Journal of Hazardous Materials 189 (2011) 69–75

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Effect of trace amounts of polyacrylamide (PAM) on long-term performance of activated sludge

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ARTICLE INFO

Article history:
Received 8 December 2010
Received in revised form 29 January 2011
Accepted 31 January 2011
Available online 1 March 2011

Keywords:
PAM
Activated sludge
Performance
SBR

ABSTRACT

This study aims at evaluating the impacts of PAM addition on activated sludge performance. Four lab-scale sequencing batch reactors (SBRs), each with a working volume of 3 L, were investigated with different PAM concentrations. Experiments were conducted with varying organic loading rate and the sludge volume index (SVI), particle size, zeta potential, specific oxygen uptake rate (SOUR), mixed liquor suspended solids (MLSS), COD and ammonium removal efficiency were monitored over a 105-day period. The results showed that all of the PAM addition not only improved the removal efficiencies of COD and ammonium, but also exhibited some advantages on sludge performance. It was found that the sludge performance of settling property, flocculation and microbial activity increased with increasing concentration of PAM. However, high level of PAM (1 mg/L) led to the formation of large amounts of loose-structure flocs, which eliminated dissolved oxygen transfer and caused the sludge disintegration, resulting in bad settleability and lower microbial activity. In this way, when the dosage of PAM was 0.1 mg/L, the sludge had the best settling property and activity.

1. Introduction

Activated sludge process, one of the ways of biological treatment, is commonly used in domestic wastewater treatment, or in the secondary treatment of the industrial wastewater, and over 90% of the municipal wastewater treatment plants use it as the core part of the treatment process [1,2]. The basic function of activated sludge process is to convert organics to carbon dioxide, water and bacterial cells [3]. Nowadays, the discharge wastewater is more complex in nature and cause harm to human's health [4]. In general, coagulation/flocculation process is used as a pretreatment prior to biological treatment in order to enhance biodegradability of the wastewater during the biological treatment [5]. The mechanism of applying coagulation/flocculation treatment is generally to remove the colloidal matter present in the wastewater and aggregate small particles into larger sized ones [6]. The compounds such as ferric chloride, aluminium chloride and/or polymer are common flocculates used in water and wastewater treatment [7]. PAM is a generic name for thousands of polymers containing acrylamide as the major constituent. These polymers have different charges (anionic, cationic, or neutral), charge densities, and molecular weights and they are amorphous and water soluble [8]. Among them, the cationic species of PAM is mostly used in activated sludge process. The flocculating mechanism of cationic PAM contains charge neutralization and bridging. On one hand, the sludge particles in water usually form anionic species, thus, charge neutralization occurs by the adsorption of the PAM chains on to the particle surface, where the cationic PAM will locally reverse the particle surface charge. On the other hand, PAM is a high molecular weight polymer, which could adsorb onto the surface of sludge particles with its long-chains where the tails and loops are extended far beyond its surface and can interact with other particles via bridging flocculation [9,10]. Based on the above mechanisms, domestic water and wastewater treatment uses PAM as a flocculate to pre-settle influent, to thicken and dewater sewage sludge [11]. However, PAM and its derivatives, pose a number of environmental problems as the intermediate products of their degradation are hazardous as their monomer is highly toxic [12,13]. The effluent of activated sludge treatment inevitably contains lower concentration of PAM, so investigating the effect of trace amounts of PAM on activated sludge performance has great significance.

The objective of the research described in this manuscript is to understand the effect of trace amounts of PAM supplementation upon sludge performances in SBR. The hypothesis is that trace

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doi:10.1016/j.jhazmat.2011.01.115

Table 1
Composition of the synthetic wastewater used in this study. Values are in mg/L except the pH.

Component	Concentration	Component	Concentration
$(NH_4)_2SO_4$	566	$FeSO_4·7H_2O$	10
KH_2PO_4	25	$ZnSO_4·7H_2O$	4.4
$KHCO_3$	125	$CoCl_2·6H_2O$	3.2
$CaCl_2·2H_2O$	300	$MnCl_2·4H_2O$	10.2
$MgSO_4$	200	$CuSO_4·5H_2O$	3.2
$NiCl_2·6H_2O$	19	EDTA	6.25
H_3BO_3	6	Glucose	625
pH	7.0–7.8		

amounts of PAM addition in SBR treating would enhance reactor performance and positively impact sludge properties manifested in terms of better settleability, increased sludge activity, etc. To the best of our knowledge, there are no published studies on the effect of trace amounts of PAM addition on sludge properties in SBR operated with organic synthetic wastewater and landfill leachate before.

2. Materials and methods

2.1. PAM and inoculums

Commercial PAM, used was Chemfloc YC250, obtained from No. 2 Sewage Treatment Plant (Changsha, China). The polymer was cationic, with high charge density and a molecular weight of 8–10 million Da. The PAM used in this experiment was added as a solution at the concentration of 0.5 g/L. The solution was prepared per 24 h because it was easy for hydrolysis. The activated sludge from the Heimifeng Landfill Leachate Treatment Plant (Changsha, China) was used to seed the reactors. The initial total suspended solid (TSS) content in the reactors was 2.097 g ss/L.

