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Post-treatment of banknote printing wastewater using polysilicate ferro-aluminum sulfate (PSFA)

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ABSTRACT

In this paper, a new kind of inorganic polymeric flocculant (IPF)—polysilicate ferro-aluminum sulfate (PSFA) was adopted to treat banknote printing wastewater. Effects of flocculants dosage on the colour and Chemical Oxygen Demand (COD) removal were examined. Experiments revealed that maximal colour removal efficiency of 98% and COD removal efficiency of 85% could be achieved at the optimal dosage of 30.33 g/L. And the colour and COD removal results treated by the PSFA flocculant were compared with those treated by aluminum sulfate. Experimental results showed that the most attracting parts of PSFA as compared with that of $Al_2(SO_4)_3$ were: (i) lower COD and colour contained effluents; (ii) less quantity and volume sludge; (iii) better dewatering behaviour and solid–liquid separation flocs; (iv) providing a possibility to eliminate the high labour intensity plate-frame pressure procedure and replace it by ordinary filtration. Therefore, the using of PSFA generally offered a lower cost of operation and maintenance choice to treat banknote printing wastewater as compared to that of $Al_2(SO_4)_3$.

1. Introduction

Banknote printing wastewater is a kind of printing and dyeing wastewater which is one of the most complicated wastewater in manufacturing industry because of its fragmented and heterogeneous character and high in Chemical Oxygen Demand (COD) and colour. Among the conventional methods, biological treatment, chemical coagulation and flocculation, chemical oxidation, and adsorption processes are the most popular ones [1] used to treat dying wastewater. Although biological treatment processes such as aerated lagoons are generally efficient in Biochemical Oxygen Demand (BOD) and Suspended Solid (SS) removal [2], they are ineffective for removing colour from the wastewater because of the low biodegradability of many chemicals and dyes. In a typical coagulation/incineration system, the coagulant requirement is, however, large [3]. Advanced oxidation processes such as ozonation, Ultraviolet (UV) and ozone/UV combined oxidation, photo catalysis (UV/TiO_2), Fenton reactive and ultrasonic oxidation are not economically feasible [4,5], while adsorption processes have the associated cost and difficulty of the regeneration process and a high waste disposal cost.

Besides, banknote printing wastewater contains high concentration of sodium hydroxide [>10 g/L] that can be recycled and reused. Therefore, ultra filtration has been adopted for treatment of wastewater by several banknote printing factories in China. However, ultra filtration, a physical chemistry separation procedure, can produce highly concentrated rejects effluents, which needs post-treatments such as coagulation [6–9], activated carbon absorption [10,11] before entering into municipal sewage treatment system. And the process flow in Nanchang banknote printing factory is illustrated as below (Fig. 1):

The process is as follows: printing wastewater and wash water are the process effluents of banknote printing works which are collected together and follow the treatment of ultrafiltration (UF) for maximum recycling of recovered waters. The permeate stream which contains water and small amount of soluble solutes and free surfactants is recycled and reused in the cleaning operations. Rejects (about 20% of the inlet volume, whose characteristics are presented in the Table 1), containing high concentrations of organic matter, non-biodegradable matter, oil, suspended and dissolved solids and alkalinity, are collected and allowed for coagulation to remove the colour. Precipitate generated from coagulation is incinerated or land filled while the clarified effluents are further treated at a municipal wastewater treatment system.

The colour of the UF rejects effluence is red or purple, depending on the production process. The main environmental impacts of the banknote printing wastewater come from the strong colour and a high concentration of dissolved solids (organic and inorganic materials), and their discharge into river may further aggravate the problem. Due to these factors, the banknote printing industry faces the challenge of balancing the environmental protection, its

