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Effect of C/N ratio and aeration rate on performance of internal cycle MBR with synthetic wastewater

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ABSTRACT

Effect of C/N ratio and aeration rate on performance of continuously operated internal circulation membrane bioreactor (ICMBR) was investigated thoroughly using synthetic domestic wastewater. The results show that COD and total nitrogen (TN) removal efficiencies were improved with the increase of C/N ratio under certain conditions ($T = 25$ ℃, MLSS = 10 g/L, aeration rate was 0.15 m^3/h, influent NH_3-N concentration was 20 mg/L, influent NO_3^--N concentration was 100 mg/L, and HRT = 12 h). Meanwhile, a steady ammonia nitrogen (AN) removal rate (~80%) was obtained when C/N ratio increased from 2:1 to 6:1. Furthermore, when C/N ratio was 6:1 and aeration rate was 0.15 m^3/h, average removal rates of COD, AN, and TN reached 98.5, 97.4, and 52.6%, respectively. Additionally, the improvement in activity of denitrifying bacteria could increase TN removal rate at a lower aeration rate. Under the optimal operation parameters (C/N ratio of 6:1 and aeration rate of 0.05 m^3/h), the high average removal efficiencies of COD (96.0%), AN (96.4%), and TN (81.0%) were obtained by ICMBR. Finally, the membrane fouling was unobvious during this experiment process.

Keywords: Internal cycle MBR; C/N ratio; Aeration rate; Total nitrogen

1. Introduction

The membrane bioreactor (MBR) is a kind of sewage and industrial wastewater treatment process used widely in recent years, which combines biological degradation process with membrane separation. Interception of the membrane can keep high sludge concentration in bioreactor and gather slow-growth nitrifying bacteria to facilitate nitrification [1]. Khan et al. [2] and Prado et al. [3] reported that MBR system possessed good and stable capacity for COD and ammonia nitrogen (AN) removal. MBR also has the advantages of short hydraulic retention time (HRT), strong shock load resistance ability, and small space occupation [4–6], which will promote the research on MBR process continuously [7]. However, the conventional MBR has a low capacity for total nitrogen (TN) removal [8]. Moreover, the membrane should be cleaned or changed because of membrane fouling, which limits development and application of MBR further [9].

Internal circulation membrane bioreactor (ICMBR) design is based upon the successful utilization of internal circulation anaerobic reactor. ICMBR have hollow fiber membrane module and perforated pipe sparger installed in the chamber. Circulation of mixed liquid in ICMBR is realized by aeration, and it can

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form the aerobic zone through aeration in upflow area and form a relative anoxic zone (not an absolute anoxic zone) in down flow area. Therefore, ICMBR has many advantages compared with conventional MBR, such as relatively high COD and TN removal efficiency, and the aeration rate can be reduced at the same time, which overcomes the problem that it is difficult for conventional MBR to control DO so as to decreasing operating cost. In addition, it is easy to realize that automated management [10]. Li et al. [11] used internal-loop airlift MBR (a form of ICMBR) for treating synthetic domestic wastewater, and they found that the average TN removals of the internal-loop airlift MBR (63.1%) were always higher than that with continuous stirred tank reactor (CSTR) configuration (18.7%). Therefore, ICMBR has a better application prospect than conventional MBR for domestic wastewater treatment.

The simultaneous nitrification and denitrification (SND) process can be achieved in a single ICMBR, which will improve TN removal performance in conventional MBR. Performance of the SND process is affected by many factors, such as reactor structure and size, sludge concentration, aeration rate, F/M ratio, C/N ratio, pH, and so on [12,13]. Among the factors, C/N ratio and aeration rate are the parameters being investigated usually for the SND, because the aeration rate is easy to handle in a full-scale wastewater treatment plant and C/N ratio is an inherent character of the wastewater affecting performance of the SND. Zhang et al. [14] reported that maximal removal efficiency for AN and TN was 90.6 and 82.2% in a Modified University of Cape Town-MBR (MUCT-MBR) system, when C/N ratio of the wastewater was around 4.0. In addition, Zhang et al. [15] reported that COD and AN removal efficiency was around 90% in a Sequencing Biofilm Batch Reactor (SBBR) system when aeration rate was 0.025–0.070 m^3/h. They also found that TN removal rate reached 73.3% when the aeration rate was 0.025 m^3/h. Therefore, C/N ratio and aeration rate will be suitable factors to promote the SND performance if they are handled appropriately.