2.2. Synthetic wastewater and raw leachate

The composition of the synthetic wastewater [14] used in this experimental was described in Table 1. The synthetic wastewater contains glucose, ammonium sulfate, potassium dihydrogen orthophosphate and trace nutrients, which has COD of 580–600 mg/L, BOD_5 of 520–540 mg/L and total nitrogen (T-N) of 130–150 mg/L. The synthetic wastewater used in this study does not contain any colloidal and suspended particles. It contains only soluble organic matter. The raw leachate used in this experimental was supplied from the Heimifeng municipal solid waste (MSW) sanitation landfill site (Changsha, China) per 10 days, and conserved at 4 °C. The average values of the principal chemical compounds concentration were summarized in Table 2.

2.3. Experimental set-up and operation

Four identical SBR reactors were operated in parallel where reactor $1^{\#}$ used as a reference (without PAM), reactors $2^{\#}$, $3^{\#}$, $4^{\#}$ were added the PAM with concentrations of 0.01, 0.1, 1.0 mg/L, respectively. The experimental reactor with its instrumentation and control system is schematized in Fig. 1. The working volume of the reactor was 3 L, with an internal diameter of 10 cm and a height of 40 cm. A complete mixture was achieved during the filling and reaction phases with a mechanical stirrer at the speed of 100 r/min. Dissolved oxygen (DO) was controlled within 2.0–4.0 mg/L during all aeration reaction stages utilized an air-compressor and three micropore aerators supplied air. The pH inside the reactor was initially controlled at a maximum set-point value between 7.3 and 6.8, adding 0.5 mol/L Na_2CO_3 or 1 mol/L HCl. An auto control system consisted of an interface card (PCL-812 PG, Advantech, USA) and two probes (one reads pH, other one reads DO and temperature) was utilized to carry out the real-time control of the thermostatic system, the aeration system and the pH system.

Table 2
Characteristics of the raw landfill leachate from the Heimifeng municipal wastes landfill site of Changsha city. Values are in mg/L except the pH.

Compound	Average ± S.D.	Compound	Average ± S.D.
COD	3876 ± 661	NO_2^--N	0
BOD_5	548 ± 236	TKN	2018 ± 512.3
NH_4^+-N	1451 ± 417	Alkalinity	9618 ± 3502
NO_3^--N	0	pH	7.67 ± 0.53

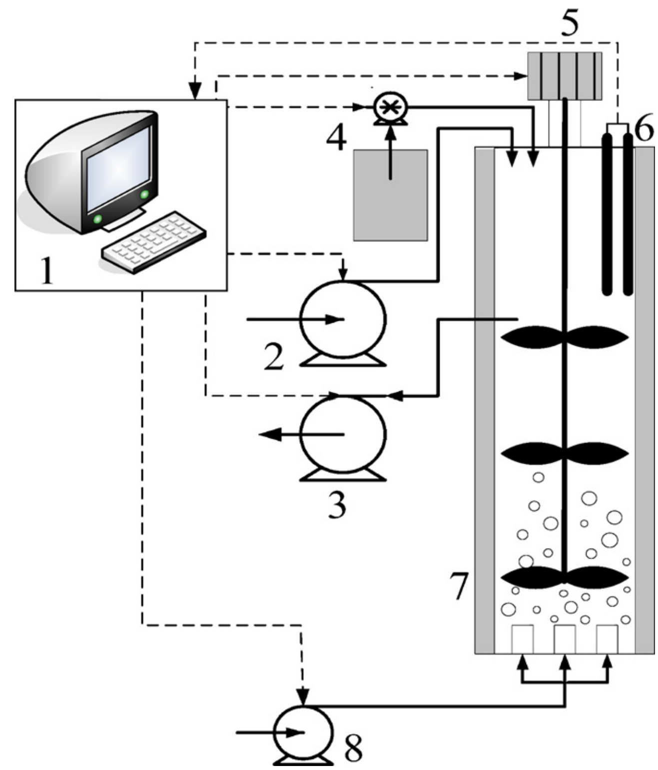

Fig. 1. Schematic representation of the 3 L lab-scale SBR (1. control system; 2. influent pump; 3. effluent pump; 4. pH controller; 5. stirrer; 6. probes (pH, DO, T); 7. jacketed SBR; 8. air compressor).

The experiment was divided into two periods. In period I, the reactors were fed with the organic synthetic wastewater as described in Table 1. Over this period, the organic loading rate was stepped up periodically by increasing the exchange volume (as equivalent feed of the classical continuous supply systems), ranging from 150 mL to 750 mL. The main purpose of this period was to study different concentrations of PAM on the performance of activated sludge. When the exchange volume was maintained as 750 mL, the PAM was absent for 10 days and then recovered. During this period, the characteristics of sludge effect by PAM were detected and compared.

In period II, the reactors were fed with a mixture of organic synthetic wastewater (the exchange volume of organic synthetic wastewater was always maintained at 750 mL) and landfill leachate from the Heimifeng Landfill Leachate Treatment Plant (Changsha, China), while the landfill leachate volume was increased from 25 mL to 250 mL. The main purpose of this period was to study the integrated effect of trace amounts of PAM and landfill leachate on the performance of activated sludge.

The entire study was carried out on a 12 h operational cycle, consisting of four phases: (1) feeding phase, which was supposed to occur in an instant; (2) reaction phase, which contained four

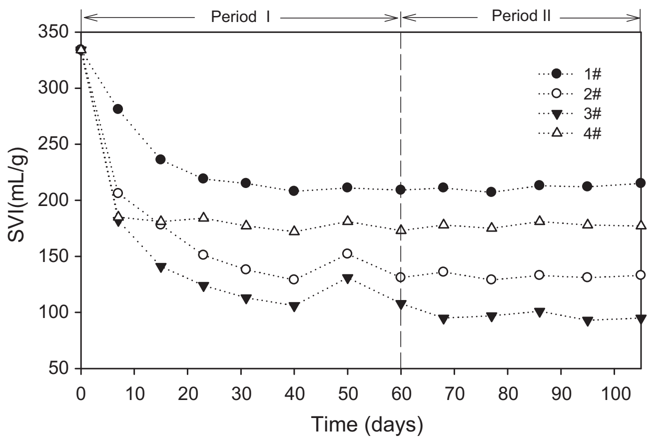

Fig. 2. SVI in the reactors.

