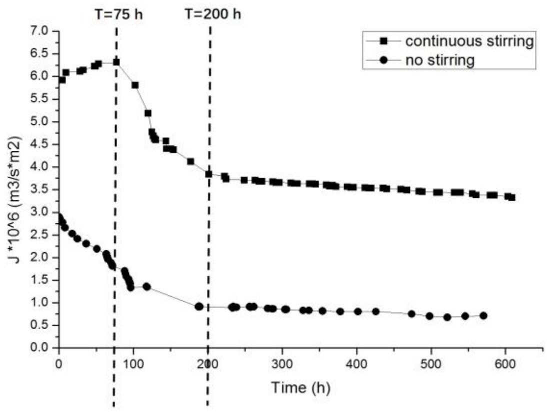

Fig.2 Variation of the membrane flux with time

One of the membrane performance influenced by agitation was the membrane flux. Fig.2 indicated that under continuous-stirring, the cross-flow shear force was mainly provided by the stirring instead of aeration, and the membrane flux reached a relatively high level of 14.68 L/h*m^2, which was 3 times the membrane flux under non-stirring as the shear at the membrane surface increased. In contrast, under non-stirring, sludge and organic macromolecules covered onto the membrane surface and thus the viscosity increased, since all the cross-flow shear force was provided by the aeration. Because of the low stress of the aeration at the membrane surface, the membrane flux was relatively low, only 4.88 L/h*m^2.

Compared to the non-stirring condition, the membrane flux fluctuated more in the first 75h under the continuous-stirring. This is due to the factor that under non-stirring, the colloidal solids, sludge particulates and dissolved matters were deposited onto the membrane surface by the trans membrane pressure, and the shear force formed by aeration was too weak to affect the deposition, leading to a steady increase of the total resistance and decrease of the membrane flux. Under continuous-stirring, larger sludge particulates were more easily washed away by the shear force under the alternation of deposition and washing, thus the membrane flux showed a slight fluctuation.

Table 2(a) Model fitting at different stages of operation under continuous -stirring

Stage	Model	Model fitting	R^2
0-75h	Membrane resistance control model	$\frac{1}{J} = -157t + 1.68 * 10^5$	0.85
75-200h	Pore blocking resistance model	$\ln J = -4.6 * 10^{-3} t - 11.64$	0.89
>200h	Cake resistance model	$\frac{1}{J^2} = 4.78 * 10^7 t + 6.02 * 10^{10}$	0.99

Table 2(b) Model fitting at different stages of operation under non-stirring

Stage	Model	Model fitting	R^2
0-200 h	Pore blocking resistance model	$\ln J = -6.23 * 10^{-3} t - 12.77$	0.98
>200h	Cake resistance model	$\frac{1}{J^2} = 2.68 * 10^9 t + 5.68 * 10^{11}$	0.93

In Table 2(a) and (b), the cake resistance model which lasts the longest period is the most important contribution compare to membrane resistance control model and pore blocking resistance model. Under continuous–stirring, the model of the whole process could be expressed as membrane resistance control model (0-75h), pore blocking resistance model (75-200h) and cake resistance model (>200h), according to the different periods. Under no–stirring, the model of the whole process could be expressed as pore blocking resistance (0-200h) and cake resistance model (>200h). The difference under continuous–stirring was reflected in 0-75h, since the influence of pore blocking was reduced by continuous–stirring, the origin membrane resistance became the main factor that affected the membrane flux. Between 75-200h, the holes on the surface of the membrane began to be blocked, leading to the pore blocking resistance becoming one of the main factors that affected the membrane flux. Although the amount of the colloidal solids was much smaller than that of the sludge flocs, the dissolved matter was still deposited onto the membrane surface and absorbed into the membrane pores, leading to an irreversible membrane fouling. These three factors all affected the total resistance although the sludge particulates were the major contribution, thus explaining the difference of the resistance models between non-stirring and continuous-stirring in the first 200h. It was also indicated that the addition of continuous–stirring could reduce the influence on total resistance by pore blocking in the initial period.

After the first 200h of operation, the sludge particulates and colloidal solids were steadily deposited in the cake on the membrane surface, leading to the increase of the total resistance and the decrease of the membrane flux. The dissolved matter was also deposited onto the membrane surface, but the concentration of dissolved matter on the membrane surface began to be higher than that in the bio-reactor, so that the dissolved matter on the membrane surface were sheared back into the bio-reactor back-diffusion. When the deposition equals the back diffusion, the effect of the dissolved matter can be ignored. So the membrane fouling after 200h was reversible. It can be relieved by backwash. The cake resistance dominates the total resistance, thus making it the key factor to prevent membrane fouling.

