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Pollutants removal and simulation model of combined membrane process for wastewater treatment and reuse in submarine cabin for long voyage

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Abstract

A laboratory scale test was conducted in a combined membrane process (CMP) with a capacity of 2.91 m^3/d for 240 d to treat the mixed wastewater of humidity condensate, hygiene wastewater and urine in submarine cabin during prolonged voyage. Removal performance of chemical oxygen demand (COD), ammonia nitrogen (NH_4^+-N), turbidity and anionic surfactants (LAS) was investigated under different conditions. It was observed that the effluent COD, NH_4^+-N, turbidity and LAS flocculated in ranges of 0.19–0.85 mg/L, 0.03–0.18 mg/L, 0.0–0.15 NTU and 0.0–0.05 mg/L, respectively in spite of considerable fluctuation in corresponding influent of 2120–5350 mg/L, 79.5–129.3 mg/L, 110–181.1NTU and 4.9–5.4 mg/L. The effluent quality of the CMP could meet the requirements of mechanical water and hygiene water according to the class I water quality standards in China (GB3838-2002). The removal rates of COD, NH_4^+-N, turbidity and LAS removed in the MBR were more than 90%, which indicated that biodegradation is indispensable and plays a major role in the wastewater treatment and reuse. A model, built on the back propagation neural network (BPNN) theory, was developed for the simulation of CMP and produced high reliability. The average error of COD and NH_4^+-N was 5.14% and 6.20%, respectively, and the root mean squared error of turbidity and LAS was 2.76% and 1.41%, respectively. The results indicated that the model well fitted the laboratory data, and was able to simulate the removal of COD, NH_4^+-N, turbidity and LAS. It also suggested that the model proposed could reflect and manage the operation of CMP for the treatment of the mixed wastewaters in submarine.

Key words: combined membrane process; wastewater treatment and reuse; back propagation neural network

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Introduction

Submarine cabin was a typical airtight space isolated from atmosphere during prolonged underwater voyage, which was similar to spacecraft and space station (Millet *et al.*, 1996). There were very high temperature (26–50°C) and limited fresh water which could not meet the requirement of mechanism and hygiene of crews in submarine (Zhang and Wang, 2007). Some regulations are established for using fresh water for long-range missions: only a glass of water for toothbrushing and washing face; and bath only once a week (Zhang, 2006). In addition, concealment of submarine was particularly important due to the special battle features, which limited the wastewater discharge (Carrasquillo, 1991; Cath, 2003). In such circumstance, the supplement of water became more important.

During long-range missions, hygiene wastewater, urine and humidity condensate can be reclaimed and reused. For example, a sorption-catalytic method with subsequent mineralizing, preservation with ionic silver and pasteurization of the product water is used for water recovery from humidity condensate (Samsonov, 1993; Colley, 1991; Samsonov and Farafonov, 1993). Water reclamation from urine is provided via distillation with sorption-catalytic purification of the condensate (Samsonov and Farafonov, 1993; Samsonov, 1994; Hutchens, 1994; Zdankiewicz, 1985, 1986; Miernik, 1991). The treatment of hygiene water would be achieved by ultra-filtration with subsequent sorption and ionic exchange (Samsonov, 1993, 1994; Colley, 1991; Samsonov and Farafonov, 1993). In January 1975, in the space station “Salyut-4”, Gubarev and Grechko used the water recovered from humidity condensate for the drinking and food preparation (Vorobyov *et al.*, 1977). The system was operational in the entire flight period. Similar systems of SRV-K types had operated on space and Salut-7 (1982–1986 for 743 d). The SRV-K system together with the supplement system provided reused water for the crew. In addition, the water recovery system purified water of supplies with an exceeded storage time limit, provided hot water for hygiene procedures. Since 1988, Zhou (2002) had begun to investigate the regenerative wastewater treatment system for the living of three men for 62 d, and realized a closed circulation of water. The transformation efficiency of humidity

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condensate regenerating to potable water reached 80%, hygiene water and urine were also entered into water regenerative circle, in which the supplement water in the circle from the external environment is 2.2 kg per day (Fu *et al.*, 2007). The above-mentioned studies all focused on the treatment and recycling of wastewater in spacecraft and space station. However, so far no experimental study on biological processes was reported to remediate mixed high-strength wastewater of submarine.

The objective of this study is to develop a combined membrane process (CMP). To authors' knowledge, the application of CMP for the treatment of submarine mixed wastewaters remains scarce. Therefore, a laboratory-scale system comprising a membrane bioreactor (MBR), security filter, ultrafilter (UF), activated charcoal filter, advanced oxidation of TiO_2/UV process and reverse osmosis (RO) equipment was run at a fixed wastewater flow rate of 2.91 m^3/d for 240 d. The effluent of chemical oxygen demand (COD), ammonia nitrogen (NH_4^+-N), turbidity and anionic surfactants (LAS) were evaluated. Furthermore, to simulate and manage the operation of CMP a model was established based on the theory of back propagation neural network (BPNN). To predict the effluent quality of CMP. The simulation model was established by using software program MATLAB which does not appear to have been used in the similar research.

1 Materials and methods

1.1 Wastewater characteristics

The wastewater in submarine could be divided into humidity condensate, hygiene wastewater and urine (Samsonov, 1993). The characteristics of the wastewater are given in Table 1. The pollutants in humidity condensate were mainly smaller molecular-weight and soluble organic compounds. Hygiene wastewater was generated from washing operations and contained a large amount of suspended substance, hair and organic pollutants such as COD and LAS. Urine was the most complicated wastewater and the pollutants of which were mainly urea, sodium chloride and different acids. The major characteristics of the mixed experimental wastewater were as follows: COD of 2120–5350 mg/L, NH_4^+-N of 79.5–129.3 mg/L, turbidity of 110.0–179.3 NTU, LAS of 4.88–5.44 mg/L.