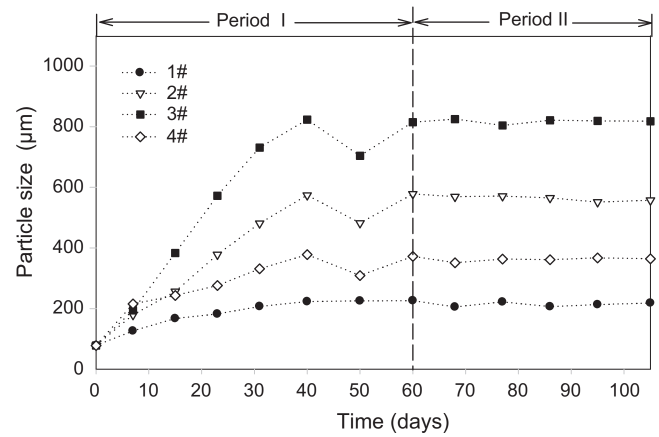

Fig. 3. Sludge particle size in the reactors.

aeration stages and four anaerobic stages running alternately for 2 h and 1 h, respectively; (3) settling phase, which was simultaneously carried out with the last anaerobic stage of the reaction phase; and (4) drawing phase, which was considered to occur in an instant like fill phase.

Moreover, all reactors were measured for the concentration of COD and ammonium over a 12 h cycle to observe the removal efficiency. The sludge volume index (SVI), particle size, zeta potential, specific oxygen uptake rate (SOUR) and mixed liquor suspended solids (MLSS) content were monitored at specific times (as mentioned in the sections when influent load rate changed).

2.4. Chemical analysis

DO and pH were measured by a DO meter (HI9143, Hanna, Italy) and a pH meter (pH meter pen, Lida, China), respectively. The SOUR was calculated using the following equation:

$$\text{SOUR}(\text{mg O}_2\,\text{g SS h}) = -60\frac{G}{X}$$

where G is the slope of the linear portion of the DO decline curve in mg/L min and X is the SS concentration in g/L.

Ammonium nitrogen (NH_4^+-N), COD, mixed liquor suspended solid (MLSS), and sludge volume index (SVI) were analyzed according to the standard methods for the examination of water and wastewater [15]. Particle size of sludge was analyzed with laser diffraction technique (Mastersizer, Malvern 2000, UK). The zeta potential of sludge was measured using a Zetasizer 3000 (Malvern Instruments Ltd., UK).

3. Results and discussion

3.1. Sludge settleability and particle size

Sludge settleability was determined by measuring the sludge volume index (SVI), which is the volume of MLSS after 30 min of settling [6]. The SVIs measured during the experiments are displayed in Fig. 2. Without PAM addition, the SVI decreased slowly. On the other hand, the use of PAM improved the settleability of the activated sludge in SBRs and lower SVIs were observed. After 7 day, reactor $3^{\#}$ and reactor $4^{\#}$ showed the lower SVI compared to reactor $1^{\#}$ and reactor $2^{\#}$, which were 183, 185, 206, 281 mL/g, respectively. After 15 days, the SVI of reactor $3^{\#}$ showed the lowest value (141 mL/g), followed by reactor $2^{\#}$ (178 mL/g), reactor $4^{\#}$ (181 mL/g) and reactor $1^{\#}$ (236 mL/g). In the later 16–40 days, the SVIs of reactor $3^{\#}$ and reactor $2^{\#}$ dropped dramatically and maintained at relatively constant values around 106 and 130 mL/g respectively in case of 0.1 and 0.01 mg/L PAM addition. Nevertheless, the SVIs of reactor $4^{\#}$ and reactor $1^{\#}$ dropped slightly and the values were around 175 and 210 mL/g, respectively. In reference to sludge settleability improvement by PAM, Wong et al. [16] reported that the use of PAM improved the sludge settling characteristics. The SVI value of cationic PAM was less than 30 mL/g in reactor to treat the pulp and paper mill wastewaters. The SVI values in this study were higher compared to the reported values. This might be due to the different treatment method and operational parameters used in the experiment.

There was a sudden rise in SVIs for all PAM addition reactors on the 50th day. The increase was due to the PAM was absent during that period. After few days with PAM added, the SVI recovered. This further demonstrated that the use of PAM can decrease SVI thereby improving the settleability of the activated sludge. During period II the reactors were fed with a mixture of organic synthetic wastewater and landfill leachate. As can be seen from the results, there was only marginal difference between the SVIs with period I and period II. Thus, this indicated that the reactors still present the good settleability even after adding the landfill leachate.

