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INVESTIGATION ON BIOMASS PERFORMANCE OF A SUBMERGED MEMBRANE BIOREACTOR FOR TREATING SOY SAUCE WASTEWATER

A pilot study for investigating a membrane bioreactor (MBR) performance in treating soy sauce wastewater was divided into three stages. At stage 1, the MBR was operated without excess sludge withdrawal, and pollutants removal increased gradually and entered into a pseudo-stable phase eventually along with increasing mixed liquor suspended solid (MLSS). At stage 2, the growth of biomass and removal of pollutants were obviously decreased by lowering temperature. At stage 3, membrane permeate flux and sludge yield under various MLSSs were studied. Additional polishing steps including coagulation and oxidation were validated to be effective to ensure the treated wastewater to meet the discharge limits.

SYMBOLS AND SHORTCUTS

DO – dissolved oxygen, $mg\cdot dm^{-3}$
HRT – hydraulic retention time
K_d – The endogenous decay rate, d^{-1}
MLSS – mixed liquor suspended solids, $g\cdot dm^{-3}$
N_{rs} – ratio of F/M, $g_{COD}\cdot g_{MLSS}^{-1}$
PAC – polyaluminium chloride
SRT – sludge retention time, day
SS – suspended solids, $g\cdot dm^{-3}$
TMP – transmembrane pressure, kPa
TP – total phosphorus, $mg\cdot dm^{-3}$
Y – real sludge yield rate, $g_{MLSS}\cdot g_{COD}^{-1}$
Y_{obs} – observed sludge yield rate, $g_{MLSS}\cdot g_{COD}^{-1}$
θ_c – SRT

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1. INTRODUCTION

Soy sauce is an ancient condiment originating from China, known for more than 1800 years. The industrial production of soy sauce by fermentation of soybean, wheat, and bran results in the discharge of large quantities of high strength liquid. Although the BOD_5/COD_{Cr} value of soy sauce wastewater is generally higher than 0.4, indicating a good biodegradability, treating the wastewater via the processes of conventional activated sludge (CAS) still seems to be a serious challenge due to the high contents of organic matter, ammonia nitrogen, salinity, and colour. The most difficult work for treating soy sauce wastewater by CAS is the reduction of colour because of the asynchronous degradation of COD and colour [1]. Moreover, the application of CAS for treating soy sauce wastewater is embarrassed by the high surplus sludge production.

In recent years, membrane bioreactor (MBR) process has gained worldwide attraction and popularity for the treatment of industrial wastewater. Compared with CAS processes, MBR process has great advantages including a smaller footprint, less sludge production and better effluent quality [2, 3]. Several researches have been carried out to study the treatment of soy sauce wastewater with MBR system [1]. The removal efficiencies of pollutants such as organic matter, ammonia nitrogen, colour, turbidity, etc., as well as the selection of optimal parameters such as concentration of dissolved oxygen (DO), hydraulic retention time (HRT), pH, MLSS, etc., have been investigated. In addition, the biophase and permeate flux variations has been analyzed. However there is still lack of systematic investigations on the effects of biomass on the removal of pollutants and the variation of permeate flux.

In treating a high strength liquid such as soy sauce wastewater, the advantages of MBR could be mainly ascribed to the prolonged sludge retention time (SRT) and high biomass concentration. In such a way, the MBR could sustain the notable fluctuation of pollutants contents and the negative impact of high contents of ammonia nitrogen and salinity. The prolonged SRT may facilitate the growth of specialized microorganisms which could decompose the refractory organics in the wastewater. The high concentrated biomass might improve colour removal by sludge adsorption [4]. Nevertheless, high concentration of mixed liquor suspended solids (MLSS) has adverse effects on membrane filtration [5, 6]. It is generally known that the extending of SRT could reduce surplus sludge production [7, 8]. Even zero surplus sludge could be achieved with MBR systems [9–11]. However, for insuring the sufficient pollutants removal and acceptable membrane filtration, complete sludge retention seems to be unreasonable when a MBR system is adopted to treat the high strength industrial wastewater. Thereafter, it is of great importance to clarify the performance of biomass for balancing its effects on pollutants removal and membrane filterability, as well as to know the real sludge yield of MBR as it is valuable for evaluating the feasibility of MBR in treating soy sauce wastewater.