Although ICMBR has been proved to have good performance of pollutant removal from different kinds of wastewater [16,17], there are few studies on the effect of C/N ratio and aeration rate on pollutant and TN removal performance in ICMBR, especially for the collaborative effect of the two parameters on a single ICMBR. In order to optimize the operating conditions of ICMBR and to provide some advices for engineering application of ICMBR technology, the effect of C/N ratio and aeration rate on the performance of ICMBR reactor was investigated thoroughly in this study.

2. Materials and methods

2.1. Set up and operation of ICMBR system

Fig. 1 shows the schematic diagram of the ICMBR system used in this study, which is mainly composed of a bioreactor and a membrane module. The bioreactor was made up of organic glass (poly(methyl methacrylate), PMMA) which had an inner barrel (14 cm in internal diameter and 18 cm in height) inside, the outer barrel was 20 cm in the internal diameter and 25 cm in height, and the total volume was around 6.2 L. The working volume of reactor was around 5.0 L. The membrane module was polyvinylidene fluoride (PVDF) micro-filtration membrane (type MOF-1b) produced by Tianjin Motimo Membrane Technology Co. Ltd., which formed a negative pressure and discharge water through the suction of the effluent peristaltic pump. Pore size of the membrane was 0.2 μm. Filtration area of the membrane was 0.5 m^2. HRT of the reactor was kept at around 12 h throughout the experiments. Temperature of the reactor was 25 ± 2℃ throughout the experiments. Intermittent manner operation of the membrane module was adopted for the effluent of ICMBR, and the ratio of suction and stop was 10:2. Inner barrel formed the aerobic zone through aeration and DO concentration of the outer barrel was relatively low, which created a suitable condition for the SND process.

Sludge for inoculum was taken from sludge return pipe (aeration tank) in a sewage treatment plant

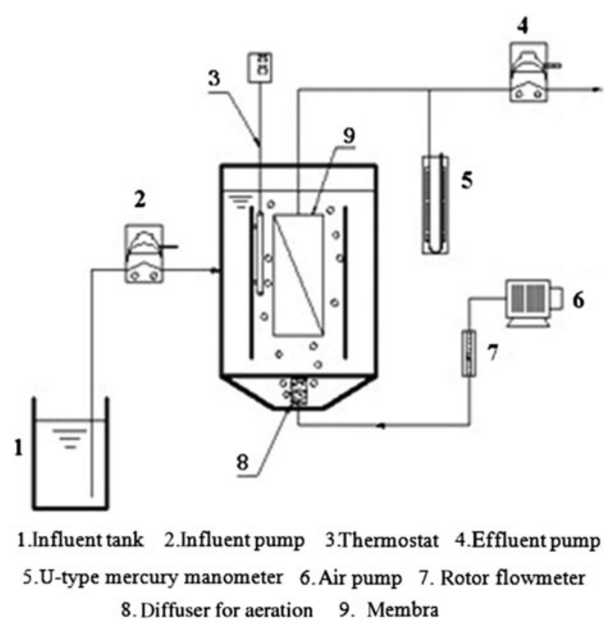

Fig. 1. ICMBR system.

treating domestic wastewater (actual operation) in Chengdu with MLSS of around 13 g/L. 0.5 L of activated sludge mixed liquor was added into the reactor, after diluting it 4 times with the synthetic wastewater which was composed of glucose, NH_4Cl, KH_2PO_4, and trace elements. Influent COD of startup enhanced from 360 to 810 mg/L, effluent COD of startup varied from 10 to 85 mg/L, and the COD removal rate was always above 85%. The experiments began after successful startup (about one month) when the sludge had good setting property and was at the stable phase.

