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Simultaneous nitrogen and phosphor removal in an aerobic submerged membrane bioreactor

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Abstract: Simultaneous nitrification and denitrification (SND) effect and phosphor removal were investigated in a one-staged aerobic submerged membrane bioreactor on pilot-scale with mixed liquor suspended solids (MLSS) 19—20 g/L. The effects of DO concentration, sludge floc size distribution on SND were studied. Test results suggested that SND was successfully performed in the membrane bioreactor (MBR) and about 70% total nitrogen removal efficiency was achieved when DO concentration was set to 0.2—0.3 mg/L. The main mechanisms governing SND were the suitable sludge floc size and the low DO concentration which was caused by low oxygen transfer rate with such a high MLSS concentration in the MBR. In the meantime, phosphor removal was also studied with polymer ferric sulfate (PFS) addition and 14 mg/L dosage of PFS was proper for the MBR to remove phosphor. PFS addition also benefited the MBR operation owing to its reduction of extracellular polymer substances (EPS) of mixed liquor.

Keywords: membrane bioreactor (MBR); flat-sheet membrane; simultaneous nitrification and denitrifiction (SND); phosphor removal; wastewater treatment; oxygen transfer rate

Introduction

Submerged membrane bioreactors (MBRs) have been increasingly popular for the treatment of municipal and industrial wastewater in recent years (Howell *et al.*, 2004). The smaller footprint, higher quality effluent and better control over biological conditions (Thomas *et al.*, 2000; Le-Clech *et al.*, 2003) provide MBRs with significant benefits over other biological wastewater treatment processes. Furthermore, many investigators have put forward a series of methods to overcome membrane fouling which is one of the biggest barriers in the MBR application. These methods include hydrodynamic control by cross-flow (Cho and Fane, 2002), air scouring (Ueda *et al.*, 1997), MBR dimensional parameters (Liu *et al.*, 2000) and operation mode such as sub-critical flux operation (Field *et al.*, 1995; Wu *et al.*, 1999), intermittent suction mode (Ahn and Song, 2000) and so on. Undertaking these countermeasures enable MBRs to run smoothly and stably without extra chemical or physical cleaning.

Due to the introduction of stringent effluent standards, in particular for nutrients, total nitrogen and phosphor removal must be taken into account during MBR designing. As far as concerns nitrogen removal, MBRs are traditionally designed with a pre-denitrification stage (Buisson *et al.*, 1997), or with post-denitrification stage (Lesjean *et al.*, 2002), or as an intermittently aerated MBR (also named as sequencing aerobic/anoxic membrane bioreactors) (Yeom *et al.*, 1999; Ahn *et al.*, 2003), or even as an anaerobic/aerobic MBR described by Zhang and his coworkers (2005). The nitrogen removal theory of the combined MBRs as mentioned above is that under aerobic conditions ammonium is oxidized to nitrite or nitrate which is later changed into nitrogen under anoxic condition. Therefore, two separate reactors (or sequences in intermittent system) are required to provide two different environmental conditions. However, recent studies have revealed that the two steps could occur at the same time in one reactor (Pochana and Keller, 1999; Holman and Wareham, 2005). The process, which can eliminate a separate denitrification tank or period (in intermittently aerated processes), is termed simultaneous nitrification and denitrification (SND). If SND can be realized in one-staged aerobic MBRs, it might help to simplify the overall process design of MBRs dramatically. MBR process will become one of most promising technologies in wastewater treatment. Many researchers also have focused on phosphorus removal using either biological or chemical processes. Genz *et al.* (2004) removed phosphor from membrane filtrates by adsorption on activated aluminium oxide and granulated ferric hydroxide. Ahn *et al.* (2003) investigated phosphor removal effect by using a sequencing anoxic/anaerobic membrane bioreactor (SAM) process and found the phosphorus removal efficiency was about 93%. These studies focused on the nutrient removal and gained profound understanding of the characteristics of nitrogen and phosphor removal in anoxic/aerobic (or other combined forms) MBRs or related biological processes. However, few investigators have described the SND effect together with the characteristic of phosphor removal in one-staged aerobic MBRs.