The daily production rates of consumed substance and product per person are given in Table 2. The submarine CMP was constructed with tanks having a capacity to accommodate the daily wastewater generated by 100 crew members, and then the quantity of experimental wastewater was 2.91 m^3/d with humidity condensate of 180 L/d, hygiene wastewater of 2530 L/d, urine of 200 L/d, respectively.

Table 1 Characteristics of wastewaters

	Substance	Content (mg/L)
Humidity condensate	COD	250–650
	Ammonia	50
	Chlorides	25
	Sulphates	< 5
	Nitrates	< 1
	Alcohols	< 300
	Acids	< 30
	Bacteria (CFU/L)	104–107
Hygiene wastewater	COD	2000–5000
	Ammonia	< 80
	Chlorides	< 10
	Sulphates	< 300
	Nitrates	< 1000
	Bacteria (CFU/L)	106–108
Urine	Urea	13000–20000
	Sodium chloride	8000–12000
	Creatine	400–1600
	Phosphorus	1300–1500
	Ammonia	70–800
	Hippuric acid	100–1600
	Uric acid	200–800
	Potassium	1500–2000
	Calcium	150–700
	Magnesium	150–250
	Sulphur	1200–1400
	Sulphates	1000–2000
	Amino acid	150–350
	Fatty acid	30–60
	Phenols	100–300

Table 2 Production rate of consumed substance and product

Consumed substance	Mass (kg/(d·person))	Product	Mass (kg/(d·person))
Oxygen	0.9	Carbon dioxide	1
Drinking water	2	Humidity condensate	1.8
Water potable in food	0.5	Urine	2
Food (dry weight)	0.6	Urine brine	0.05
Water for hygiene	6	Hygiene wastewater	25.3
		Water in feces	0.25
		Feces (dry weight)	0.05

1.2 Laboratory-scale setup and operation

A schematic representation of the submarine CMP is shown in Fig. 1. The system mainly consisted of an MBR, a security filter, a ultrafilter, an activated charcoal filter, an advanced oxidation of TiO_2/UV process and a reverse osmosis equipment. The MBR had a working volume of 525 L with a dimension of 1.06 × 0.50 × 0.99 m^3 (length × width × height) and was installed with three submerged hollow-fiber PVDF microfiltration (MF) membrane modules (Tianjin Motian Membrane Engineering & Technology Co., Ltd., China). The MF membrane modules are characterized with a pore size of 0.22 μm and an effective surface area of 2 m^2. Dissolved oxygen (DO) concentration of the MBR was maintained at 1.2–3.0 mg/L by adjusting the airflow to between 10 and 15 mg/h. The liquid level was controlled by a level controller equipped with a level sensor, while the reactor temperature was maintained at 20°C through auto-controlled heat exchangers. Security filter was made up of a spiral wound stainless steel filter element with 400 mm long, and filtering precision was 5 μm. Ultrafilter was installed with a inter compression type hollow fiber polyacrylonitrile (PAN) membrane module cuboidally with a square section of 50 mm^2 and a height of 480 mm, the molecular weight cut-off of which was 30000 dalton. The membrane

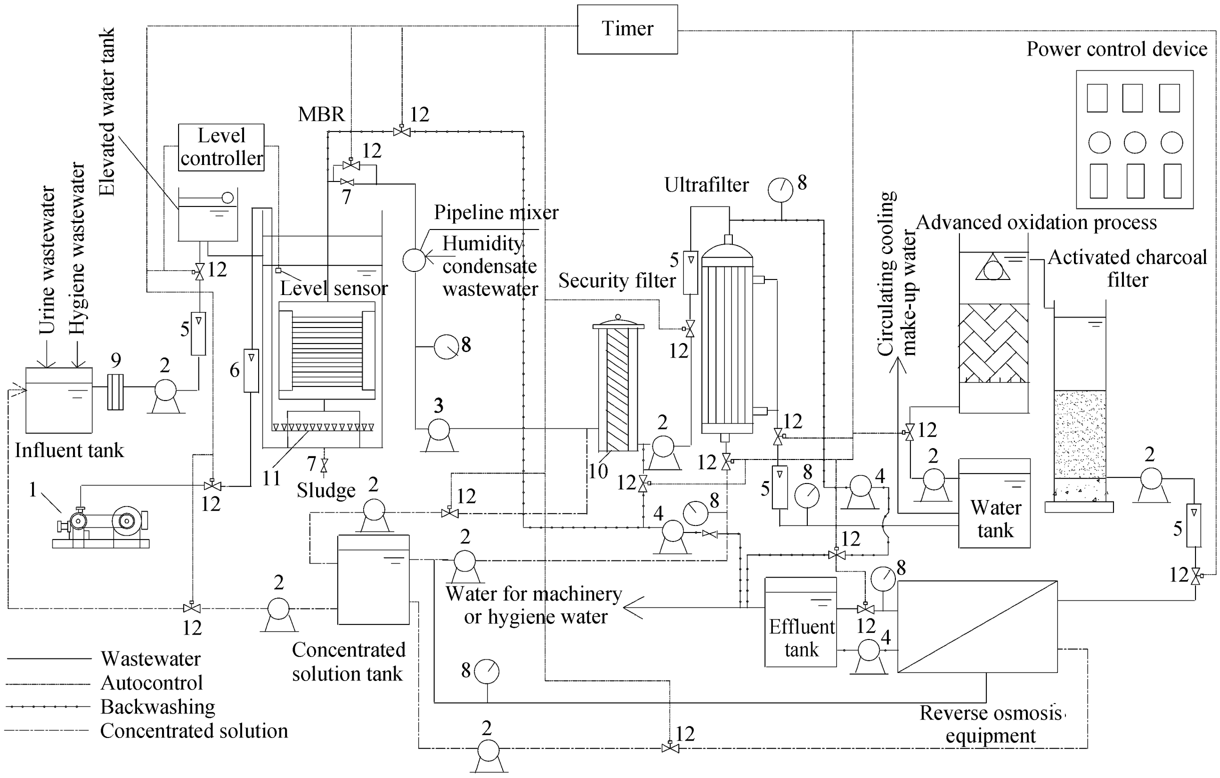

Fig. 1 Treatment and reuse system for submarine. (1) air compressor; (2) peristaltic pump; (3) vacuum pump; (4) backwashing pump; (5) liquid flow meter; (6) gas flow meter; (7) valve; (8) pressure indicator; (9) quick opening hair catcher; (10) filter element; (11) air diffused device; (12) electron-magnetic valve.