Since the cake resistance model is the main part of the whole process, it is important to compare its value with and without stirring. The slope of the model under continuous–stirring was $4.78*10^7$, which is much smaller than that under non-stirring ($2.68*10^9$): The flux decreased more slowly under continuous–stirring than under non-stirring, because J is the denominator. It indicated that the cake resistance was much less than that without stirring, leading to a higher membrane flux during this period.

b. Influence of agitation on the membrane flux

As proven before, the agitation caused by stirring can improve the membrane flux through reducing the total resistance. To further study which part of the resistance is mainly influenced by the agitation, the total resistance distribution was studied in detail.

Table 3 Resistance distribution (R value in m^{-1} to be multiplied by 10^{13})

	R	R_m	R_p	R_c	R_i
continuous -stirring	3.59	1.60	1.44	0.45	0.10
non-stirring	10.28	1.60	7.93	0.43	0.32

R= total resistance; Rm= membrane resistance; Rp= polarization resistance; Rc= cake resistance; Ri= inner resistance.

In Table 3, the total resistance (R) is mainly made up of membrane resistance (R_m), polarization resistance (R_p), cake resistance (R_c) and inner resistance (R_i). The R_c and R_i occupied a small proportion so that they can be ignored in the following discussion. It is obvious that the total resistance mainly lies on the R_p and R_m, which are also the reason of membrane fouling. The total resistance in the non-stirring system is much higher than that in the continuous-stirring system, reaching $10.28*10^{13}$ m^{-1}, 2.86 times the latter one. Considering there is no obvious difference between R_m in both systems, the R_p became the most important factor in the total resistance. Therefore, the difference in the total resistance between the non-stirring and continuous-stirring system lies in the difference in the polarization resistance of the two systems, R_p of continuous-stirring ($1.44*10^{13}$ m^{-1}) is 5.51 times smaller than that of non-stirring ($7.93*10^{13}$ m^{-1}). Besides, according to Xue et al. [19], in most cases, polarization resistance occupies the highest part of total resistance, over 60%. Based on the results and former studies, agitation can inhibit the polarization to reduce the total resistance and improve the membrane flux.

B. Influence of agitation on the removal of COD and NH_4^+-N

It is reported that the total COD and NH_4^+-N removal were influenced by operational factors such as membrane filtration and biodegradation. To study the effect of additional continuous stirring on COD and NH_4^+-N removal, the contribution of both membrane filtration and biodegradation were investigated.

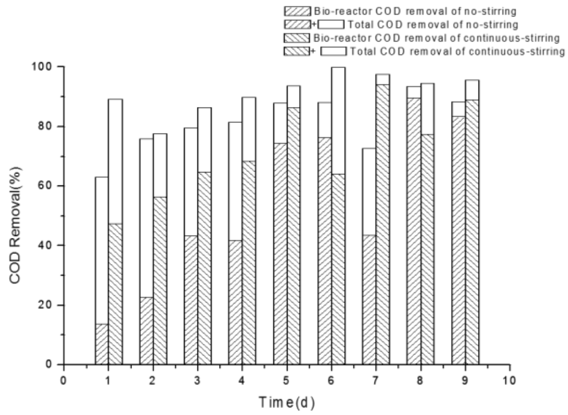

Fig.3 COD removal under non-stirring and continuous –stirring

As shown in Fig.3, biodegradation (striped bars) and microfiltration (blank bars) simultaneously contributed to the total COD removal. During the start-up period, the COD removal caused by biodegradation was relatively low in both groups. The reason may rely on the low biodegrability in the initial period. In this period, the COD was mainly removed by the microfiltration which concentrated the organic matters in the reactor. However, under continuous stirring, the COD removal caused by biodegradation increased from 50% to 85% in the following 5 days and was maintained at over 85% in the mid-to-late stage, while the non-stirring group only reached over 70% at the same time. Under continuous stirring, the COD of the effluent was 40-60 mg/L when the COD of influent was 800-1000 mg/L. It is clear that the COD removal efficiency significantly increased under continuous–stirring.

Continuous stirring has some positive effects on the COD removal. The effects can be listed as follow:

1) The continuous-stirring improved the biodegradation through improving the contact area between microorganism and organic matters in the bioreactor, leading to the enhancement of mass transfer process. The COD of the food wastewater can hence be degraded more readily and completely.

2) The continuous-stirring improved the membrane flux. A better degradation caused less organic load for the membrane to filtrate, further increased the capacity of COD removal. Besides, a lower organic load is also good for improving the membrane permeability.