In the present work, a pilot scale MBR system was adopted to treat soy sauce wastewater. The effects of sludge concentration on the removal of pollutants and the permeate flux have been investigated. Sludge yield of the MBR was studied in detail. Due to the fact that the discharging limitation has becoming more stringent in China, the effectiveness of MBR for treating soy sauce wastewater was revaluated according to the discharge criteria, and additional polishing steps including coagulation with polyaluminium chloride (PAC) and oxidation with sodium hypochlorite have been studied. The experimental results were aimed to direct the practical engineering application of MBR for treating the kind of high strength industrial wastewater as soy sauce wastewater.

2. EXPERIMENTAL

Soy sauce wastewater. The pilot experiment was performed in a soy sauce factory, Heshan Donggu Flavoring and Food Co., Ltd. located in Guangdong Province of China. The factory is one of the biggest condiment production base with more than 150 years history in China. The amount of daily wastewater is about 1500 $m^3 \cdot d^{-1}$. The wastewater treatment plant has been built involving an anaerobic biological degradation tank followed with a sequential batch reactor (SBR) and a decolouring process with sodium hypochlorite. In the study, the raw wastewater is taken from the anaerobic tank effluent of the existed wastewater treatment plant. The main characteristics of the MBR influent are given in Table 1.

Table 1

Influent quality of MBR and discharge limits (DEPG, 2001)

Parameters	COD [$mg \cdot dm^{-3}$]	NH_4^+-N [$mg \cdot dm^{-3}$]	Colour [mg Pt-Co$\cdot dm^{-3}$]	Turbidity [NTU]	pH	Salt content [%]
Raw wastewater	500–900	77–174	370–500	ca. 100	5.6–7	0.40–1.35
Discharge limits	≤90	≤10	≤50	≤70	6–9	–

Apparatus. Scheme of the experimental setup for the aerobic submerged MBR is shown in Fig. 1. The reactor is made of PVC and its working volume is 450 dm^3. The reactor is driven by two peristaltic pumps (model No. 7520-57, Cole-Palmer, USA). One of the peristaltic pumps is for feeding raw wastewater and the other is for drawing membrane permeate. Two bundles of “outside-in type” modified polypropylene hollow fiber UF membrane (Jingcheng Membrane Co., Ltd., KunShan, China) with an effective surface area of 8 m^2 are set in the reactor. The membrane fibers are horizontally submerged in the reactor with a length of 800mm. The nominal pore diameter of the membrane is 0.2 μm. Air diffusers are mounted underneath the membrane in the reactor providing a constant air flow to mix the suspensions, scour the membrane surface for fouling reduction and maintain the dissolved oxygen (DO) concentration of >2.5 $mg \cdot dm^{-3}$. HRT is controlled at 48 h throughout all stages of the experiment. Na_2CO_3 solution is added to maintain pH at about 7 in the reactor using a pH controller (model Liquitron DP 5000, LMI Milton Roy, USA) and an electromagnetic dosing pump (model UL#A752 -393SI, LMI Milton Roy, USA) because pH of the reactor would decrease in the test.