To further understand the sludge settleability of the reactors, the sludge particle sizes with a volume mean diameter (VMD) were measured. According to the results listed in Fig. 3, after 7 days, with no PAM added, the sludge particle size reached a VMD of 126 μm. When PAM was added at 0.01 mg/L, sludge particle size reached a VMD of 179 μm. At the highest PAM dose of 1 mg/L, the sludge grew to a VMD of 215 μm. There was a clear difference in the size of the sludge between PAM and nonPAM-dosed systems with sludge formed with PAM being consistently larger and a general trend for higher polymer doses to have larger flocs. However, in the later period, higher concentration of PAM (1 mg/L) led to the formation of large amounts of loose-structure flocs, which caused the sludge disintegration and small particle size can be seen (378 μm). Meanwhile, the particle size of reactor $3^{\#}$ and reactor $2^{\#}$ increased significantly with time and reached up to 823 and 574 μm, respectively. Zhe et al. [17] draw a similar conclusion when they investigated the effect of cationic PAM on the floc size. They reported that flocs size induced by cationic PAM with C448 and C498 increased to 731 μm and 796 μm, respectively.

Also, there was the similar tendency during the period of PAM was absent. The particle size gained a sudden drop during that period and then recovered after a period of stable PAM addition. During period II, the reactors still presented the stable particle size after adding the landfill leachate. Since sludge settleability was positively influenced by increased particle size [18], the lower SVIs were achieved by PAM addition could be owing to its ability of increasing particle size.

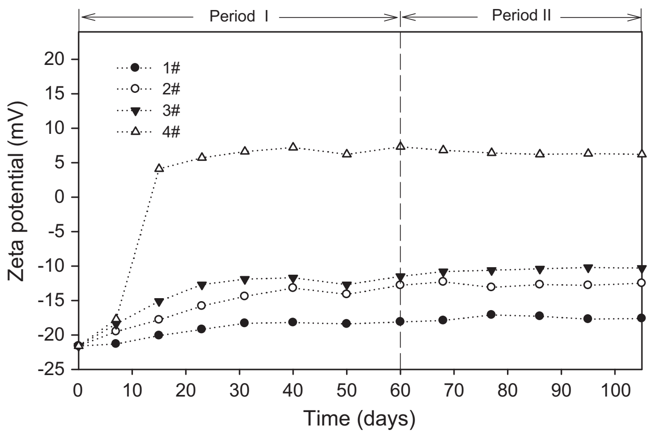

Fig. 4. Zeta potential in the reactors.

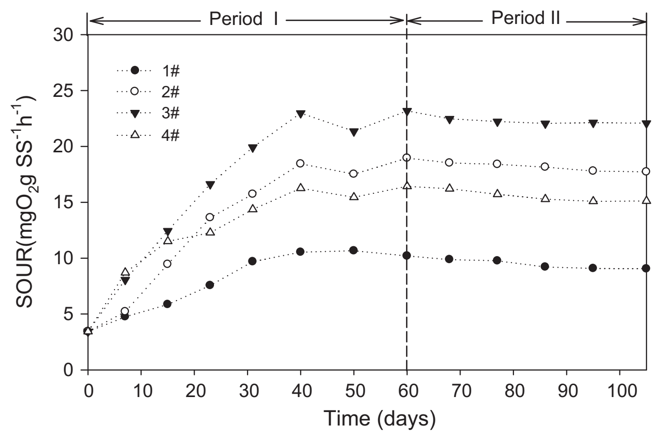

Fig. 5. SOUR in the reactors.

In the present study, it suggested that the use of PAM could decrease SVI and increased particle size thereby improved sludge settling property. The optimal dosage of PAM was 0.1 mg/L, and higher concentration of PAM (1 mg/L) might have negative effects on sludge settleability.

Flocculants with high molecular weights are usually long enough and contains a sufficient number of free functional groups, which can act as bridges, bringing many suspended solids together and building larger flocs [19,20]. The PAM, used in this study, was a high molecular weight polymer (molecular weight was 8–10 million Da). Thus, the bridging flocculation occurred with its long-chains where the tails and loops are extended far beyond its surface and can interact with other particles [9,10]. On the other hand, the PAM used in the experiment was cationic. The role of cationic PAM is to increase the adsorption of flocculants on the surface of sludge particles by diminishing the negativity of the charged particles. However, higher amount of the PAM may lead to competition during bridging between the flocculants and adverse effect on the surface of sludge particle. The effectiveness of flocculants adsorption depends on the amount of polymer adsorbed per unit area of the surface of the particles [21]. But the surface of particle is insufficient for flocculants adsorption when creating high competition amongst the PAM [8]. At low concentration of PAM, the adsorbent site for particle adhesion is available. The mechanism of charge neutralization was dealt with in the following section.

3.2. Zeta potential

Fig. 4 summarized the zeta potential with different concentrations of PAM together with the values of control SBR. Prior to starting-up, the sludge particles have moderate negative charge and corresponding zeta potential (−21.6 mV). As observed, the zeta potential increased quickly after PAM addition compared to nonpolymer-dosed system. After 7 days, the zeta potential showed the values of −19.5 to −17.7 mV from 0.01 to 1 mg/L PAM, while the nonpolymer-dosed system reached a zeta potential at −21.3 mV. After that, the zeta potential increased gradually with time and reached up to −13.2, −11.7, and +7.2 mV, respectively in case of 0.01, 0.1, and 1 mg/L PAM addition by the end of period I. In contrast, the zeta potential of reactor $1^{\#}$ increased slightly and gained the relatively constant value around −18.2 mV.

In the case of cationic polymer, the optimum flocculation was linked to the reduction of the magnitude of the zeta potential to zero by charge neutralization [22]. In general, the optimum regions of −10 mV and +3 mV were recommended for flocculation performance whereby the particles were stable [23,24]. In our present study, the zeta potential of reactor $3^{\#}$ was nearest to optimum flocculation region, which implied best flocculation. Reactor $2^{\#}$, with lower concentration of PAM (0.01 mg/L), showed a lowly increase in zeta potential. Higher concentration of PAM led to the excessive increase in zeta potential.