2.2. Experimental wastewater

The domestic wastewater was simulated by tap water (15 L/d), glucose (112, 224, 336, 449, 561, and 673 mg/L at C/N varied from 1:1 to 6:1, respectively), NH_4Cl (76 mg/L), KNO_3 (163 mg/L), and KH_2PO_4 (18 mg/L) which were used as carbon, nitrogen, and phosphorous sources, respectively, supplemented by trace nutrient elements such as $CaCl_2$ (2 mg/L), $FeSO_4{\cdot}7H_2O$ (1 mg/L), $MnCl_2{\cdot}4H_2O$ (1 mg/L), and $MgSO_4{\cdot}7H_2O$ (3 mg/L). The pH of the synthetic wastewater was adjusted at 7.8 ± 0.2 with $NaHCO_3$.

2.3. Analytical methods

During continuous operation period, samples were taken from influent and effluent of ICMBR once a day and were analyzed immediately after the filtration by 0.45 μm filter paper. Soluble COD, AN, NO_3^--N, and NO_2^--N were measured in accordance with National Environmental Protection Agency of China [18]. TN was calculated by addition of AN, NO_3^--N, and NO_2^--N. The pH was monitored by pH electrode. Differential pressure gauge was used to measure the transmembrane pressure (TMP) of the membrane module.

3. Results and discussion

3.1. C/N ratio

AN and NO_3^--N concentrations were controlled at around 20 and 100 mg/L in the influent of ICMBR. C/N ratios were 2:1, 3:1, 4:1; 5:1, and 6:1, respectively. Aeration rate of the reactor was controlled at 0.15 m^3/h stably with the change of C/N ratio, and the corresponding MLSS at different C/N ratio was 10.2, 14.3, 8.7, 9.9, and 11.9 g/L, respectively. Emission amount of sludge was 0.1L/d (when C/N ratio was 3:1). All points in the figure below were obtained based on 7 d pseudo-steady data under each operation condition.

3.1.1. Effect of C/N ratio on COD removal

COD removal performance of bioreactor in treating different kinds of wastewater is very important for the measurement of the bioreactor performance because discharge standard for COD is strict in China. Therefore, COD removal performance of ICMBR with the variation of C/N ratio is shown in Fig. 2. It could be seen from Fig. 2 that COD measured values were all slightly lower than the corresponding theoretical values, which is probably caused by the moisture absorption of the industrial glucose used for synthesizing the simulated wastewater. C/N ratio rose from 2:1 to 6:1 gradually, and corresponding influent COD concentration increased from 219 to 672 mg/L.

Average effluent COD concentration was always below 50 mg/L (The first grade A standards of Discharge Standard of Pollutants for Municipal Wastewater Treatment Plant in China) during the experiments, and COD removal rate enhanced from 84.7 to 98.5%. Therefore, COD removal performance of ICMBR was affected little by the increase of C/N ratio at the given range. Khan et al. [19] also reported that the effect of C/N ratio on COD removal was unobvious and COD removal rate in MBR system was above 95% with organic loading rate of 3 kg/m^3/d. The results suggest that COD removal rate was improved with the increase in C/N ratio, which might be due to the enhancement of heterotrophic bacteria proliferation and denitrifying bacteria activity with the improvement of COD concentration.

3.1.2. Effect of C/N ratio on AN removal

AN is another key control factor for the discharge of wastewater in China, which must be handled well in treating AN-containing wastewater.

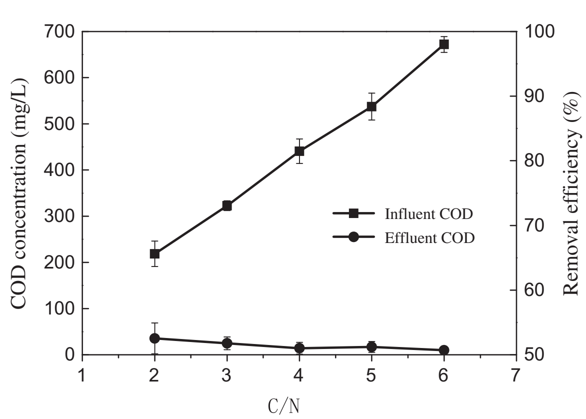

Fig. 2. COD removal performance of ICMBR under different C/N ratio.