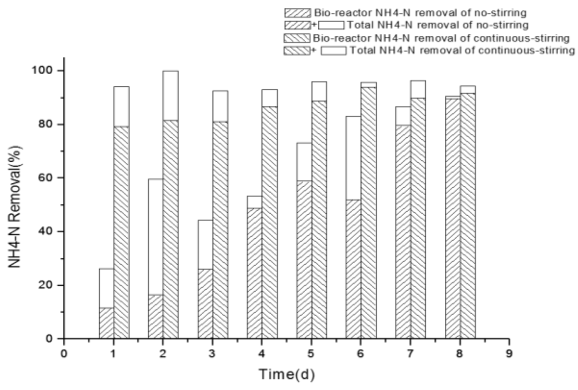

Fig.4 Ammonia nitrogen removal under non-stirring and continuous -stirring

Since the removal of NH_4^+-N and COD is influenced by some common factors, a similar conclusion can be drawn for the NH_4^+-N removal, although it was slightly different from the COD removal. Fig.4 shows that under continuous-stirring, the NH_4^+-N removal caused by biodegradation was at a high level for the whole period, between 80% and 85%. The total NH_4^+-N removal exceeded 90%. In contrast, the non-stirring group has a lower NH_4^+-N removal (around 85%) and it increased slowly with time. That means agitation has a stronger and more direct effect on the NH_4^+-N removal than on the COD removal. Under continuous-stirring, the ammonia nitrogen concentration of the effluent was below 1 mg/L. It is showed that the ammonia nitrogen removal efficiency under continuous–stirring (over 90%) was higher than that under non-stirring (around 85%).

C. *Morphology of aerobic granular sludge*

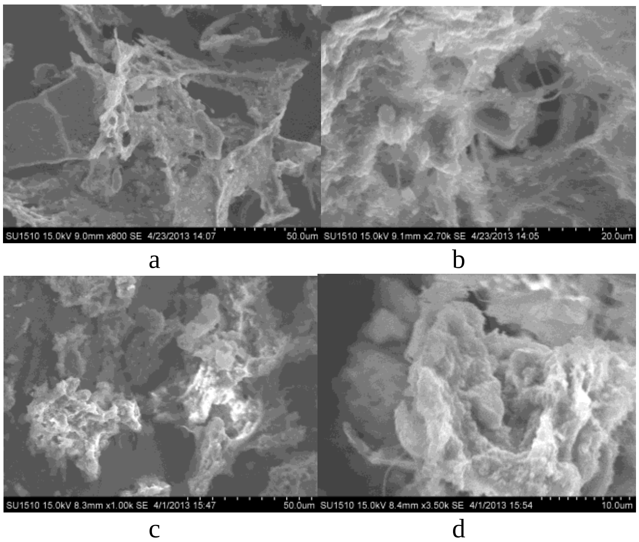

Fig.5 Comparison of morphology of aerobic granular sludge under continuous –stirring (a,b) and non-stirring (c,d) after 30 days' operation

The SEM images were obtained with scanning electron microscopy (SEM) and shown in Fig. 5. The granular sludge was obtained from the bioreactor. Chen [20] have proven that the shear force has a direct influence on the formation of granular sludge. In this study, Fig.5 indicates that under continuous-stirring, because of the high stress of the shear force, most of the aerobic granular sludge was of flaky structure (Fig.5 (a)). On the surface of the flaky structure, the bubbles more easily entered the granular structure under such a higher shear force, thus the tunnel structure formed and the amount of binding sites for microorganisms increased (Fig.5 (b)). On the contrary, under non-stirring, for lack of shear force, most of the aerobic sludge granules were of irregular block structure (Fig.5 (c)). Mostly the surface of granule showed ravine and the tunnel structure is hard to form (Fig.5 (d)), leading to less microorganisms binding sites compare to the structure under continuous-stirring. The different structures under the two different agitation conditions could be a reason to explain why the COD and NH_4^+-N removal efficiency under continuous–stirring is higher than that under the non-stirring.

IV. CONCLUSION

This work studied the capacity and biodegradation problems caused by membrane fouling, through applying an immersed membrane bioreactor with additional continuous stirring as an enhancement of agitation. The removal rate of COD and NH_4-N can reach 93% and 90%, respectively. The membrane performance was simultaneously improved by continuous–stirring. The resistance caused by the concentration polarization decreased 2.86 times under continuous–stirring, and the membrane flux reached 14.68 L/h*m^2, 3 times the control value. It is proven the bioreactor contributes most in the degradation of the organic matter. The membrane concentrates the organic matter and activated sludge to maintain them at a high concentration. Because of the flaky structure formed under continuous–stirring, the binding chance for microorganisms could increase. This work could provide an effective technique for the control of biofouling in MBR.

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