modules were characterized with an effective surface area of 2 m^2. Activated charcoal filter was put into operation downstream which was made up of PMMA (polymethyl methacrylate) hollow cylinder with the inner diameter 86 mm, the height 800 mm, among which the effective height of activated carbon was 300 mm and the height of supporting layer (quartz sand) was 70 mm. Reverse osmosis equipment was installed with a hollow fiber Tri-Acetate (CTA) membrane module to assure considerably high total salt rejection, with a dimension of Φ 100 × 1016 mm and an effective membrane area of 1.5 m^2.

The mixed wastewater (including hygiene wastewater and urine) was collected in an influent tank. First, the mixed wastewater crossed the quick opening hair catcher before entering into MBR to remove abundant hair in hygiene wastewater. Subsequently, the wastewater was treated in a membrane bioreactor as the principal cleaning unit. Air was injected into the reactor to scour the membranes and to provide oxygen for biological degradation. It was free of turbidity and considerably reduced in microbes. Humidity condensate had almost the same COD as the microfiltration permeate. Hence both mixed in pipeline mixer were treated in a second step by a security filter to remove large suspended particles ensuring adequate membrane permeability subsequently. The effluent of security filter was treated in an ultrafilter which was basically a pressure-driven process that separated on the basis of molecular diameter. For UF permeate stored in a water tank, one part was used as circulating cooling make-up water and another part was treated in advanced oxidation of TiO_2/UV process to remove refractory organics. Then the activated charcoal filter received the treated effluent from the advanced oxidation of TiO_2/UV process, where the majority of pollutant had already been removed from the liquid phase. Therefore, only the more persistent pollutants were presented, creating a challenge for removal. Prior to storage, the effluent from the activated charcoal filter was treated by a low pressure RO unit with hollow fiber membrane modules to retain mainly salts. The concentrated solutions of security filter, UF and RO were deposited in a concentrated solution tank. The RO permeate met the class I water quality standards in China (GB3838-2002), thus it can be used as hygiene water or machinery water. An automatic vacuum effluent system was used to control the system working volume at a constant value. Pressure indicators, flow meters, and electron-magnetic valve were installed to monitor and control the operation of the system. The operating conditions of CMP are summarized in Table 3.

1.3 Analytical methods

Standard methods (APHA, 1995) were used to determine COD, NH_4^+-N, turbidity, and LAS, which were referenced the class I water quality standards in China (GB3838-2002). Effluent COD, NH_4^+-N, turbidity had the prescribed limited values in standards, which were 15 mg/L, 0.15 mg/L, 3 NTU and 0.2 mg/L, respectively. Software MATLAB was applied in numerical analyses.

Table 3 Operating conditions of combined membrane process

	Component	Value
Membrane bioreactor	Sludge retention time (d)	300
	Hydraulic retention time (h)	3–5
	Dissolved oxygen (mg/L)	2–4
	Membrane flux (L/(m^2·h))	20
	Water flux (L/h)	114
	Gas/water ratio	20:1–30:1
	Operation pressure (MPa)	0.02–0.05
Security filter	Filtering precision (m)	5
	Pressure difference (kPa)	< 58.8
	Influent SS (mg/L)	< 1000
Ultrafilter	pH	2–13
	Temperature (°C)	$\leqslant$ 45
	Operation pressure (MPa)	$\leqslant$ 0.2
	Filtration mode	Cross flow
	Backwashing time (s)	30
	Frequency of backwashing (times/h)	4
Activated charcoal filter	pH	6–9
	Turbid (NTU)	$\leqslant$ 5
	Filtration rate (m/h)	3–5
	Hydraulic retention time (min)	4.8–7.2
	Backwashing intensity (L/(s·m^2))	12
Advanced oxidation of TiO_2/UV	TiO_2 amount (g/L)	5
	Illumination time (h)	3
	pH	8
Reverse osmosis	pH	5–6
	Temperature (°C)	25 ± 1
	Operation pressure (MPa)	1.5 ± 1
	Recovery rate (%)	$\leqslant$ 50
	Desalinization ratio (%)	$\geqslant$ 90–95
	Expansion ratio (%)	28

2 Results and discussion

2.1 Effect of CMP on COD removal

The changes in COD removal and effluent COD values with time are illustrated in Fig. 2. The COD concentration of mixed wastewaters was varied from 2120 to 5350 mg/L. Corresponding to this variation in the influent strength, the average value of MBR effluent COD decreased gradually to 275.8 mg/L, the COD removal rate became stable since day 50, and remained constant at a rate of approximately 95% throughout the experiment. Such high removal rate showed that biological effect played an important role in the elimination of COD. It was noted that MBR holds strong anti-shock loading capability, implying its convenience and endurance in practical operation and the security in treated water quality. The security filter significantly reduced COD with an removal rate from the liquid phase of 20%–28%, and the average COD effluent remained around 221.9 mg/L. The ultrafilter stage of the plant removed approximately 37%–57% of COD in the liquid phase, the average value of effluent COD decreased to 168.4 mg/L, and the permeate COD showed downtrend along with running time due to the cake layer formed on the membrane surface with the increase of running time, which could react in solution to induce the influent COD of ultrafilter. The effluent COD of advanced oxidation of TiO_2/UV process fluctuated from 5.2 to 32.6 mg/L, with an average 13.2 mg/L. The average COD elimination rate in the advanced oxidation process (AOP) unit was 92%. The subsequent process-activated charcoal filter eliminate organic compounds effectively; the effluent COD value was always lower than 20 mg/L and typically averaged in the range of 2.5–13.1 mg/L, and the COD removal rate could reached 68%. The RO membrane reduced the COD by the removal rate of approximately 95%, the permeate COD decreased to 0.38 mg/L averagely, fluctuated from 0.19 to 0.85 mg/L, which fully met the requirements of the class I water quality standards in China (GB3838-2002) for mechanical or hygiene water.