AN removal performance in ICMBR with the improvement in C/N ratio is shown in Fig. 3. The measured AN concentration was also slightly lower than the theoretical value, but the impact on the results was insignificant. Average influent AN concentration was 21 mg/L in the variation range of C/N ratio from 2:1 to 6:1.

AN concentration in the effluent of ICMBR was always lower than 2 mg/L, and corresponding AN removal rate was maintained above 92%, with the highest value of 99.6% and the mean value of 97.4%. The results showed that the AN removal in ICMBR system was affected little by changing the C/N ratio in the given level, which was similar with COD removal. AN removal was achieved mainly by the activity of nitrifying bacteria and the interception of the membrane could make the nitrifying bacteria multiply, while the interception effect on the AN was very small [20]. At the same time, heterotrophic bacteria consumed part of nitrogen for reproducing themselves to remove certain AN. Kumar et al. [21] also found that when C/N ratio rose from 0.9 to 9.3, the AN removal in MBR system was relatively stable and removal efficiency was always maintained above 80%, which agreed with the results shown in the experiments.

3.1.3. Effect of C/N ratio on TN removal

TN is one of the important indexes of water quality which can induce the extent of water eutrophication. TN removal performance in ICMBR with the improvement of C/N ratio is shown in Fig. 4. The TN concentration is calculated by the sum of AN, nitrate, and nitrite nitrogen concentration. The TN concentration in the effluent rose with the increase of C/N ratio, and it reached its maximum value of 116 mg/L when C/N ratio was 2:1.

With the continuous increase in C/N ratio, the TN concentration in the effluent presented obvious linear decreasing tendency ($y = -14.9x + 146.1$, $R^2 = 0.994$) within the given range. Correspondingly, the TN removal rate was the lowest (4.5%) when C/N ratio was 2:1, and then it began to increase and reached 52.6% when C/N ratio was 6:1. Thus, it is clear that the denitrification process was obviously affected by C/N ratio. Since there were not enough electron donors (provision of organic matters) under the low C/N ration conditions [22], the denitrification process would be inhibited seriously. Additionally, since the membrane interception and sufficient carbon sources would be achieved by increasing C/N ratio, the denitrifying bacteria could reproduce themselves gradually which results in the gradual increase in the TN removal rate. Zhao et al. [23] also found the SND effect was the best when C/N ratio was increased from 6:1 to 8:1, which was similar to the results obtained in this study. It could be concluded that when C/N ratio was 6:1, the removal efficiencies of COD, AN, and TN were above 95, 95, and 50%, respectively.

3.2. Aeration rate

COD concentration of the influent was controlled at around 680 mg/L and aeration rate was 0.15 m^3/h (lasted 7 d), 0.10 m^3/h (lasted 7 d), and 0.05 m^3/h (lasted 7 d), respectively.

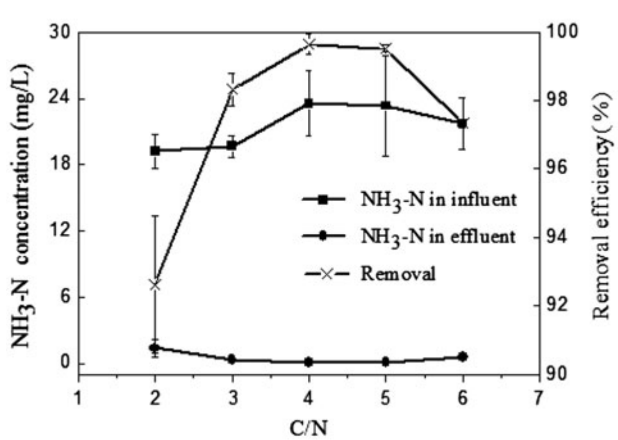

Fig. 3. AN removal performance of ICMBR under different C/N ratio.