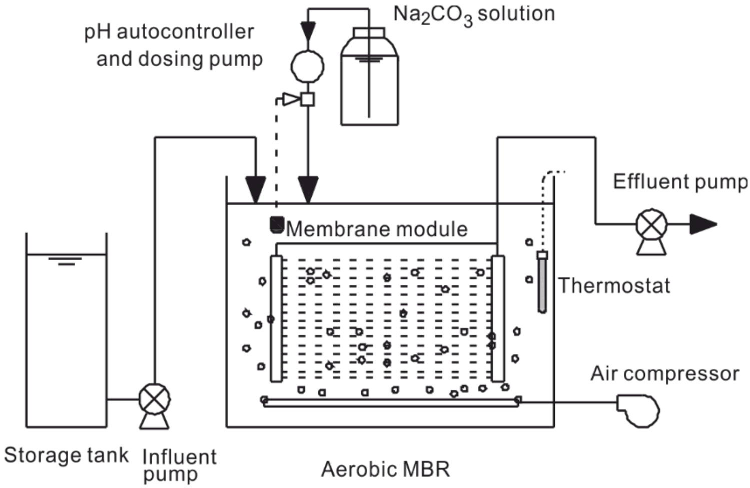

Fig. 1. Schematic diagram of experimental apparatus

Experimental procedure. After the inoculation and acclimation of sludge, which was collected from a local municipal wastewater treatment plant, the experiment divided into three stages was started. At the first stage (68 days), the MBR was operated without surplus sludge withdrawal for investigating the effect of sludge concentration on the pollutants removal. The ambient temperature in that period was kept at 30±2 °C by an adjustable heater. At the second stage (35 days), the effect of temperature on the performance of MBR was investigated between the 11th November and the 15th December, during which the local temperature was low, ca. 7 °C. The liquid temperature was adjusted by the heater in this experiment, and sludge withdrawal was not conducted in that period. At the third stage, the variation of membrane permeate flux and observed sludge yield was detected under two different approximate steady biomass concentrations maintained by periodical sludge withdrawal (100 days for MLSS 11.7 $g\cdot dm^{-3}$ and 212 days for MLSS 8 $g\cdot dm^{-3}$). The temperature in this stage was controlled similarly as in the first stage and the transmembrane pressure (TMP) was maintained at about 0.01 MPa. The additional polishing process including coagulation with polyaluminium chloride (PAC) and oxidation with sodium hypochlorite (NaClO, 13 wt. %) was conducted in the third stage. The results after treating wastewater were compared with the discharge limits of water pollutants (DB 44/26-2001) which was established in Guangdong, China (Table 1).

While the membrane was clogged (permeate flux lower than 10 $dm^3\cdot m^{-2}\cdot h^{-1}$), ex situ physical cleaning (jetting the module with tap water under moderate pressure) and chemical cleaning (soaking the module in 2% sodium hydroxide and 1% sodium hypochlorite mixed solution for 12 h) were applied to restore the membrane permeability.

Analytical methods. Chemical oxygen demand (COD), total phosphorus (TP), NH_4^+-H, MLSS and mixed liquor volatile suspended solids (MLVSS) were analyzed using standard methods [12]. pH was measured with a pH meter (pHS-3C, China). Dissolved oxygen (DO) and temperature were determined using a portable DO meter combined with a temperature probe (JBP-607 DO, China). Colour was determined by the platinum-cobalt colorimetry in mg Pt-Co·dm^{-3}.

All the tests were repeated twice under the identical conditions and the mean of the two measurements was used.

3. RESULTS AND DISCUSSION

3.1. EFFECT OF MLSS CONCENTRATION ON POLLUTANTS REMOVAL

COD values in membrane permeate and its removal rates in function of MLSS concentration are shown in Fig. 2a. In the first stage, the maximum MLSS concentration in MBR could reach 19.934 g·dm^{-3} which was similar to the value obtained byTeck et al. [8]. COD removal rates increased rapidly with the increasing MLSS concentration to ca. 6 g·dm^{-3}. Thereafter, the increasing trend slowed down. The COD values varied around 120 mg·dm^{-3} as MLSS concentration was higher than 8 g·dm^{-3}. The lowest permeate COD value was about 116 mg·dm^{-3} which could not meet the discharge limits [13].

As widely known, slow-growing microorganisms such as nitrifying bacteria can be enriched in MBR and high, and stable ammonia removal can be achieved in treating industrial wastewater with high ammonia content [11,14]. In the present work (Fig. 2b), NH_4^+-N removal rate was higher than 80% after the successful inoculation and acclimation of sludge. Along with the increasing of MLSS concentration, NH_4^+-N removal rate increased gradually to more than 90%. While MLSS exceeded 9 g·dm^{-3}, the variation of NH_4^+-N in permeate entered into a pseudo stable phase and NH_4^+-N in permeate remained steadily at less than 10 mg·dm^{-3}. Chiemchaisri et al. [15] reported that MBR could remove nitrogen through manipulations of its biological process. Suwa et al. [16] found that simultaneous nitrification and denitrification could occur under a high DO concentration in an MBR.

As can be seen in Figure 2c, colour removal increased rapidly as MLSS concentration increased from 3.1 g·dm^{-3} to 7.5 g·dm^{-3}, and entered into a slower increasing phase for MLSS concentration higher than 7.5 g·dm^{-3}. The colour of 93 mg Pt-Co·dm^{-3} corresponding to the removal rate of 78.9% was eventually achieved at the maximum MLSS of 19.934 g·dm^{-3}.