The purpose of the present study is to investigate the SND effect, to discuss conditions and parameters governing SND efficiency, to study the phosphor

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removal by means of chemical precipitation in a one-staged submerged flat-sheet membrane bioreactor.

1 Materials and methods

1.1 Experimental setup

The experimental study was performed using two kinds of MBR: a pilot scale and a laboratory scale MBR.

The pilot-scale submerged membrane bioreactor (Fig.1), which was located in Quyang Domestic Wastewater Treatment Plant of Shanghai, has a total volume of 1.44 m^3 being 0.8 m $\times$0.9 m $\times$2.0 m (length$\times$width$\times$height). It is divided into a riser zone and two down-comer zones by two baffle sheets. Thirty flat sheet membrane modules are located in the riser. The membranes are made of polyether-sulfone (PES) membrane with 0.23 μm pore. The effective filtration area for each module is 0.7 m^2. Air is supplied through an axial perforated tube which is below the membrane modules in order to supply oxygen demanded by the microorganisms and induce a cross-flow velocity along the membrane surface. The water level was maintained constant by using control system to command the operation of influent valve and suction pump. The effluent flow rate and the trans-membrane pressure (TMP) were monitored with a water meter and a pressure gauge, respectively.

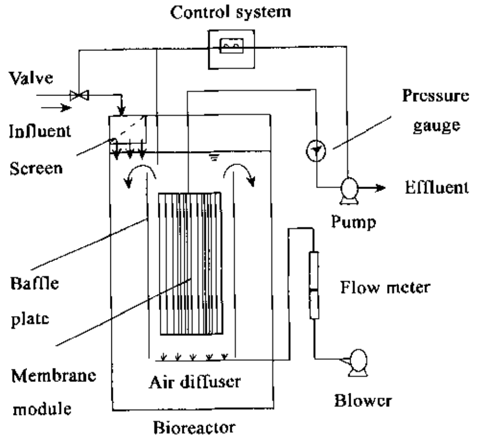

Fig.1 Schematic flow diagram of the MBR on pilot-scale

Domestic wastewater from the wastewater treatment plant was used in the study. After filtration through a stainless steel screen mesh of 0.9 mm, raw wastewater was supplied into the bioreactor and most of the organic pollutants contained in the raw wastewater were decomposed through biodegradation by the activated sludge. The membrane-filtered effluent was then intermittently obtained by suction using a pump connected to the modules.

The laboratory scale MBR (Fig.2) was geometrically similar to the pilot scale one. It was used to verify the effect of sludge floc size on SND and the influence of polymer ferric sulfate addition on membrane filtration operation. The sludge used in these tests was taken from the pilot-scale MBR. The MBR has a total volume 6.3 L in which a flat-sheet membrane with effective membrane area 0.098 m^2 was immersed.

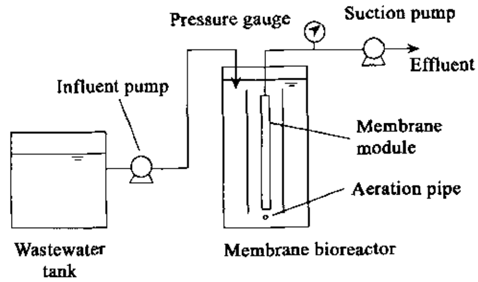

Fig.2 Schematic flow diagram of the MBR on lab-scale

1.2 Analytical items and methods

The analytical methods from Chinese NEPA standard methods (Chinese NEPA, 1997) were adopted for the measurements of COD, total phosphor, total nitrogen, nitrate, nitrite, ammonia and pH in the influent, supernatant liquor and membrane effluent, total suspended solids (SS), volatile suspended solids (VSS) in the bioreactor. The supernatant samples were obtained by filtrating the mixed liquor of the MBR through filter paper with mean pore size 0.45 μm. DO concentration was measured by a probe of dissolved oxygen meter.

1.3 Definition of steady-state period

The steady-state period was a period after 40 d when the biomass concentration was 19—20 g/L and comparatively stable.