By the end of period I, there was a sudden decrease in zeta potential and then recovered. This was ascribed to that the PAM was absent during that period. After few days with PAM added, the zeta potential recovered. During the period II, the zeta potential got a little higher than period I. This was due to the landfill leachate which had toxic substances such as heavy metals (Cu, Cr, Zn, Ni, etc.) could cause charge neutralization to the sludge particle surface.

The above information pointed towards a correlation between zeta potential and various sludge properties. The use of PAM could result in zeta potential close to the optimum flocculation region thereby improved sludge flocculation property. The optimal dosage of PAM was 0.1 mg/L, and higher concentration of PAM (1 mg/L) might have negative effects on sludge flocculation. Similar behavior has been reported by Patience et al. [23], who found the zeta potential of flocs increased with the increasing of cationic PAM concentration. The sludge particles in water usually form anionic species, thus the electrostatic attraction occurred between the negatively charged particles and the positively charged PAM adsorption on the particles [25,26]. However, higher concentration of cationic PAM might lead to adverse effect where the surface of particle became positively charge. When this phenomenon occurred, bridging of particle during flocculation was assumed to difficult. This was because flocculation process was improved due to a decline in the charge density by the supplied cations, leading to inter-particle bridging between sludge particles [27]. Low concentration of PAM might affect the charge neutralization process due to the surface of particle was negatively charged, the cationic PAM was not being able to absorb on the surface of particle [8]. It was suggested that repulsive force might have occurred. The polymer chain might not be able to overcome the force and failed to extend the chain far enough to attach on the particle surface. Optimize concentration of PAM could cause the surface of the particles to obtain enough charge to bridge with other particles.

3.3. Specific oxygen uptake rate (SOUR)

Specific oxygen uptake rate (SOUR) tests of mixed liquor had been conducted periodically in order to investigate the impact of PAM on microbial activity and oxygen transfer. Fig. 5 presents the SOUR in four SBR reactors. The SOUR increased with time. All of the PAM addition resulted in a higher increase (3.43–22.97 mg O_2 g SS^{-1} h^{-1}) as compared to that of no flocculant addition (3.43–10.53 mg O_2 g SS^{-1} h^{-1}), which indicated

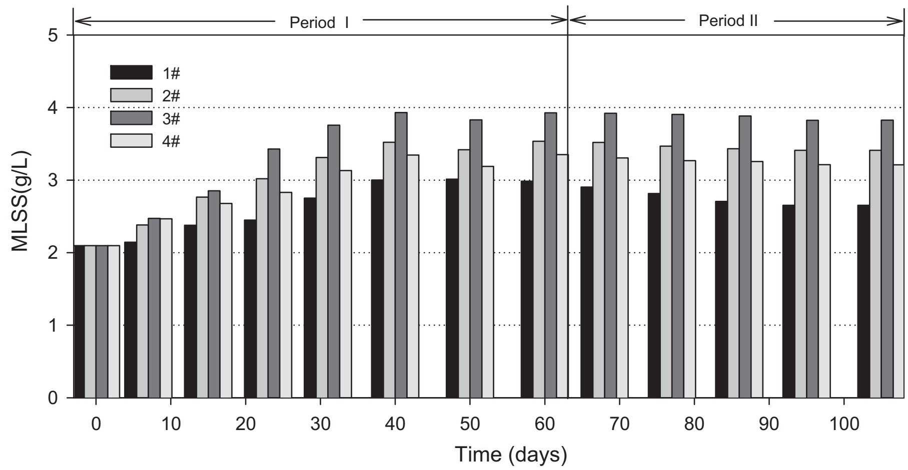

Fig. 6. MLSS in the reactors.

PAM addition led to a high oxygen transfer. The SOUR values in this study (3.43–22.97 mg O_2 g SS^{-1} h^{-1} is equivalent to 4.29–28.71 mg O_2 g VSS^{-1} h^{-1}) were relatively higher than those reported in the literature [28] for PAC addition to treat high strength wastewater (3.3–12.7 mg O_2 g VSS^{-1} h^{-1}).

As observed, after 7 days, the SOUR in reactor $3^{\#}$ and reactor $4^{\#}$ was higher than reactor $1^{\#}$ and reactor $2^{\#}$. However, after 15 days, the values of SOUR were as follows: reactor $3^{\#}$ > reactor $2^{\#}$ > reactor $4^{\#}$ > reactor $1^{\#}$. These indicated that the optimal dosage of PAM to improve SOUR was 0.1 mg/L (reactor $3^{\#}$). On day 40, the PAM was absent for 10 days, a lower SOUR value was observed during this period. However, after a period of stable PAM addition, the system recovered its performance. This phenomenon further demonstrated that PAM addition enhances microbial activity. A previous study showed that the PAM, which had the chemical characteristics of specific chains can stabilize particles fixation, builds composite materials [29]. The rough surface provided by the PAM for microbial growth, thereby improved the microbial activity and oxygen transfer. However, the charge reversal and competition during bridging between the flocculants that caused flocs breakup as well as eliminated dissolved oxygen when there was excessive or overdosing of PAM. During period II, SBR was initially fed with a mixture of organic synthetic wastewater and landfill leachate, and the SOUR values in all reactors showed a slight decrease compared to period I. The microbial population in activated sludge was a heterogeneous community, the landfill leachate which had toxic substances such as heavy metals (Cu, Cr, Zn, Ni, etc.) and certain compounds such as volatile fatty acid inhibited the sludge activity [30–32].