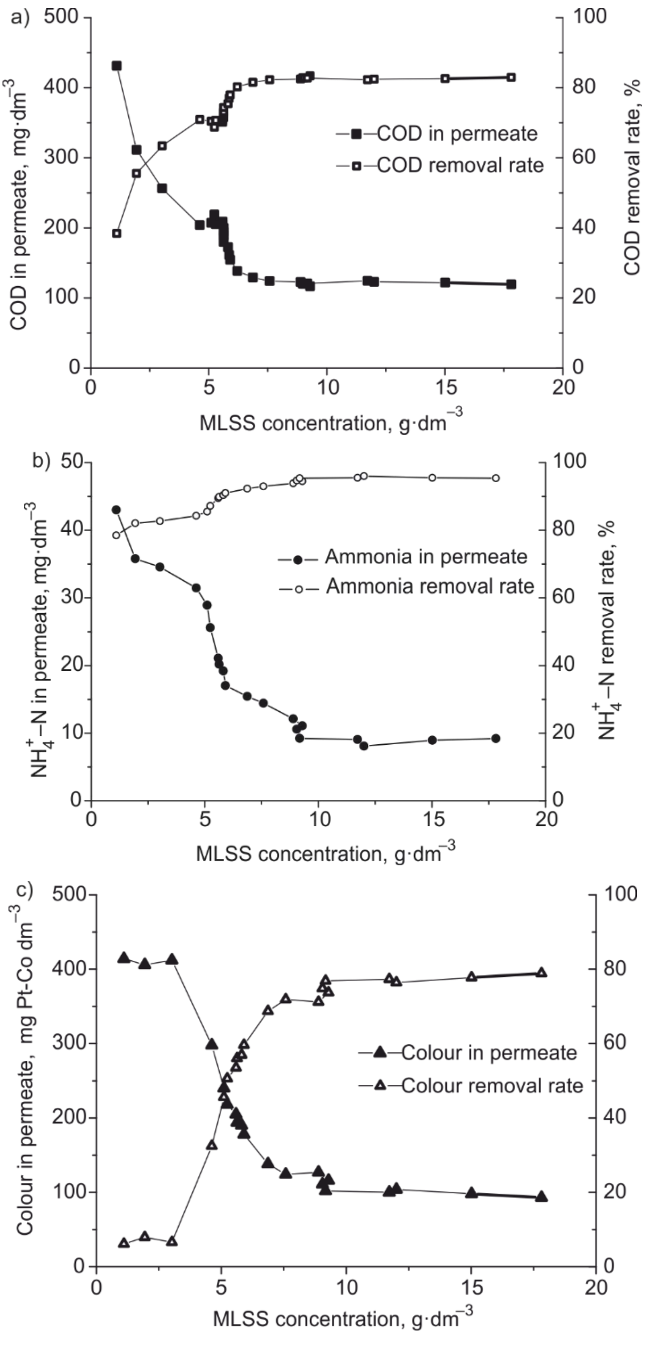

Fig. 2. Dependences of a) COD, b) NH_4^+-N, c) colour in permeate and its removal rate on the MLSS concentration

The colour matter in soy sauce wastewater was regarded to be generated from the Maillard reaction in the fermentation process and the manual addition of caramel pigments for product preparing. CAS processes are generally considered to be ineffective to remove the colour in soy sauce wastewater. Tian et al. [17] reported that one SBR could reduce 19.4% of the colour in soy sauce wastewater and two SBR in series could reduce 38.9%. The present colour removal rate is clearly far higher than that published in the literature. The reason could be that the main mechanism inducing colour removal includes two aspects as biological degradation and biomass adsorption. Brik et al. [4] reported a distinct relationship between sludge growth and colour removal as a MBR was applied to treat textile wastewater. They observed that an increasing sludge growth rate yielded higher colour removal efficiencies and recommended sludge growth rates above 0.3 $g\cdot dm^{-3}\cdot d^{-1}$, corresponding to the sludge age of 50 days at the sludge concentration of 15 $g\cdot dm^{-3}$, to attain maximum colour removal efficiencies.