1.4 Operating conditions

The pilot-scale MBR was operated in sub-critical flux zone during the run of the whole experiments. The cross-flow velocity around the membrane surfaces was maintained at 0.2—0.4 m/s by air scouring. A suction mode of 10 min on and 2 min off was also adopted to control membrane fouling. The hydraulic retention time (HRT), the sludge retention time (SRT) and the mixed liquor suspended solids (MLSS) of the bioreactor (during steady-state period) was 2.2 h, 40 d and 19—20 g/L respectively. The temperature of the experiments varied between 10—18℃.

2 Results and discussion

2.1 Filtration performance

Critical flux is a key concept in membrane filtration operation. Below the critical flux (also called as sub-critical flux) there is no severe fouling, however, above the critical flux (named as supra-critical flux) dramatic fouling will occur. The critical flux value was determined in the experimental unit using filtration operations based on gradually increasing the permeate flux value while monitoring changes in trans-membrane pressure (Defrance and Jaffrin, 1999). The critical flux of the experiment is

27—36 L/(m^2·h) and thus the MBR must be operated below 27 L/(m^2·h) which belongs to sub-critical flux. Membrane flux varied in the range of 21—27 L/(m^2·h) (mainly 25 L/(m^2·h)) and trans-membrane pressure increased from 4 kPa to 60 kPa during 120 d run. During the experiment, MBR was not cleaned physically or chemically except normal air scouring. Variations of trans-membrane pressure, flux are shown in Fig.3. On the day 96 of the operation, aeration failed for several hours while filtration was continued. As a result, cake layer accumulated more rapidly, thereby increasing TMP and filtration resistance. Accordingly, on the day 96, the MBR was aerated for 12 h without pump suction in order to recover the TMP. From the day 113, the fouling rate was higher compared with the previous run.

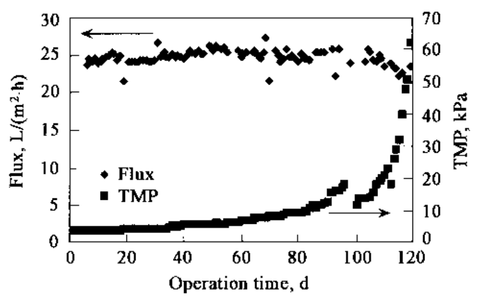

Fig.3 Changes of membrane flux and trans-membrane pressure (TMP) during 120 d run

2.2 SND efficiency and nitrogen mass balance

About 70% total nitrogen removal efficiency was achieved when DO concentration was set to 0.2—0.3 mg/L. The effluent TN concentration ranged from 13 to 19 mg/L with influent TN concentration 39—70 mg/L.

The nitrogen loss in the MBRs can be due to three aspects: ammonia stripping, assimilation and dissimilation by bacteria. Ammonia stripping can be neglected because ammonia at ambient temperature mostly lie in water as ammonium ion with the pH of the mixed liquor in the MBRs ranging from 6.9 to 7.4. Assimilation is responsible for 25% —30% of the nitrogen loss in biological wastewater treatment processes (Benefield and Clifford, 1980). In the meantime, the endogenous respiration can compensate a lot for the nitrogen loss in the MBR with long SRT 40 d (Hao *et al.*, 1997). Therefore, the dissimilation by bacteria mainly contributes to nitrogen loss in the MBRs.

2.3 Factors affecting SND

Three main factors influence the SND process, i. e. carbon source, DO concentration and floc size (Pochana *et al.*, 1999). Many investigators (Li and Lv, 2001; Xie *et al.*, 2004; Hao *et al.*, 1996) have studied the effect of carbon source on SND and the results are always the same—a higher C/N ratio can achieve a better SND efficiency. The pilot-scale MBR was used to treat the real domestic wastewater, the C/N ratio ranged from 6 to 8 all the time. Therefore, the main parameters affecting SND in the present study are owing to DO concentration and floc size.