2.2 Effect of CMP on NH_4^+-N removal

The changes in NH_4^+-N removal and effluent NH_4^+-N with time are illustrated in Fig. 3. NH_4^+-N was one of the most important oxygen-consuming contaminants, which mainly came from the excrements of crews. The NH_4^+-N in humidity condensate and hygiene wastewater was relatively stable, and the concentration of mixed wastewaters ranged from 79.5 to 129.3 mg/L, with the average value of 100.7 mg/L. Results showed that the effluent of each process unit corresponded to the variation in the influent strength. The average NH_4^+-N value of MBR effluent decreased gradually to 5.74 mg/L, and the NH_4^+-N removal rate remained constant with a rate of approximately 94%. Such high removal rate showed that biological conversion played an important role in the elimination of NH_4^+-N. It can be assessed that there was well nitrification in the MBR due to an ample oxygen, an appropriate pH value and a relatively long sludge retention time. The security filter significantly reduced NH_4^+-N with an removal rate of 1.32%, and the average NH_4^+-N effluent remained around 5.67 mg/L. The ultrafilter removed approximately 4.12% of NH_4^+-N, and the average effluent NH_4^+-N decreased to 5.43 mg/L. Subsequently, the effluent NH_4^+-N of TiO_2/UV process changed from 1.05 to 6.23 mg/L with the average 2.51 mg/L, and the NH_4^+-N elimination rate in the TiO_2/UV unit averaged at 53.9%, which was higher than the other process units of the CMP system, because the produced hydroxyl radicals are highly active which can transform NH_4^+-N to N_2 and H_2O. The concentration of NH_4^+-N was only reduced by an average of 0.49% during the treatment of activated charcoal filter, and the average of effluent NH_4^+-N was 2.49 mg/L. Because the pore size of membrane was lower than nanometer size and only the water molecule could cross through, the subsequent-reverse osmosis equipment had well elimination to NH_4^+-N. The effluent NH_4^+-N of advanced RO changed from 0.03 to 0.18 mg/L, with the average concentration 0.06 mg/L and the average elimination rate 97.5%. The effluent NH_4^+-N also fully met the requirements of the class I water quality standards in China (GB3838-2002). These analysis indicated that the NH_4^+-N elimination rates of the security filter and the ultrafilter were very low (< 5%), suggesting that it was inability to remove NH_4^+-N for the membrane interception. Also, activated charcoal filter could not adsorb such small molecule substance effectively, therefore, the NH_4^+-N elimination of the CMP primarily depended on the biological effect.

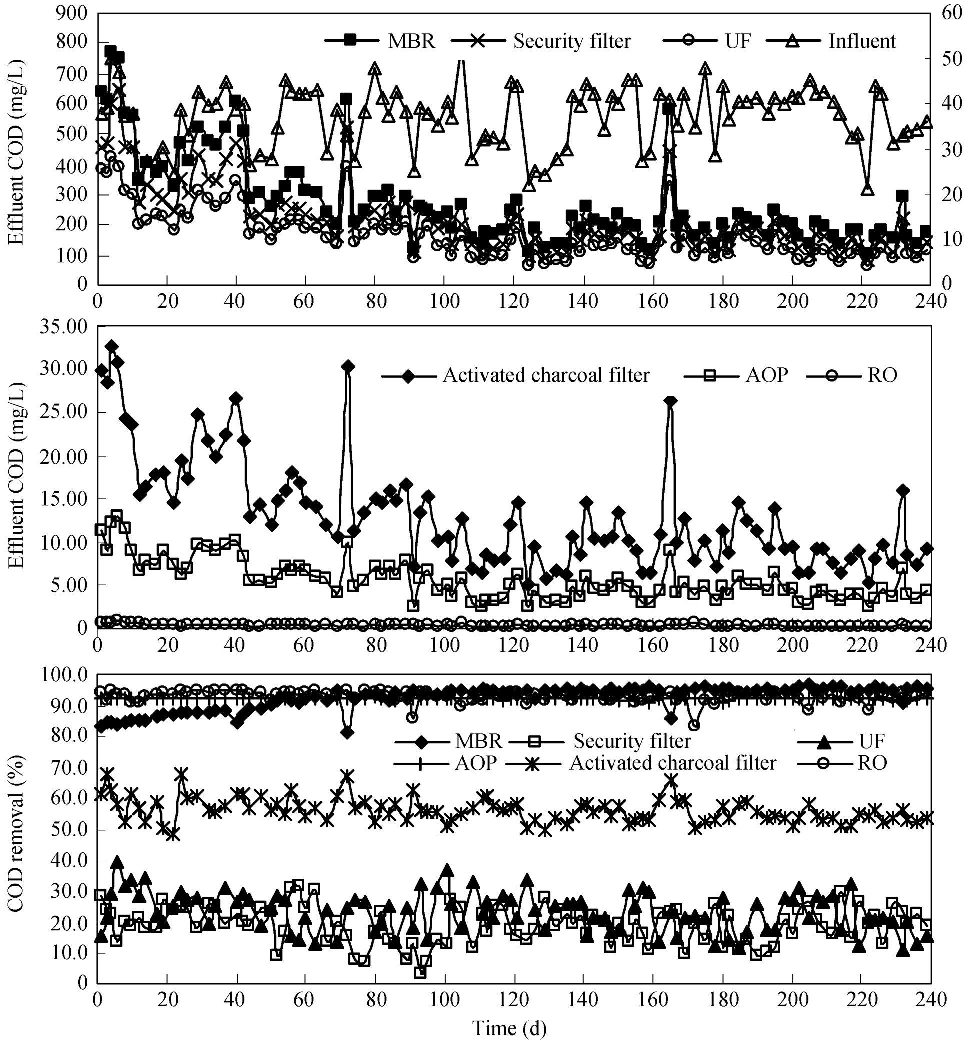

Fig. 2 COD removal in CMP system.