During this experiment, the salinity of 0.40–1.35% seemed to have no effect on the performance of MBR. The observation was supported by Hamoda and Al-Attar [18]. They found that the organic (COD or TOC) removal efficiency and the effluent quality of the activated sludge system were not deteriorated by high salinity even as 3 $g\cdot dm^{-3}$. Sridang et al. [3] also reported that there was no effect of salinity on the performance of MBR in treating seafood processing wastewater. This can be attributed to the acclimation of sludge for selecting salt tolerant microbial species.

3.2. EFFECT OF TEMPERATURE ON MBR PERFORMANCE

During the second stage, liquid sample measurement for data yielding was implemented once every 3 days (Fig. 3). The liquid temperature decreased rapidly from 25 °C to the lowest value of 10 °C on day 6, and then increased gradually to 25 °C on day 25, thereafter maintained at around 25°C until the end of the period. It could be seen that, the reduction of temperature caused a decline of MLSS concentration from 11.9 $mg\cdot dm^{-3}$ to 10.4 $mg\cdot dm^{-3}$, and eventually recovered to the initial value along with the temperature increasing. Compared with the increasing trend of MLSS in the present work except the period of temperature reduction, the narrow variation of MLSS without any sludge withdrawal indicated an inhibited biomass growth caused by temperature reduction. Considering the observed sludge yields which will be shown in the following section, a clear decay of sludge growth in the period could be assumed.

As to the removal of pollutants, different variations were observed in the period. The COD removal rate showed little change in the first 6 days and expeditious decrease after day 6 from 72% to 60% on day 9. The low COD removal rate was kept steadily till day 28, after which the rate increased to about 72% on day 35. The NH_4^+-N removal rate decreased from the initial 89% to about 77% on day 10 and recovered to 84% on day 14, thereafter NH_4^+-N removal rate varied around 85%. Comparatively, colour removal rates were affected by temperature more significantly. Along with the decreasing temperature, the colour removal rate decreased clearly from the initial one of about 75% to the lowest rate of 26% on day 17, and thereafter a gradually increasing trend was observed till day 28 to about 77%.

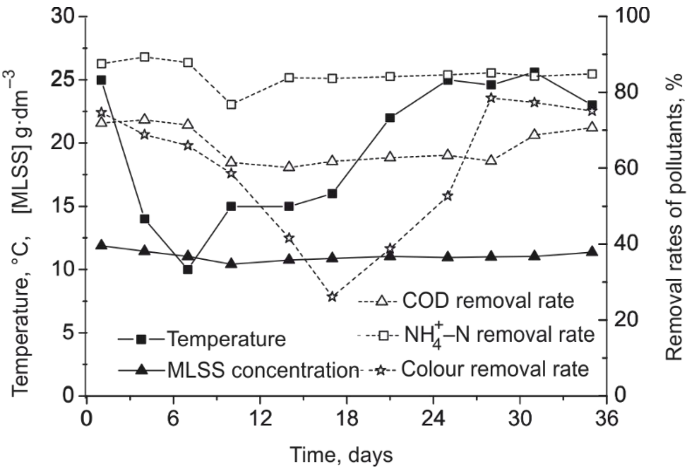

Fig. 3. Time dependences of temperature of liquid,
MLSS concentration, and the removal rates of COD, NH_4^+-N, colour

A rule could be attained from Fig. 3 that, the decrease of pollutants removal occurred later than the temperature reduction, and the recovery of pollutants removal also occurred later than the temperature recovery. This indicated that, on the one hand, the MBR could sustain a short period of temperature reduction; on the other hand, acclimation for several days was inevitable to resume the decreased bioconversion efficiency. The effect of temperature on NH_4^+-N removal was relatively smaller than the other two. A support research was carried out by Chae and Shin [19] who reported that remarkable differences were not found in nitrification efficiency (79–88%) at differing temperatures (13–25 °C). The relatively significant decrease of colour removal resulting from temperature lowering might be attributed to the colour removal mechanism which includes not only biological degradation but also adsorption on biomass. The sludge growth rate was restrained by temperature reduction; hence the adsorption of colour onto biomass was reduced [4].