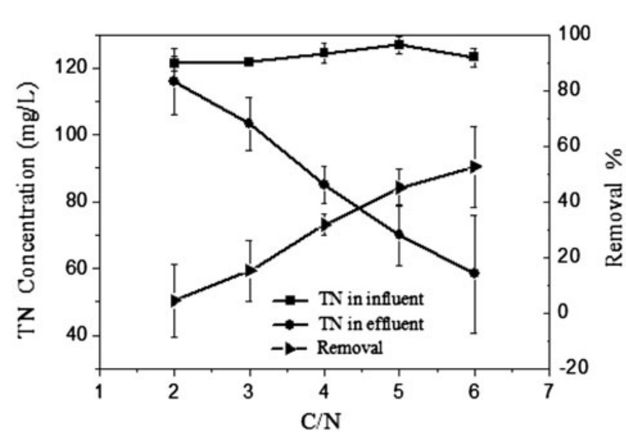

Fig. 4. TN removal performance of ICMBR under different C/N ratio.

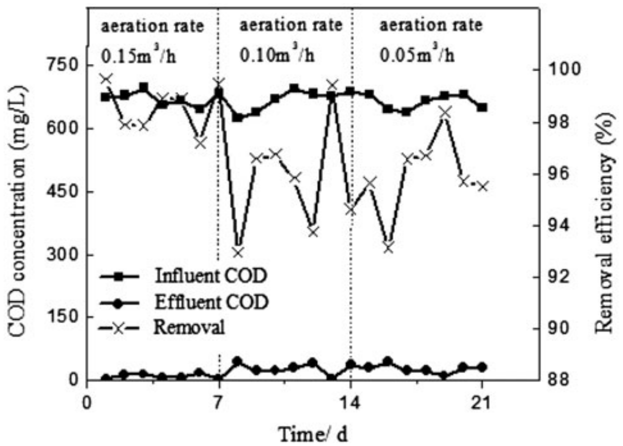

Fig. 5. COD removal performance of ICMBR under different aeration ratio.

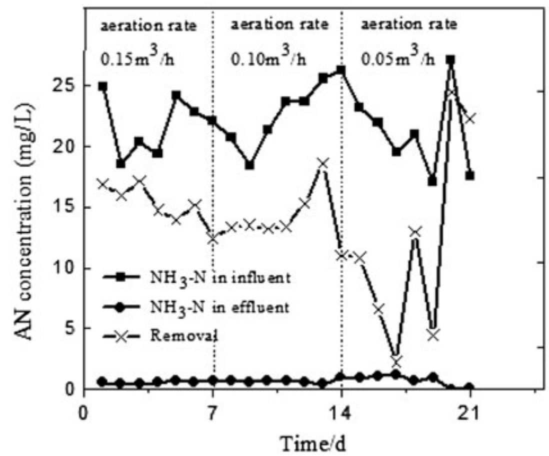

Fig. 6. AN removal performance of ICMBR under different aeration ratio.

3.2.1. Effect of aeration rate on COD removal

Aerobic bacteria need oxygen for the removal of COD, so the aeration rate is a key parameter for the removal of COD. Reduced aeration rate can reduce operating cost. COD removal performance in ICMBR with the decrease of aeration ratio is shown in Fig. 5.

Average COD concentration in the effluent was 9.9 mg/L and average removal rate of COD was 98.5%, when the aeration rate was 0.15 m^3/h. When the aeration rate was 0.1 m^3/h, the removal rate of COD decreased obviously at first, being only 93%. Then, it rose to 95.70% on average, and the average COD concentration in the effluent was 28.6 mg/L. When the aeration rate was 0.05 m^3/h, the change of COD removal rate was similar to the above-mentioned changes, being 96.0% on average and the average COD concentration in the effluent was 26.8 mg/L. This showed that ICMBR system could keep relatively high COD removal efficiency when the aeration rate was 0.05 m^3/h, which was probably because oxygen was not the only electron acceptor, but nitrate and nitrite could also serve as the electron acceptors for decomposing organic matters. In addition, MBR could stably remove 5–15% COD through the interception of the membrane [24]. It also indicated that ICMBR system can be operated at relatively low aeration rate and decrease the operating cost under the premise of ensuring low COD concentration in the effluent wastewater.