2.3 Effect of CMP on turbidity removal

The turbidity of wastewater can meet the organoleptic requirement, and simultaneously decreased the content of organic compounds and toxic and harmful substances partially. Low turbidity water can make bacterial virus out of protecting function and to be killed much easily in the process of disinfection, hence turbidity was a necessary inspection item of treated water in all previous version of water quality standards, the qualified rate of which must reach 95% in these standards. Therefore, turbidity is essential index for wastewater reuse.

The changes in turbidity removal and effluent turbidity with time are illustrated in Fig. 4. The turbidity of mixed wastewaters was 110.0–181.1 NTU, and the average was 147.0 NTU. The results of trials showed that the average value of MBR effluent turbidity decreased gradually to 5.1 NTU and the turbidity removal rate remained at 96.5%. The interception of microfiltration membrane played a central role in the elimination of turbidity, and the removal efficiency was hardly changed by influent quality, temperature and microbial activity. The security filter significantly reduced turbidity with a removal rate of 46.7%, and the average turbidity effluent remained around 2.7 NTU. The ultrafilter removed approximately 96.5% of turbidity, the average effluent turbidity decreased to 0.0–0.3 NTU. Subsequently, the effluent turbidity of TiO_2/UV process fluctuated from 0.0 to 0.1 NTU which could not be detected basically and was better than the requirements of the above mentioned standards.

2.4 Effect of CMP on LAS removal

Among the different surfactants, anionic surfactants were the most widely used primary surfactants in personal care products. Accurate measurement of these surfactants in wastewater or environmental samples had always been a challenge due to their foaming and peculiar smell (> 0.5 mg/L). The changes in LAS removal and effluent LAS values with time are illustrated in Fig. 5. The LAS concentration in mixed wastewaters was 4.9–5.4 mg/L, and the average was 5.1 mg/L. The result of trials indicated that the average value of MBR effluent LAS decreased gradually to 0.16 mg/L and the LAS removal rate remained at a range of 94.5%–99.4% throughout the experiment. Such high removal rate showed that biological effect played an important role in the elimination of LAS. The LAS elimination rate in the security filter was almost zero. Corresponding to the variation in the influent strength, the

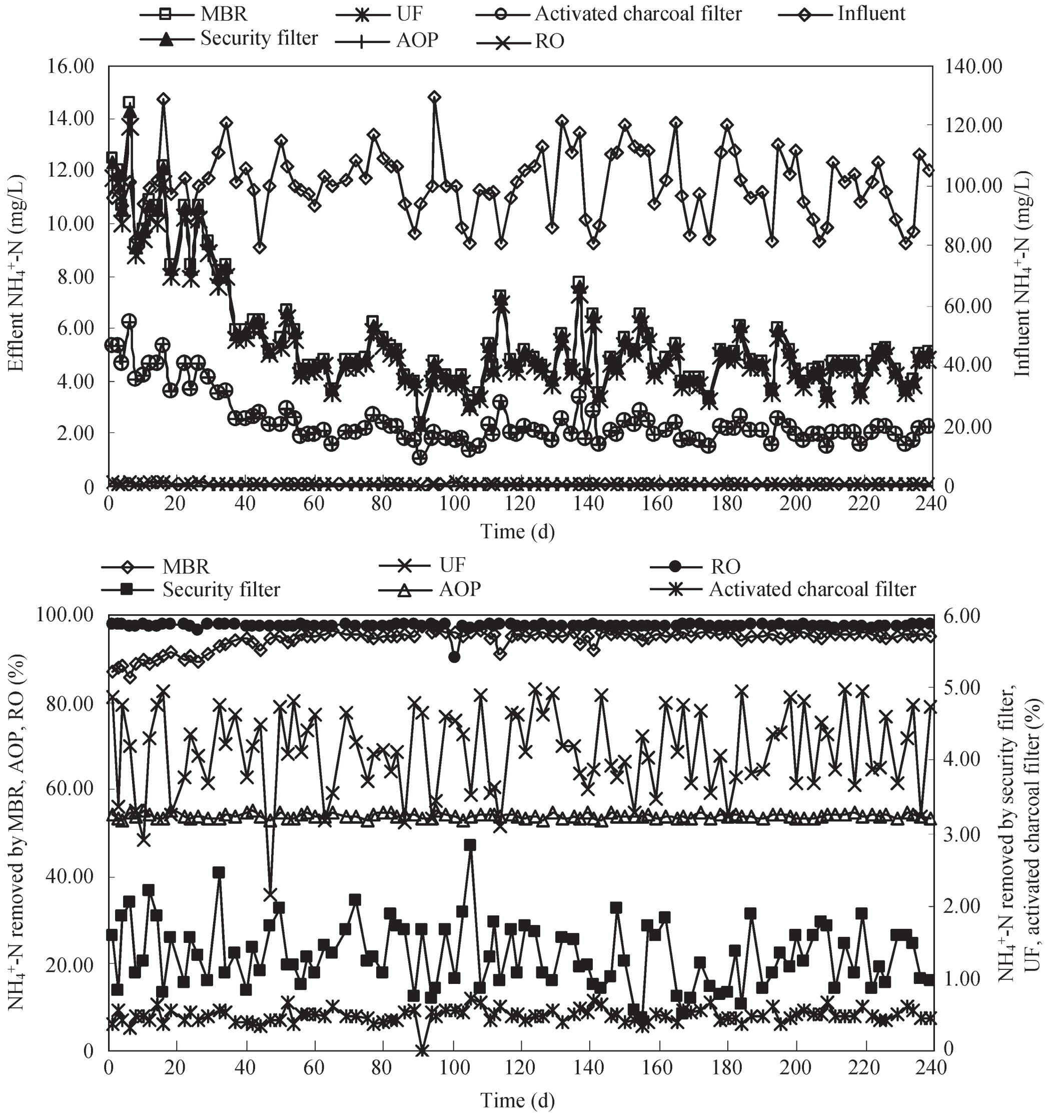

Fig. 3 NH_4^+-N removal in CMP system.