It was also observed that the concentration of MLSS increasing steeply in all of the PAM addition compared to nonpolymer-dosed system (Fig. 6). After 47 days operation, by the end of period I, the MLSS content in reactor $3^{\#}$ was maintain in 3930 mg/L followed by reactor $2^{\#}$ (3423 mg/L), reactor $4^{\#}$(3289 mg/L), and reactor $1^{\#}$ (3015 mg/L). Also, the MLSS content showed a similar tendency as SOUR during the period II. This observation was in agreement with improved sludge activity and oxygen transfer upon PAM addition.

3.4. Removal of COD

After inoculation, the reactors were fed with the organic synthetic wastewater in period I. The organic loading rate was stepped up by increasing the exchange volume, ranging from 0.015 to 0.295 kg COD m^{-3} day^{-1}. Then the exchange volume of organic synthetic wastewater was maintained at 750 mL, while the landfill leachate was increased from 25 mL to 250 mL in period II. The organic loading rate was ranging from 0.36 to 0.96 kg COD m^{-3} day^{-1} during this period.

The variation in COD removal is shown in Fig. 7. The results indicated that dosing PAM significantly increased the COD removal. After 40 days, the efficiencies of COD removal were as follows: reactor $3^{\#}$ (97.03%) > reactor $2^{\#}$ (94.67%) > reactor $4^{\#}$ (94.31%) > reactor $1^{\#}$ (91.84%). An interesting observation was the slight decrease of COD removal efficiency for systems dosed with PAM during the period PAM was absent. Nevertheless, after a period of stable influent PAM, the system recovered its performance. The explanation for this observation was linked to adding the PAM which could result in higher COD removal. After 105 days of operation, by the end of period II, the SBR was operated with 250 mL landfill leachate and an organic loading rate of 0.96 kg COD m^{-3} day^{-1}. Under such conditions, the reactor still presented stable behavior, which reached high COD efficiency. These results are better than those of Ref. [16], who found that 93% of COD reduction after cationic PAM used in the treatment of pulp and paper mill wastewaters.

Based on the results mentioned above, the PAM addition showed two main advantages: it improved the removal efficiency of COD compared to nonpolymer-dosed system, and the optimal dosage of PAM was 0.1 mg/L, which meant higher concentration of PAM (1 mg/L) might have some negative effects on organic (COD) removal. For the high influent COD, PAM was still effective for landfill leachate treatment, and the reactor presented stable behavior. In particular, the higher COD removal efficiency observed with PAM addition correlated well with it could improve microbial activity and oxygen transfer (Section 3.1 SOUR).

3.5. Removal of ammonium

Fig. 8 shows the ammonium removal efficiency in two periods. In period I, the influent ammonium load was ranging from 0.012 to 0.06 kg N m^{-3} d^{-1}. In the first two weeks, it could be observed that the removals with and without PAM addition were comparable. The efficiency obtained was achieved more than 98%. After 40 days of operation, the removal efficiency showed the 79.53% (no PAM) and 85.37%, 91.23%, and 84.87%, respectively (for 0.1, 0.01, and 1.0 concentration of PAM). The implication was that dosing 0.1 mg/L PAM showed best efficiency to ammonium removal. Also, there was a similar tendency for the efficiency of the ammonium removal as COD removal in the same stage. During the period II, the reactors were treated with high influent ammonium load (0.12–0.40 kg N m^{-3} d^{-1}) with organic synthetic wastewater and landfill leachate. As presented in Fig. 7, the removal efficiency of ammonium in period II decreased significantly compared to

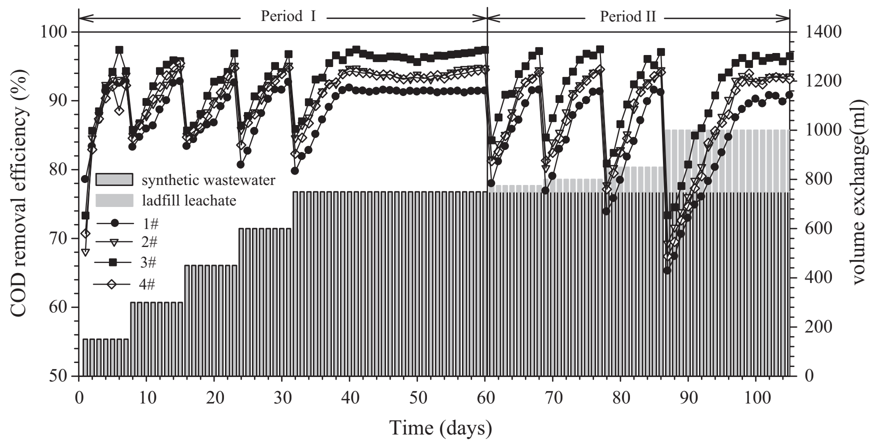

Fig. 7. COD removal efficiency in the reactors.