2.3.1 DO concentration effect on SND

Three different DO concentrations were set to test their effects on SND. In Runs 1, 2 and 3, DO concentration was maintained at 0.4—0.5, 0.2—0.3 and 0.15—0.2 mg/L respectively. The variations of TN, ammonia, nitrate, nitrite in influent and effluent are shown in Fig.4a.

During the test, nitrite concentration always ranged irregularly but all below 0.6 mg/L (as shown in Fig.4d). Run 1 showed the nitrate of effluent was higher than Run 2 and Run 3 (Fig.4c). It indicated that nitrification was the prevalent reaction while denitrification was retarded under DO concentration 0.4—0.5 mg/L. In Run 3, under DO concentration 0.15—0.2 mg/L nitrification was restrained and ammonia of the effluent was higher accordingly (Fig.4b). When DO concentration was 0.2—0.3 mg/L in Run 2, ammonium and nitrate of the effluent were comparatively low and the SND effect was better comparing with the other two runs.

The DO concentration as mentioned above could be easily achieved in the MBR even at high intensity aeration. The main reason attributing to the low DO was that MLSS was much higher in the flat sheet membrane bioreactor compared with conventional activated sludge system. Therefore, in the MBR, the high intensity aeration not only maintained a proper cross-flow velocity around the membrane surface to keep MBR running stably, but also induced a suitable environment (low DO) for SND due to its high MLSS concentration.

In order to verify the relationship between high MLSS and oxygen transfer rate, the α value which is the ratio of oxygen transfer rate in mixed liquor and that in pure water as shown in Eq. (1) was tested.

$$\alpha = \frac{K_{La}}{K_{L0}} \quad (1)$$

Where K_{La} and K_{L0} are oxygen transfer rates in mixed liquor and pure water respectively (h^{-1}).

As shown in Fig.5, the α value decreased from 0.57 to 0.16 as MLSS increased from 4.5 to 20 g/L. So the result well explained that the DO concentration in the MBR with high MLSS could be maintained at a low level. Fig.5 also shows that the relationship between α value and MLSS accorded with α=0.8293 exp (-0.0849 MLSS), where MLSS was calculated by g/L.

2.3.2 Effect of floc size on SND

The underlying physical explanation is that a substantial anoxic mass fraction exists in the center of the biomass flocs resulting from an oxygen diffusion