average value of ultrafilter effluent LAS decreased gradually to the range of 0.00–0.26 mg/L, the LAS removal rate was approximately 15.2% from the liquid phase. Advanced oxidation of TiO_2/UV process decomposed LAS to CO_2 and H_2O and eliminated secondary contamination with the LAS removal rate more than 90%. The effluent LAS of the CMP system was 0.0–0.05 mg/L which was better than the requirements of the above mentioned standards.

2.5 Simulation model of CMP system

A successful back propagation neural network (BPNN) requires internal parameters determination such as network architecture and initial weights to meet the required performance (Cigizoglu and Alp, 2006; Ghedira and Bernier, 2004). Finding a suitable architecture and the corresponding weights of the network is a complex task due to the lack of theoretical parameters or optimal values. Arithmetic models were established by using the software MATLAB to simulate the performance of CMP in terms of effluent COD, NH_4^+-N, turbidity and LAS, which are illustrated in the topological architecture of BPNN (Fig. 6). These models are based on the BPNN theory which is composed of a set of elements of calculation (layers) connected to each other and linear regression techniques. Figure 6 shows two nine-level and two five-level networks, and each network comprises input, hidden and output layers. Neurons in the hidden and the output layers calculate their inputs by performing a weighted sum of the outputs that was received from the previous layer. Their outputs however are calculated by transforming their inputs using a transfer function. The model program was debugged and perfected through adjusting node numbers of the hidden layers. The number of the hidden layers is shown in Fig. 6 according to the results of interactive programming in which a satisfactory approximation of the model to the steady-state experimental data between day 121 and day 239 was able to be achieved. In addition, the appropriate number of neurons in hidden layers was chosen to obtain a reliable network.

The most widely used transfer functions are the log-sigmoid (logsig) transfer function (Eq. (1)), the tan-sigmoid (tansig) transfer function (Eq. (2)) and the linear

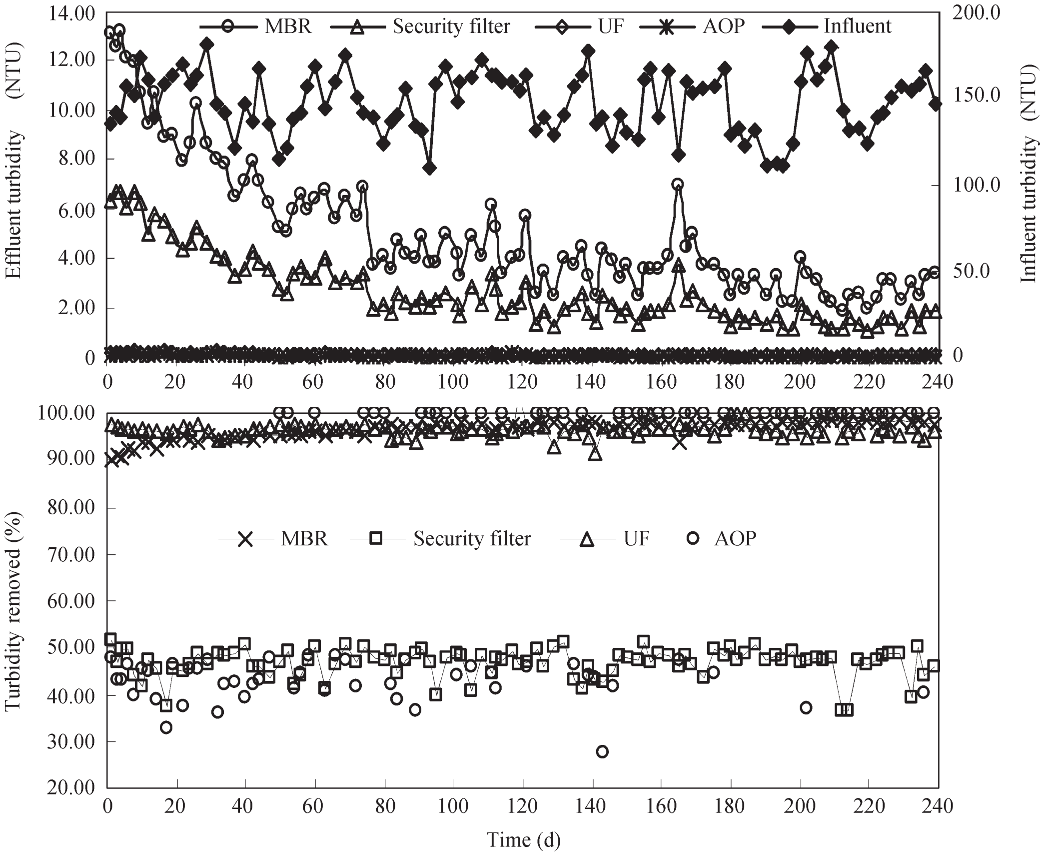

Fig. 4 Turbidity removal in CMP system.