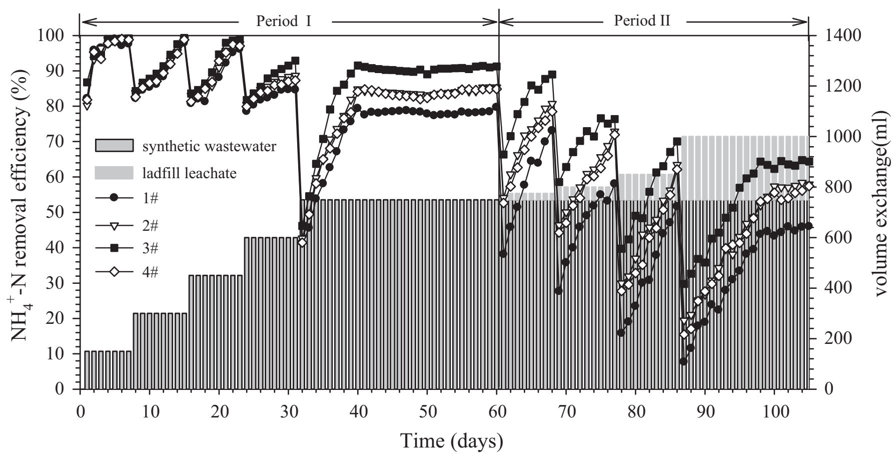

Fig. 8. Ammonium removal efficiency in the reactors.

start-up period. By the end of period II, it could be seen that the poor NH_4^+-N removal efficiency was evident with reactor $3^{\#}$ (64.35%) followed by reactor $2^{\#}$ (57.94%), reactor $4^{\#}$(57.42%), and reactor $1^{\#}$ (45.96%). Aguilar et al. [6] also observed that the efficiency of the removal of ammonia nitrogen was very low (4–17%) in the coagulation–flocculation process which used $Fe_2(SO_4)_3$, $Al_2(SO_4)_3$ and polyaluminium chloride as coagulants.

These results indicated that the PAM addition gives positive effect on ammonium removal only when the influent load was low, and the optimal dosage of PAM was 0.1 mg/L. For the high influent ammonium, adding PAM is not effective for landfill leachate treatment.

In our present study, the pH was always within the range of 6.8–7.3 for all reactors. Under these conditions the NH_4^+ ion would predominate. It would either form a part of salt dissolved in the water or it might be joined by electrostatic attraction to the surface of the negatively charged colloidal particles [33]. The removal of these particles by coagulation–flocculation in turn caused the reduction of ammonia nitrogen associated to them. However, the capacity of PAM to remove ammonia from landfill leachate was limited, which was due to high NH_4^+-N concentration in the influent caused inhibition of the nitrification step [34].

4. Conclusions

The study examined the effect of trace amounts of PAM supplementation on the performance of activated sludge in SBRs through long-term experiments. The results indicated that all the PAM addition resulted in higher COD and ammonium removal efficiency compared to nonpolymer-dosed system. After adding the landfill leachate, the reactors still represented stable behavior to removal COD, whereas the capacity of PAM to remove ammonium was limited. This was due to high NH_4^+-N concentration in the influent caused inhibition of the nitrification step. In this study, the PAM addition achieved lower SVI values and higher SOUR values thereby implied better settleability and high oxygen transfer. The sludge particle size, which increased with time, appeared to be dependent of PAM addition. However, high level of PAM (1 mg/L) resulted in bad settleability and lower microbial activity, which was ascribed to both of competition during bridging between PAMs and huge electronic exclusion. In this way, when the dosage of PAM was in the middle concentration (0.1 mg/L), the sludge had the best settling property and activity.

Acknowledgments

We are grateful for the financial support of the National Natural Science Foundation of China (50478053 and 30970105), the Program for Changjiang Scholars and Innovative Research Team in University (IRT0719), the Xiangjiang Water Environmental Pollution Control Project subjected to the National Key Science and Technology Project for Water Environmental Pollution Control (2009ZX07212-001-02 and 2009ZX07212-001-06) and the National Basic Research Program (973 Program) (No. 2005CB724203).

References

[1] M. Gavrilescu, M. Macoveanu, Process engineering in biological aerobic wastewater treatment, Acta Biotechnol. 19 (1999) 111–145.

[2] B. Saziye, Comparison between alternating aerobic–anoxic and conventional activated sludge systems, Water Res. 41 (2007) 2220–2228.

[3] Y. Liu, Chemically reduced excess sludge production in the activated sludge process, Chemosphere 50 (2003) 1–7.

[4] T.A. Özbelge, Ö.H. Özbelge, S.Z. Başkaya, Removal of phenolic compounds from rubber–textile wastewaters by physico-chemical methods, Chem. Eng. Prog. 41 (2006) 719–730.

[5] O.S. Amuda, I.A. Amoo, O. Ajayi, Performance optimization of coagulant/flocculant in the treatment of wastewater from a beverage industry, J. Hazard. Mater. 129 (2006) 69–72.

[6] M.I. Aguilar, J. Sáez, M. Lloréns, Nutrient removal and sludge production in the coagulation–flocculation process, Water Res. 36 (2002) 2910–2919.

[7] O.S. Amuda, I.A. Amoo, Coagulation/flocculation process and sludge conditioning in beverage industrial wastewater treatment, J. Hazard. Mater. 141 (2007) 778–783.

[8] Y.C. Ho, I. Norli, F.M. Abbas, N. Morad, Alkarkhi, Characterization of biopolymeric flocculant (pectin) and organic synthetic flocculant (PAM): a comparative study on treatment and optimization in kaolin suspension, Bioresour. Technol. 101 (2010) 1166–1174.

[9] T. Li, Z. Zhu, D. Wang, C. Yao, H. Tang, Characterization of floc size, strength and structure under various coagulation mechanisms, Powder Technol. 168 (2006) 104–110.

[10] M.S. Nasser, A.E. James, The effect of polyacrylamide charge density and molecular weight on the flocculation and sedimentation behaviour of kaolinite suspensions, Sep. Purif. Technol. 52 (2006) 241–252.