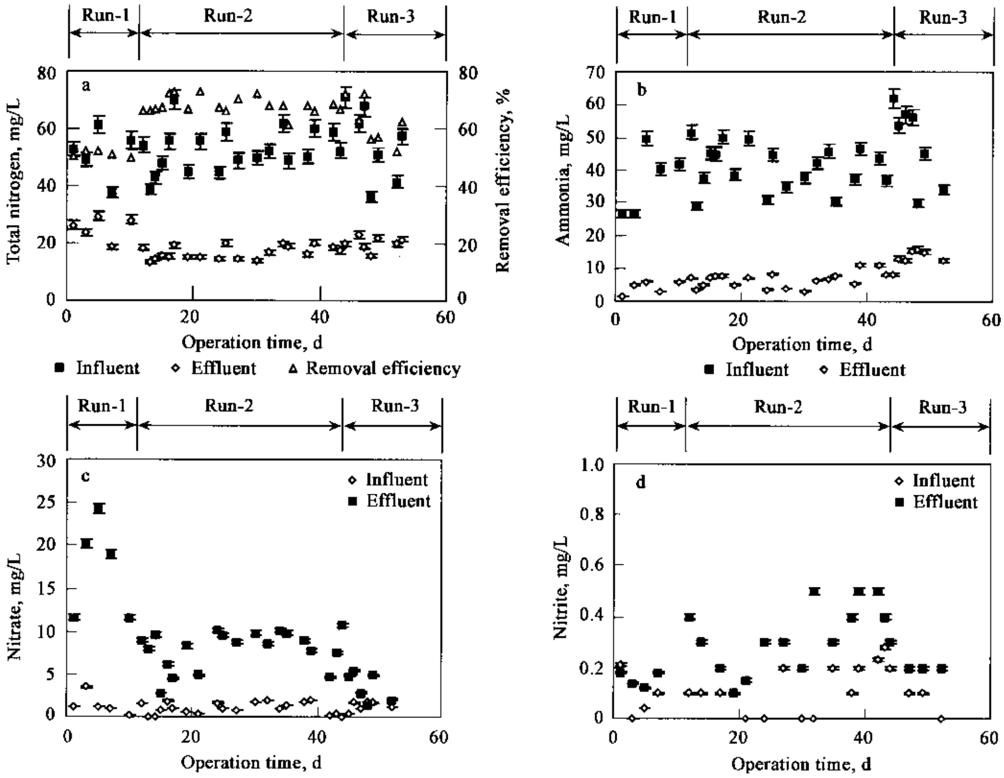

Fig.4 Variations of TN (a), ammonia (b); nitrate (c), nitrite (d) in influent and effluent

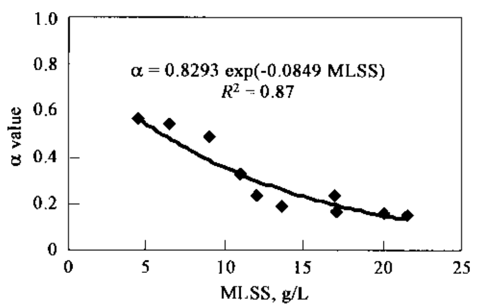

Fig.5 Relationship between α value and MLSS

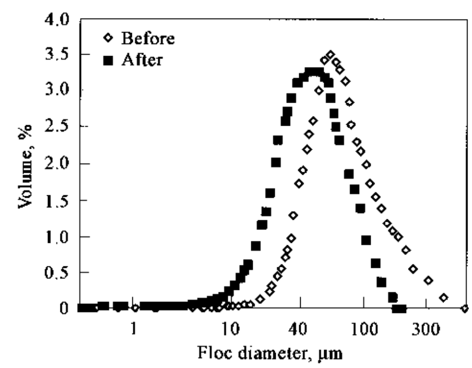

Fig.6 Floc size distribution before and after blending

limitation into the flocs. The median floc sizes as measured in the activated sludge of the MBR were 70—84.5 μm which was suitable for the SND according to the research of Pochana *et al.* (1999). The experiment to determine floc size effect was conducted in the lab-scale MBR. The first run was performed by using primitive sludge of the pilot-scale MBR with the usual floc size (median 70 μm) while the second run was operate with smaller flocs (median 40 μm). The sludge for the second experiment was obtained by blending the sample sludge taken from the pilot-scale MBR (2000 r/mim) for 15 min. The floc size distribution of the two experiments is shown in Fig.6.

The influent concentration of these two experiments was 60 mg/L ammonia, 600 mg/L COD which was synthesized by ammonium chloride, glucose and other mineral substances. 2.5 L sludge sample and 2.5 L synthetic wastewater were put into the lab-scale MBR. During the experiments, the DO concentration was maintained at 0.4—1.0 mg/L. The ammonium, nitrate and nitrite were measured in the two experiments in order to analyze the nitrification rate and the SND effect as shown in Fig.7.

The nitrification rate (within 140 min) of the two sludge samples were 0.029 mgNH_4-N/(gVSS·min) (before blending) and 0.030 mgNH_4-N/(gVSS·min) (after blending) and that suggested the blending did not affect the viability of the biomass. However, the effluent average nitrate of the MBR was 18.9 mg/L (after blending) and 11.2 mg/L (before blending). Effluent nitrite of the both experiments were the same (ranging from 0.1—0.6 mg/L) and so were the ammonia of them (varying from 3.8—5.0 mg/L).

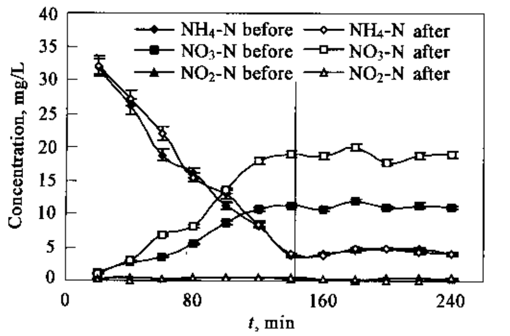

Fig.7 Variations of ammonia, nitrate and nitrite during the test

Therefore, the sludge sample before blending performed well concerning the SND effect.