3.2.2. Effect of aeration rate on AN removal

Nitrification is an important step for AN removal and nitrifying bacteria involved in this reaction is aerobic. Therefore, aeration rate is also a key parameter for the removal of AN. AN removal performance in ICMBR with the decrease in aeration ratio is shown in Fig. 6.

Average AN concentration in the effluent was 0.6 mg/L and average removal rate was 97.4% when the aeration rate was 0.15 m^3/h. When the aeration rate was 0.10 m^3/h, average AN concentration in the effluent was 0.7 mg/L and average removal rate was 97%. When the aeration rate was 0.05 m^3/h, average AN concentration in the effluent was 0.7 mg/L and average removal rate was 96.4%. With the decrease in the aeration rate from 0.15 to 0.05 m^3/h, the decrease in average AN removal rate could be ignored because DO concentration in the water could meet the normal growth and activity demand of nitrifying bacteria under the given aeration rate (0.15–0.05 m^3/h).

Usually, nitrification will be weakened with the decrease in the aeration rate. The activity of nitrifying bacteria was greatly reduced and the nitrification did not go smoothly when the aeration rate was too low. Zhao et al. [23] found that when aeration rate was 0.35 m^3/h in MBR system which had 150 L in volume, the nitrification reaction was affected because of low aeration rate and the removal efficiency of AN was just 77%. The operating cost could be reduced by decreasing aeration rate in a certain range (from 0.15 to 0.05 m^3/h in this experiment) under the premise of ensuring low AN concentration in the effluent wastewater.

3.2.3. Effect of aeration rate on TN removal

Since improvement of denitrification is an important step for TN removal and denitrifying bacteria involved in this reaction is anoxic. The aeration rate

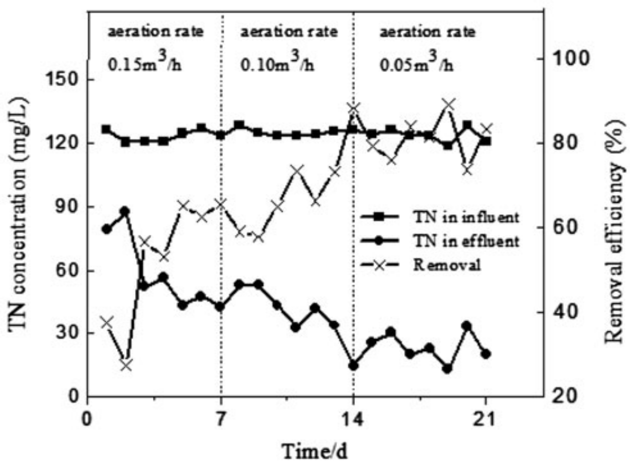

Fig. 7. TN removal performance of ICMBR under different aeration ratio.

will affect removal performance of TN. TN removal performance in ICMBR with the decrease in aeration ratio is shown in Fig. 7.

Average TN concentration in the effluent was 58.4 mg/L and average removal rate was 52.6%, when the aeration rate was 0.15 m^3/h. When the aeration rate was 0.10 m^3/h, average TN concentration in the effluent was 38.9 mg/L and average removal rate was 69%. When the aeration rate was 0.05 m^3/h, average TN concentration in the effluent was 23.7 mg/L and average removal rate was 81.0%. With the decrease in the aeration rate from 0.15 to 0.05 m^3/h, average TN concentration in the effluent decreased gradually and average removal rate rose gradually. Denitrification was the main way to achieve TN removal in ICMBR. At the same time, heterotrophic bacteria consumed part of nitrogen for reproducing themselves to remove some of TN. It was known that denitrification was affected by the aeration rate and higher aeration rate would destroy anoxic condition of ICMBR. It was more difficult to form anoxia environment under higher aeration rate in ICMBR reactor. Therefore, TN concentration in the effluent was much higher and the removal rate was lower under higher aeration rate. It was clear that decrease in aeration rate facilitated the removal of TN, but too low aeration rate would affect AN removal efficiency. The aeration rate should be reduced as much as possible under the premise of meeting the demand for activity and growth of nitrifying bacteria. It could not only improve the activity of denitrifying bacteria to increase TN removal rate, but also reduce the energy consumption and operating cost. This also verifies the opinions made by He et al. [25].