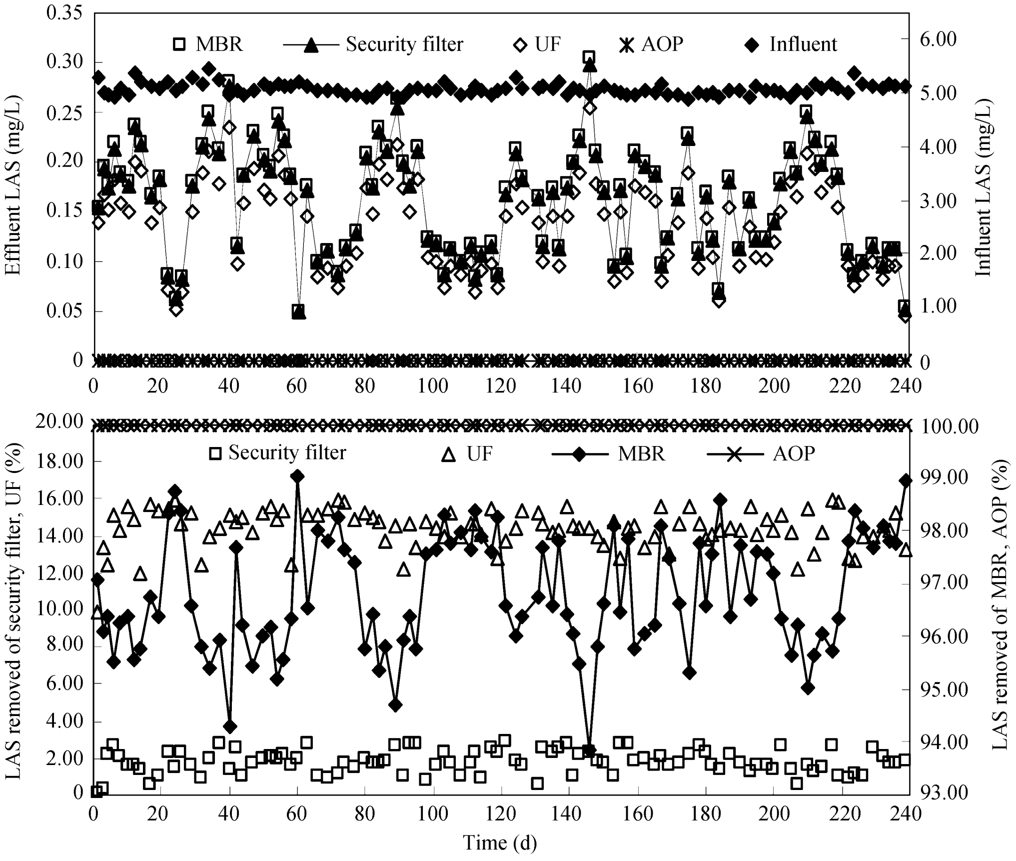

Fig. 5 Anionic surfactants (LAS) removal in CMP system.