[11] Stichting Toegepast Onderzoek Waterbeheer, An Investigation into the Environmental Impact of Polyelectrolytes in Waste Water Treatment Plants, Stichting Toegepast Onderzoek Waterbeheer, Utrecht, Netherlands, 1995, pp. 95–17.

[12] C. Rudén, Acrylamide and cancer risk-expert risk assessments and the public debate, J. Food Chem. Toxicol. 42 (2004) 335–349.

[13] E. Campos, M. Almirall, J. Mtnez-Almela, J. Palatsi, Feasibility study of the anaerobic digestion of dewatered pig slurry by means of polyacrylamide, Bioresour. Technol. 99 (2008) 387–395.

[14] Z.Y. Xu, G.M. Zeng, Z.H. Yang, Y. Xiao, M. Cao, H.S. Sun, L.-L. Ji, Y. Chen, Biological treatment of landfill leachate with the integration of partial nitrification, anaerobic ammonium oxidation and heterotrophic denitrification, Bioresour. Technol. 101 (2009) 79–86.

[15] APHA, Standard Methods for the Examination of Water and Wastewater, American Public Health Association, New York, 1995.

[16] S.S. Wong, T.T. Teng, A.L. Ahmada, A.Z. uhairi, G. Najafpour, Treatment of pulp and paper mill wastewater by polyacrylamide (PAM) in polymer induced flocculation, J. Hazard. Mater. B135 (2006) 378–388.

[17] Z. Zhe, L. Tao, L. Jiajuan, W. Dongsheng, Y. Chonghua, Characterization of kaolin flocs formed by polyacrylamide as flocculation aids, Int. J. Miner. Process. 91 (2009) 94–99.

[18] R. Henderson, E. Sharp, P. Jarvis, S. Parsons, B. Jefferson, Identifying the linkage between particle characteristics and understanding coagulation performance, Water Sci. Technol. 6 (2006) 31–38.

[19] I.L. Shih, Y.T. Van, L.C. Yeh, H.G. Lin, Y.N. Chang, Production of a biopolymer flocculant from *Bacillus licheniformis* and its flocculation properties, Bioresour. Technol. 78 (2001) 267–272.

[20] Y.D. Yan, S.M. Glover, G.J. Jameson, S. Biggs, The flocculation efficiency of polydisperse polymer flocculants, Int. J. Miner. Process. 73 (2004) 161–175.

[21] M.G. Rasteiro, F.A.P. Garcia, P. Ferreira, A. Blanco, C. Negro, E. Antunes, The use of LDS as a tool to evaluate flocculation mechanisms, Chem. Eng. Process. 47 (2008) 1323–1332.

[22] E. Sharp, P. Jarvis, S.A. Parsons, B. Jefferson, The impact of zeta potential on the physical properties of ferric-NOM flocs, Environ. Sci. Technol. 40 (2006) 3934–3940.

[23] M. Patience, J. Addai-Menash, J. Ralston, Investigation of the effect of polymer type on flocculation, rheology and dewatering behaviour of kaolinite dispersions, Int. J. Miner. Process. 71 (2003) 247–268.

[24] D. Dihang, P. Aimar, J. Kayema, S.N. Koungou, Coagulation and flocculation of laterite suspensions with low levels of aluminium chloride and polyacrylamids, Chem. Eng. Process. 47 (2008) 1509–1519.

[25] K.B. Girma, V. Lorenz, S. Blaurock, F.T. Edelmann, Coordination chemistry of acrylamide, Coord. Chem. Rev. 249 (2005) 1283–1293.

[26] C. Huang, J.R. Pan, Coagulation Approach to Water Treatment. Encyclopedia of Surface and Colloid Science, Marcel Dekker Inc., New York, 2002.

[27] M. Takeda, I. Koizumi, J.H. Matsuoka, M. Hikuma, Factors affecting the activity of a protein bioflocculant produced by *Nocardia amarae*, J. Ferment. Bioeng. 74 (1992) 408–409.

[28] Y. Satyawalia, M. Balakrishnan, Effect of PAC addition on sludge properties in an MBR treating high strength wastewater, Water Res. 43 (2009) 1577–1588.

[29] P. Walker, T. Kelley, Comparison of a static gravity screen-roll press combination separator to a PAM-assisted gravity belt thickener system for swine waste slurry solids separation, Bioresour. Technol. 96 (2005) 571–576.

[30] M.I. Sher, W.B. Arbuckle, Z. Shen, Oxygen uptake rate inhibition with PACT sludge, J. Hazard. Mater. B73 (2000) 129–142.

[31] M. Gutieĭ rrez, J. Etxebarria, L. de las Fuentes, Evaluation of wastewater toxicity: comparative study between Microtox and activated sludge oxygen uptake inhibition, Water Res. 36 (2002) 919–924.

[32] S.A. Ong, P.E. Lim, C.E. Seng, Effects of adsorbents and copper (II) on activated sludge microorganisms and sequencing batch reactor treatment process, J. Hazard. Mater. B103 (2003) 263–277.

[33] Metcalf and Eddy, Inc., Ingenieria de aguas residuales. Tratamiento, vertido y reutilizacion, Mc Graw Hill Interamericana de Espana S.A., Madrid, 1995.

[34] A.C. Anthonisen, R.C. Loehr, T.B.S. Prakasma, E.G. Srinath, Inhibition of nitrification by ammonia and nitrous acid, J. Water Pollut. Control Fed. 48 (1976) 835–852.