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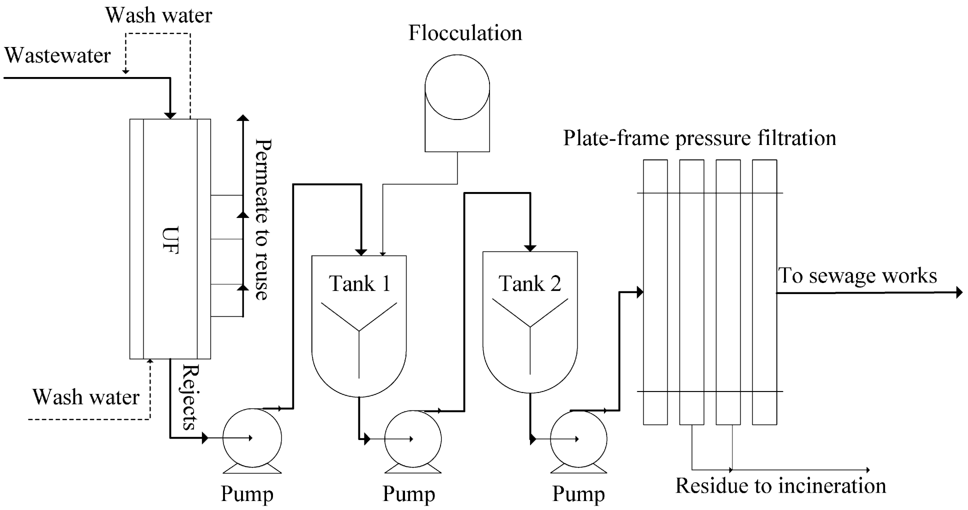

Fig. 1. Schematic diagram of the membrane ultrafiltration, coagulation and plate-frame pressure filtration procedure in Nanchang banknote printing factory.

economic viability and sustainable development. Therefore, these effluents should be treated before discharging.

There are many reports on the use of inorganic polymeric flocculants (IPFs) in water or wastewater treatments. However, few reports have been found on the use of PSFA in banknote printing wastewater treatments. The objective of this research was to investigate the efficiency of coagulation and flocculation processes for the removal of colour and the majority of organic matters from banknote printing UF rejects using a new kind of flocculants—poly-silicate ferro-aluminum sulfate. Flocculating performance comparison between polysilicate aluminum sulfate and other flocculant such as aluminum sulfate was also carried out.

2. Materials

2.1. Wastewater samples

Wastewater used in the experiments was obtained from Nanchang banknote printing factory (Jiangxi province, China), the mean characteristics of which were presented in Table 1. The samples were withdrawn from the end of the ultrafiltration equipment, after concentrated by the ultra filtration membrane and the effluents were used without any dilution. The wastewater was highly coloured and also had high COD and pH values.

2.2. Reagents

All of the chemicals used were of analytical reagent grade. $Na_2SiO_3 \cdot 9H_2O$ (MW 284.22) and $FeSO_4 \cdot 7H_2O$ (MW 278.02) were obtained from Shanghai Yanchen chemical Co, Ltd, China; $Al_2(SO4)_3 \cdot 18H_2O$ (MW 666.41) was purchased from Guangdong Xilong chemical factory, China; 98% sulfuric acid were provided by Hongdu reagent chemical factory, Nanchang, China.

Table 1
Characteristics of UF (ultra filtration) concentrated wastewater from Nanchang banknote printing factory.

Parameter	Value	Discharge standard[a]
COD_{Cr} (mg/L)	79800	100
Colour (times)	1000	180
SS (mg/L)	4900	800
Petroleum (mg/L)	1000	100
Temperature (°C)	45	–
pH	13	6–9

[a] According to integrated wastewater discharge standard (second grade of GB8978-1996, China).

3. Experiments and analytical methods

3.1. Preparation of flocculants

The flocculants used in this study were preparation as follows: firstly, 1.7 g sodium silicates 99.96 g aluminum sulfate and 27.80 g ferric sulfate were mixed in a glass reactor (1 L); secondly, 230 mL 98 wt.% sulfuric acid was added to the reactor, then 230 mL tap water was slowly poured into the reactor and the solution were stirred for a few minutes; finally, the solution were aged for 1.5 h to get the final flocculation PSFA. The flocculation was solid and white or yellow in colour, and its properties do not change for at lest 3 months.

In practical coagulation experiments, 20 wt.% PSFA stock solutions were prepared with tap water (density: 1.0866 g/mL, pH: 0.41). $Al_2(SO_4)_3$ used as a comparison in this study was dissolved in tap water to make a 20 wt.% coagulant solution (density: 1.1238 g/mL, pH: 2.51).