3.3. EFFECT OF MLSS CONCENTRATION ON MEMBRANE PERMEATE FLUX

Due to the retention of suspended solids (SS) by membrane, MBR has the advantage of a higher biomass concentration than most CAS processes. Nevertheless, the concentration of biomass could also influence membrane permeability [5, 6]. The examination of membrane permeate flux under two different MLSS concentration, i.e. 8 ^{3} and 11.7 g·dm^{-3}, as shown in Fig. 4 revealed that, higher MLSS concentration could cause faster decrease of permeate flux. Similar results have been reported in previous [6], and the reason may be attributed to: 1) more particles accumulate on the membrane [9], 2) sludge accumulation and thickening occurs in the spaces among the fibres [10], 3) high filtration resistance results from high viscosity [20].

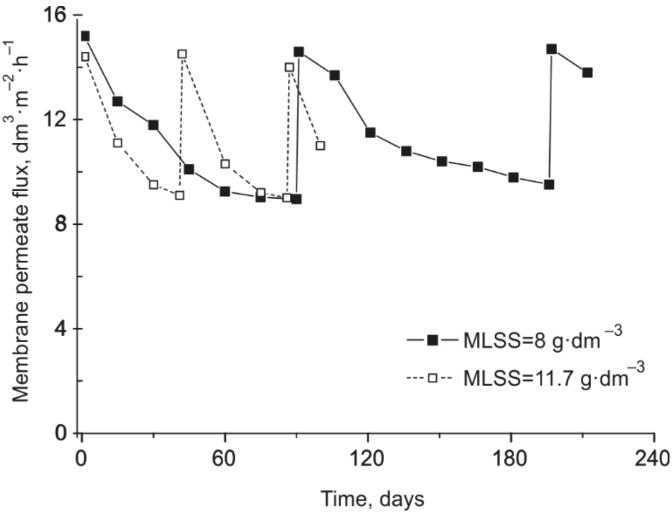

Fig. 4. Time dependences of permeate flux under MLSS of 8 and 11.7 g·dm^{-3}

The duration of decreasing of permeate flux from the initial 15 to 10 dm^3·m^{-2}·h^{-1} under MLSS of 8 g·dm^{-3} was nearly twofold higher than that under MLSS of 11.7 g·dm^{-3}. Tian and Zuo [1] recommended that the optimum MLSS concentration of MBR for treating soy sauce wastewater was 8–9 g·dm^{-3}. Wu and Huang [5] reported that the MLSS concentration of higher than 10 g·dm^{-3} had significant correlation with membrane filterability; nevertheless, MLSS concentration lower than 10 g·dm^{-3} had almost no effects on the membrane fouling rate and could be recommended to treat soy sauce wastewater by MBR.

3.4. SLUDGE YIELD

For the steady state system, the relationship of SRT and food-to-microorganism (*F*/*M*) rate can be described by the following equation [21]:

$$\frac{1}{\theta_c} = YN_{rs} - K_d \tag{1}$$

where N_{rs} = *F*/*M*, *Y* is the real sludge yield rate, θ_c is the SRT and K_d is the endogenous decay rate.

The reciprocals of SRT were plotted versus the values of N_{rs} (Fig. 5a), *Y* = 0.295 $g_{MLSS}\cdot g_{COD}^{-1}$ and K_d = 0.0222 d^{-1} were calculated.