2.4 TP removal

Polymer ferric sulfate (PFS) was adopted to remove phosphor in the MBR. Before the phosphorus removal experiment was carried out, a series of tests in the lab-scale MBR had been conducted to verify the effect of coagulant addition on membrane filtration operation. Test results suggested the membrane fouling rate was reduced after adding the coagulant into the MBR (data not shown). The extracellular polymer substances (EPS) concentration, which was regarded as a key factor influencing membrane fouling (Chang and Lee, 1998), was also measured before and after PFS addition. The EPS concentration before and after the coagulant addition (dosage 10 mgFe/L (influent) measured by total ferric ion) was 228.8 mgVS/g MLSS and 199.5 mgVS/g MLSS respectively. The test results as mentioned above indicated that PFS addition was also a positive measure to keep MBRs running smoothly.

The PFS dosage measured by total ferric ions during Runs 1, 2, 3, 4 was 24, 0, 10 and 14 mg/L, respectively. Total phosphor concentration in influent, effluent and permeate is shown in Fig.8. TP in effluent with no PFS addition ranged from 1.1 to 3.8 mg/L as influent TP concentration was fluctuated between 3.8—10.6 mg/L. The effects of PFS dosage 24 mg/L (Run 1) and 14 mg/L (Run 4) on TP removal were just

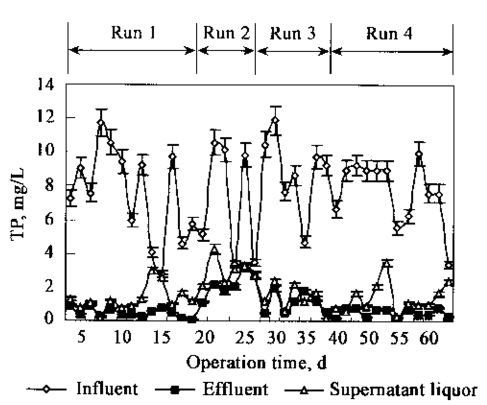

Fig.8 Variations of TP in the MBR with different PFS addition

the same and the effluent TP all ranged from 0.1 to 0.9 mg/L. However, Run 3 with 10 mg/L PFS addition performed worse with effluent TP 0.5—2.0 mg/L. Therefore, 14 mg/L PFS dosage was recommended as a proper dosage concerning TP removal. It was also observed that the effluent TP was always lower than that of supernatant liquor, which suggested that the membrane also contributed to the TP removal due to its fine pores.

3 Discussion

In the MBR, the low DO environment for SND was mainly due to the high MLSS concentration. The high MLSS concentration in turn affected oxygen transfer rate. The main mechanisms can be explained theoretically as follows.

In gas-liquid reactors, mass transfer rate can be expressed as Eq. (2) (Jiang, 1989):

$$\frac{K_{La}D_T^2}{D_L}=0.6\left(\frac{\mu_L}{\rho_L D_L}\right)^{0.5}\left(\frac{gD_T^2\rho_L}{\sigma}\right)^{0.62}\left(\frac{gD_T^2\rho_L^2}{\mu_L^2}\right)^{0.31}\varepsilon_G^{1.1} \quad (2)$$

Where D_T is the diameter of the reactor(m); D_L is the diffusion coefficient(m^2/s); σ is the surface tension (N/m); ρ_L is the liquor density (kg/m^3); μ_L is the liquor viscosity (Pa·s); ε_G is the fractional gas hold-up value (%); g is the gravitational acceleration velocity (m/s^2). D_L can also be written as the function of other related factors as shown in Eq. (3).

$$D_L=\frac{K_B T}{4\pi\mu_L R_0} \quad (3)$$

Where K_B is Boltzmann constant; T is the absolute temperature (K); R_0 is the average radius of sludge particles (m).