In summary, the removal efficiency of TN should also be taken into consideration for a suitable aeration rate. SND effect was best when the aeration rate was 0.05 m^3/h in the experiments.

3.3. Membrane fouling

Although the capital and energy cost of ICMBR has been reduced, membrane fouling which is a major problem for MBRs exists as a black box due to the complex nature of fouling layers [26]. The gel layer formed during the operation of MBR owing to the concentration polarization causes membrane fouling and affects the normal operation of the reactor [27]. Therefore, it is very important for monitoring and analysing membrane fouling. Membrane fouling is analyzed by monitoring the TMP parameter and scanning electron microscopy (SEM). The photos of SEM on membrane fiber are shown in Fig. 8.

The TMP of MBR did not change obviously (about 35 mm Hg) during the operating process, which showed that MBR did not have serious membrane fouling during the period. The membrane flux remained steady at about 2.5 L/(m^2 h) during the experiment. The outside surface of the membrane was covered with pol-

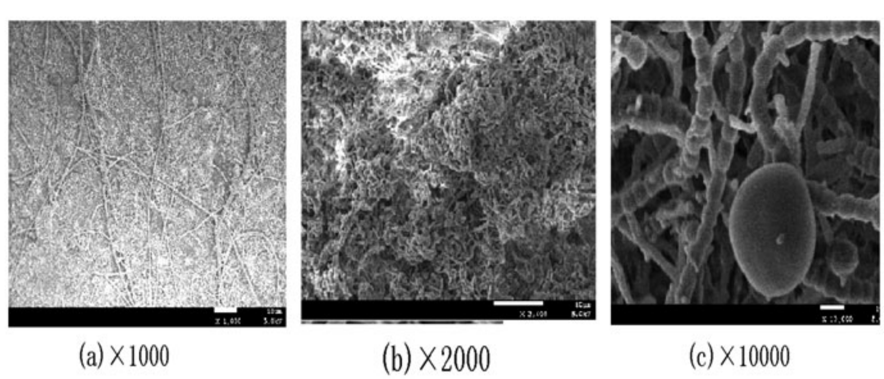

Fig. 8. Photos of SEM on membrane fiber.

luted materials, where micro-organisms stuck together and formed the gel layer. The micro-organisms attached on the gel layer mainly comprised of cocci, bacillus, and filamentous bacteria, which showed that the membrane fouling was mainly caused by the adhesion of mass micro-organisms on the membrane surface. But the porosity of the gel layer was relatively large, so it had little impact on the pressure. As the experiment influent wastewater was synthetic wastewater not actual wastewater, a new membrane was used for the experiment, so that the gel layer thickness was smaller and the membrane fouling was insignificant. Moreover, HRT of ICMBR reactor was much longer so that it could alleviate membrane fouling by reducing accumulation of soluble microbial products (SMP) and growth of biomass [28]. In addition, continuous influent and intermittent effluent was adopted in ICMBR reactor, which played an important role in alleviating membrane fouling [29].

4. Conclusions

By analyzing the effect of different C/N ratio on IC-MBR performance, it was known that AN removal was not affected by C/N ratio and removal rate was maintained above 92%. Removal of COD and TN was affected by C/N ratio, and COD removal rate rose with the increase in C/N ratio. TN removal rate decreased with the decrease in C/N ratio because of lack of sufficient electron donors at low C/N ratio, so denitrification effect was affected. However, the high C/N ratio required adding more carbon sources, which would increase operating cost. Therefore, a suitable C/N ratio was existent. It was known that the suitable C/N ratio was 6:1 at the given range in this experiment. ICMBR system could reduce the aeration rate and guarantee the effluent wastewater quality in certain range so as to reducing the operating cost. ICMBR reactor also could alleviate membrane fouling at a certain degree, reducing the operating cost as well.

Acknowledgments

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