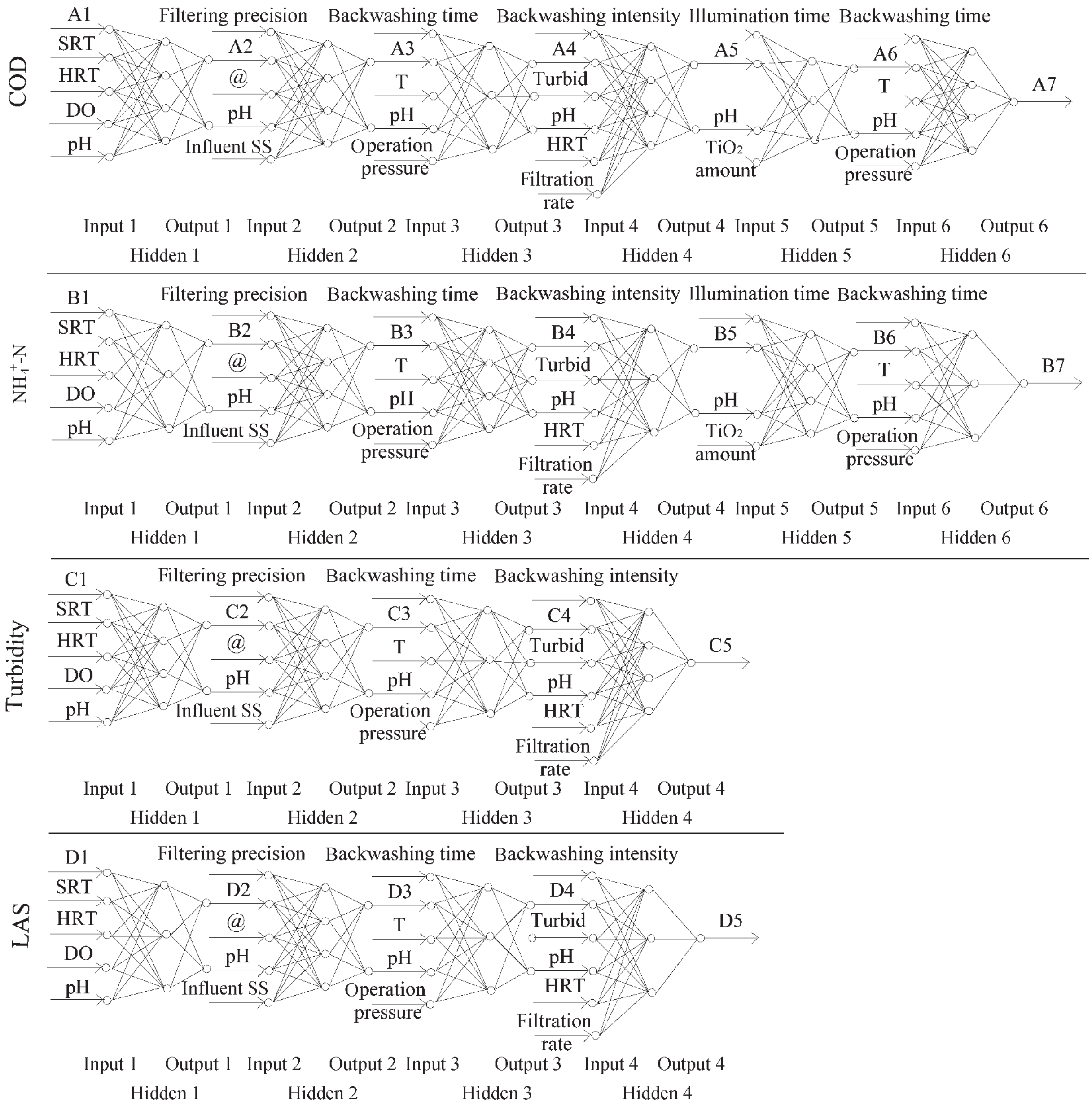

Fig. 6 Topological architecture of back propagation neural network (BPNN) model. @: pressure difference; A1: effluent COD of influent tank; A2: effluent COD of MBR; A3: effluent COD of security filter; A4: effluent COD of ultrafilter; A5: effluent COD of activated charcoal filter; A6: effluent COD of advanced oxidation of TiO_2/UV process; A7: effluent COD of reverse osmosis equipment. B1–B7 are the effluent NH_4^+-N which are comparative to A1–A7; C1–C5 and D1–D5 are the effluent turbidity and LAS, respectively which are comparative to A1–A5, respectively. SRT: sludge retention time; HRT: hydraulic retention time.

(purelin) transfer function. Logsig function produces outputs in the range of 0–1, tansig function produces outputs in the range of –1~+1 and purelin function produces outputs in the range of $-\infty\sim+\infty$ (Delgrange *et al.*, 1998; Niemi *et al.*, 1995). Momentum component (mc) is 0.9; study rate (lr), 0.8; increased coefficient of study rate (lr_inc), 1.05; decreased coefficient of study rate (lr_dec), 0.7; and joint weight, stochastic in [-1, 1].

$$f(x) = \frac{1}{1+e^{-x}} \quad (1)$$

$$f(x) = \frac{e^{x}-e^{-x}}{e^{x}+e^{-x}} \quad (2)$$

The BPNN was trained using selected parameters from data sets between day 1 and day 120 and was subsequently validated using independent data sets between day 121 and day139. The measured data and simulated results are shown in Fig. 7. Overall, the model well fitted the laboratory data in terms of reactors effluent COD, NH_4^+-N, turbidity and LAS. The average error of COD and NH_4^+-N is 5.14% and 6.20% respectively, and the root mean squared error of turbidity and LAS is 2.76% and 1.41%, respectively. This indicated that the simulation model built on the BPNN theory is a feasible and practical means to simulate and predict the pollutants removal by the CMP.

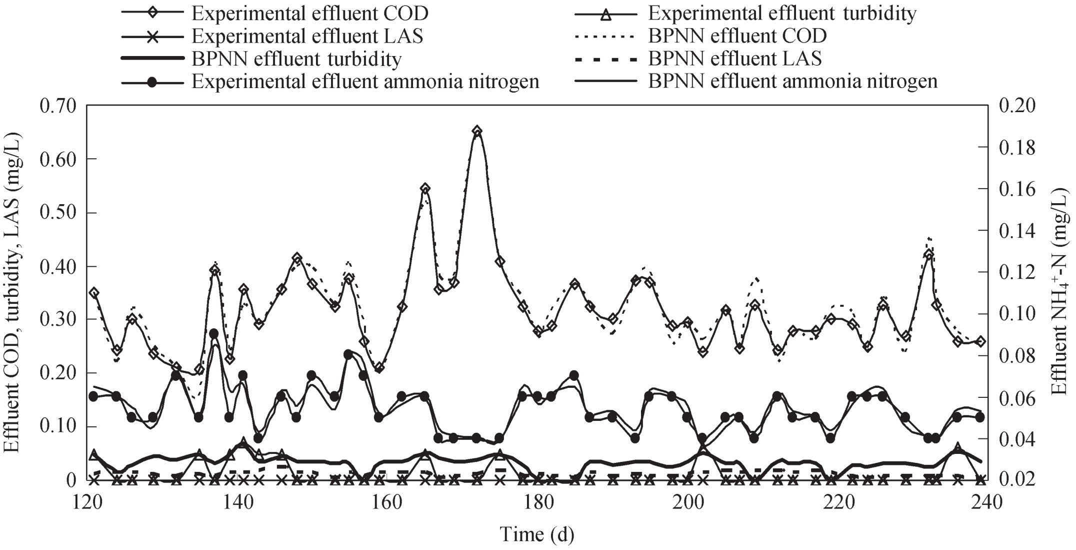

Fig. 7 Comparison of the simulated results with steady state experimental data.

3 Conclusions

A novel CMP system was demonstrated for the treatment and reclamation of mixed wastewater and the following conclusions can be revealed.

(1) The effluent COD, NH_4^+-N, turbidity and LAS of the CMP could fully meet the requirements of the class I water quality standards in China (GB3838-2002), hence the CMP system has application prospects in the treatment and recycling of wastewater in submarine.

(2) MBR hold strong anti-shock loading capability implied its convenience and endurance in practical operation and the security in treated water quality. The elimination of COD, NH_4^+-N, turbidity and LAS were more than 90% throughout the experiment, such high removal rate showed that biological degradation played an indispensable and important role in the field of wastewater treatment and recycling.

(3) Advanced oxidation of TiO_2/UV process had an obvious effect on the removal of pollutants, the elimination rate of COD, NH_4^+-N, turbidity and LAS were 92%, 53%, 46% and 90%, respectively.

(4) Artificial neural network can be a useful tool for CMP system to simulate the process performance. The developed BPNN produced high reliability with the average error of COD 5.14% and NH_4^+-N 6.20% and the root mean squared error of turbidity 2.76% and LAS 1.41%. This indicated that the simulation model built on the BPNN theory is a feasible and practical means to simulate and predict the pollutants removal by the CMP system.

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