ε_G is directly proportional with other factors as described in Eq. (4) (Zhang, 1985).

$$\varepsilon_G\propto C\left(\frac{D_T^2\rho_L g}{\sigma}\right)^{1/8}\left(\frac{D_T^3\rho_L^2 g}{\mu_L^2}\right)^{1/12}\frac{\mu_{OG}}{(D_T g)^{1/2}} \quad (4)$$

Where C is a constant related to setup structure, μ_{OG} is the apparent gas velocity (m/s). Combining Eqs. (2), (3) and (4), K_{La} can be represented as follows:

$$K_{La}\propto 0.17\frac{C^{1.1}K_B^{0.5}T^{0.5}\mu_{OG}^{1.1}g^{0.89}\rho_L^{1.1}}{R_0^{0.5}\sigma^{0.76}\mu_L^{0.8}D_T^{0.14}} \quad (5)$$

On the right side of Eq.(5), K_B and g are constant parameters. In a certain MBR setup, C, D_T and μ_{OG} (at a certain aeration intensity) are invariable. With the assumption that sludge floc size keeps the same, Eq. (5) can be rewritten as Eq.(6).

$$K_{La}\propto\frac{T^{0.5}\rho_L^{1.1}}{\sigma^{0.76}\mu_L^{0.8}} \quad (6)$$

According to Drop-weight Method (Feng *et al.*, 2005), σ can be expressed as Eq. (7).

$$\sigma = \frac{mg}{\pi r} \quad (7)$$

Where m is the drop weight (kg); r is the radius of measuring pipe (m).

On the basis of Eq. (7), the following equation can be gained.

$$\frac{\sigma_1}{\sigma_2} = \frac{m_1}{m_2} \quad (8)$$

Where σ_1 is the unknown liquor surface tension (N/m), σ_2 is the pure water surface tension (N/m) which can be calculated by Eq.(9).

$$\sigma_2 = (75.6 - 0.14t) \times 10^{-3} \quad (9)$$

Where t is the relative temperature (℃).

Through Eq. (9) the pure water surface tension 73.5×10^{-3} N/m (at 15℃) is worked out. The surface tensions of the different MLSS 10, 20 and 40 g/L were measured through Drop-weight Method, and the values were 74.5×10^{-3}, 76.8×10^{-3} and 72.3×10^{-3} N/m at 15℃, respectively. The results indicated that the surface tensions of various MLSS were very close to that of pure water. Therefore, the surface tension of different MLSS is approximately considered to be the same as the value of pure water.

Accordingly, Eq.(6) can be simplified as Eq.(10).

$$K_{La} \propto \frac{T^{0.5} \rho_L^{1.1}}{\mu_L^{0.8}} \quad (10)$$

At a certain temperature (for example 15℃), when the MLSS ranged from 4.5 to 20 g/L, the liquor viscosity increased from 1.25×10^{-3} to 4.5×10^{-3} Pa·s (measured by a revolving viscosity meter) with liquor density changing from 1.0045×10^3 to 1.020×10^3 kg/m^3. Consequently, K_{La} was reduced significantly according to Eq.(10).

The floc size distribution also influenced the SND effect. The blending procedure minimized anoxic micro-zones which were within the flocs and thus reduced SND efficiency. The larger floc sizes could provide larger anoxic micro-zones for the better SND effect.

As for TP removal, two factors affected the removal process. One was the chemical precipitation and the other was the membrane function. The membrane also contributed to the TP removal due to its fine pores. At the same time, the coagulant addition also benefited the MBR operation owing to its reduction of EPS of the mixed liquor.

4 Conclusions

The one-staged aerobic MBR for the treatment of domestic wastewater could achieve SND effect with low DO and suitable sludge floc size. About 70% total nitrogen removal efficiency was achieved when DO concentration was set to 0.2—0.3 mg/L. The high MLSS decreased the oxygen transfer rate and provided the low DO environment. Phosphor was also successfully treated by means of PFS addition and 14 mg/L dosage of PFS was proper for the MBR to removal phosphor. PFS addition also benefited the MBR operation due to its reduction of EPS concentration which accordingly decreased membrane fouling rate.

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