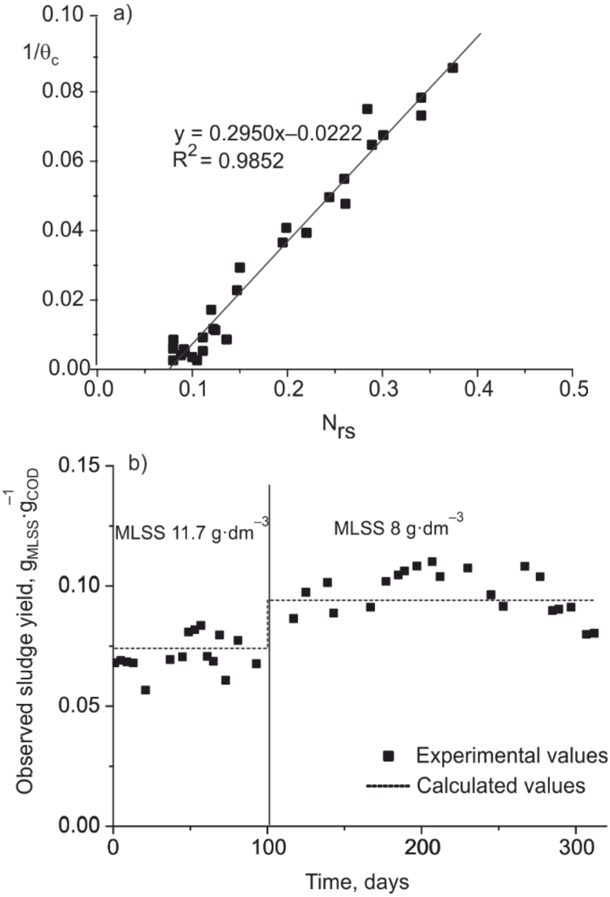

Fig. 5. The reciprocals of SRT ($1/\theta_c$) vs. N_{rs}; *Y*, and K_d calculated from the slopes and intercepts based on Eq. (1) (a), as well as observed experimental and calculated sludge yields based on Eq. (2) under MLSS of 11.7 and 8 $g\cdot dm^{-3}$ (b)

The relationship between the observed sludge yield (Y_{obs}) and SRT can be described by the following expression [21] :

$$Y_{obs} = \frac{Y}{1 + K_d \theta_c} \qquad (2)$$

The SRT values corresponding to MLSS of 8 and 11.7 $g \cdot dm^{-3}$ were 96 and 145 days, respectively. Based on Eq. (2), calculated Y_{obs} for the two MLSS concentration were 0.094 and 0.074 g_{MLSS} g_{COD}^{-1}, respectively. The observed sludge yield was plotted against time as shown in Fig. 5b. It can be seen that the observed sludge yields varied around the calculated values.

In the CAS process for treating domestic wastewater, sludge yield and endogenous decay are normally in the range of 0.30–0.50 $g_{VSS} \cdot g_{COD}^{-1}$ and 0.06–0.20 d^{-1}, respectively. The sludge yield, Y of the present MBR was slightly lower than the lower value for CAS. The endogenous decay coefficient, K_d obtained in the present work, was nearly one-third of the lower value for CAS. Huang et al. [22] reported that, the sludge decay coefficient decreased exponentially with the SRT and this phenomenon might relate to oxygen transfer in the bioreactor. Similar low sludge yield (0.22 $g_{VSS} \cdot g_{COD}^{-1}$) and endogenous decay coefficient (0.05 d^{-1}) were achieved by Teck et al. [8] who used MBR to treat industrial wastewater with a SRT of 300 days. The low sludge yield and endogenous decay coefficient may be due to that most of the energy provided by the substrate consumed is deemed to be used mainly for cell maintenance, i.e. proved the maintenance energy concept [7].

A decreasing tendency of the observed sludge yield for increasing sludge concentration has also been reported [7, 10]. Similar results present other authors who reported the sludge yields of MBR for treating textile wastewater (0.06–0.1 $g_{MLSS} \cdot g_{COD}^{-1}$) [4] municipal wastewater (0.12 $g_{MLVSS} \cdot g_{COD}^{-1}$) [10], synthetic wastewater (0.11 $g_{MLVSS} \cdot g_{COD}^{-1}$) [7], seafood wastewater (0.09 $g_{MLVSS} \cdot g_{COD}^{-1}$) [3], laundry wastewater(0.14–0.16 $g_{TSS} \cdot g_{COD}^{-1}$) [2]. Considering that the treatment of surplus sludge may account for up to 60% of the total plant operation cost [9], the low observed sludge yield in the present study may greatly improve the feasibility of MBR in treating soy sauce wastewater.

3.5. ADDITIONAL TREATMENT FOR WASTEWATER DISCHARGE

Although the pollutants removal by the MBR were high, the concentrations of organic matter and colour were still not adequate for discharging, making subsequent polishing steps mandatory. Coagulation with polyaluminium chloride (PAC) and oxidation with sodium hypochlorite (concentration of NaClO – 13 wt. %, Dongguan Dosheng Synthetic Material Inc., China) were adopted for reducing the residual organic matter and colour in the wastewater.

As can be seen in Fig. 6a, along with the increasing of PAC dosage, variations of COD and colour removal as increase first and decrease afterward were observed. The coagulation dosage of 80 $mg \cdot dm^{-3}$ seemed to be the optimum as that the effluent COD and colour were less than 90 $mg \cdot dm^{-3}$ and around 60 mg Pt-Co$\cdot dm^{-3}$, respectively. As the solution containing 13% of sodium hypochlorite was added for further decolouring

(Fig. 6b), the colour removal was improved with the increasing dosage of sodium hypochlorite. While the solution dosage was larger than 1 $cm^3 \cdot dm^{-3}$, the effluent colour was less than 40 mg Pt-Co$\cdot dm^{-3}$ which could meet the discharge limits [13].

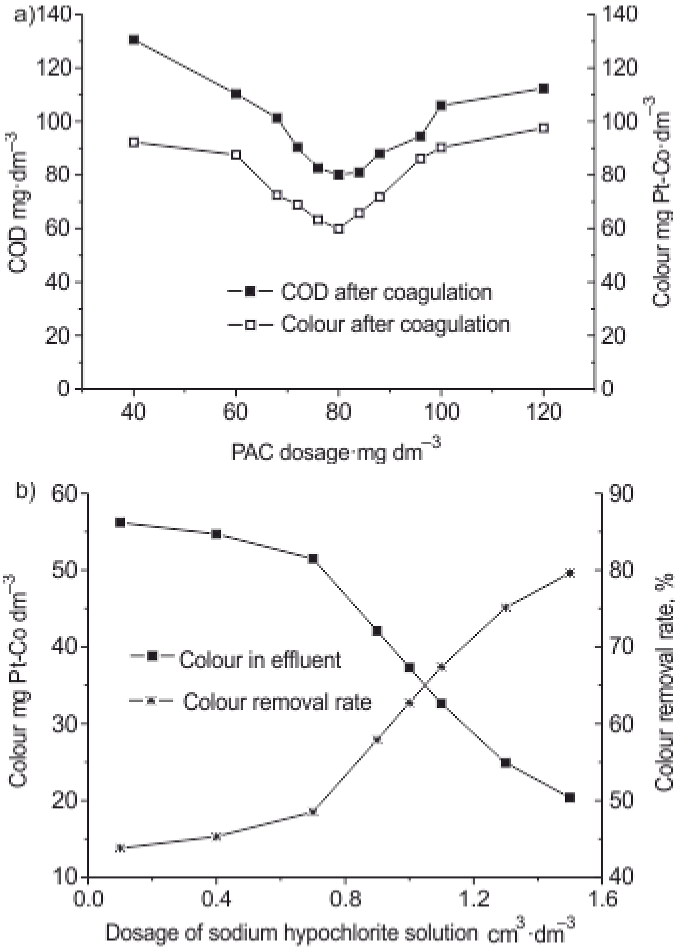

Fig. 6. Dependences of COD and colour after coagulation on the polyaluminium chloride dosage (a), as well as dependences of colour and colour removal rate after oxidation on the dosage of sodium hypochlorite solution (concentration of NaClO – 13 wt. %) (b)

As coagulation with PAC [23, 24] and oxidation with sodium hypochlorite [25] were widely used in conventional wastewater treatment process, and the total cost in the present work was calculated to be less than 0.3 China yuan per ton wastewater treated, the additional polishing steps seemed to be suitable for treating the soy sauce wastewater following the MBR technology.

4. CONCLUSIONS

A pilot MBR was adopted to treat soy sauce wastewater. The concentration of MLSS could influence the removal rates of pollutants and the membrane permeate flux clearly, and MLSS lower than 10 $g \cdot dm^{-3}$ seemed to be beneficial to balancing its effects on pollutants removal and membrane filterability. Temperature reduction caused the activity and growth of biomass to decline, subsequently affect the removal of pollutants. The observed sludge yields corresponding to MLSS of 8 and 11.7 $g \cdot dm^{-3}$ were 0.094 and 0.074 $g_{MLSS} \cdot g_{COD}^{-1}$, respectively. To meet the discharge limits, coagulation and oxidation following MBR were verified to be effective to reduce the residual COD and colour.

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