3.2. Coagulation–flocculation test procedures

The raw wastewater (UF rejects or UF concentrated wastewater) was used directly without any pH adjustment. Coagulation–flocculation studies were performed with batch cylinder test cylinder columns. Namely, 50 mL raw water and certain dosage of coagulant were added to a 100 mL of cylinder column and shaken by hand for 5 times.

3.3. Analytical methods

After the Coagulation–flocculation test procedure, the result suspension solution was filtered. Filtration was done under atmospheric pressure using a filtrate cloth provided by Nanchang banknote printing factory to simulate practical solid–liquid separation process.

The filtrate was analyzed for pH, COD, colour, while the filter residues were subjected to moisture content rate and residual yield measurements. The pH was measured by pH-3 meter (Rex, Shanghai Precision Scientific Instrument Co., Ltd, China) while the COD value was determined in accordance with the dichromate method of the Standard Methods for the Examination of Water and Wastewater (China National Standard, GB11914-89). The colour measurement followed the dilution multiple method (Standard Methods for water quality-determination of colourity, GB11903-89, China). The filter residue was collected and weighed ($w1$), then dried in a vacuum oven at 105 °C to constant weight ($w2$), and was analyzed for moisture content [%moisture content = $(w1 - w2)/w1 \times 100\%$] and sludge yield ($w2$). The flocculation performance of PSFA and aluminum sulfate was further compared with column settling test, in which flocculants and wastewater were mixed (shaking by hand for 5 times) and allowed for settling and the rate of mud-line fall (or float) was recorded. All analyses were carried out in triplicate.

4. Results and discussion

4.1. Determination of optimal dosage for chemical coagulation

Removing UF rejects effluence's colour was the main objective of this study. So colour removal efficiency was chosen as the control index to determine the optimal dosage for coagulation. Besides, other parameters such as COD removal and pH value were selected as auxiliary indexes. The flocculants dosage was optimized with respect to the colour removal efficiency. Flocculants dosage was increased from 0 to 8 mL with a fixed amount of wastewater of 50 mL. The reduction or removal of colour and COD efficiencies were calculated from the colour and COD initial concentration in the raw wastewater and final concentration in the filtrate. The results obtained at room temperature (20 °C) and at the UF rejects' outlet temperature (45 °C) were shown in Figs. 2–4.

Fig. 2 showed pH depression during the coagulation procedure. This curves displayed a steady decrease in pH value as the flocculants dosage increased. Similar trend was observed when the raw wastewater was heated to 45 °C.

While Fig. 3 showed that at room temperature, the addition of flocculants led to a significant increase in efficiency. Colour removal reached a plateau of 98% at a flocculant dose of 6 mg/L or above. Further increasing the PFSA dosage only revealed minor impacts on colour removal efficiencies. It could be seen that increasing flocculants dosage does not always improve the reduction or removal rates. From this figure, it could observe that about 98.0% colour was removed at the pH range of 3–5 and the remained colour was 20 times. Fig. 3 also showed that at 45 °C, the curve displayed nearly the same general behaviour as at room temperatures.

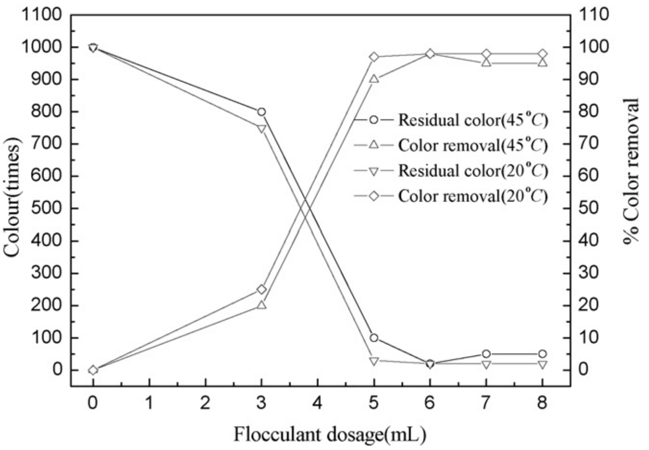

Fig. 3. Colour removal versus flocculant dose of PSFA.

The effect of flocculant addition on COD removal was depicted in Fig. 4, which showed that the COD removal efficiency gradually increased from 0% to 85% with PFSA dose increasing from 0 to 7 mL. Further increasing PFSA dosage only led to poorer COD removal efficiency. Therefore the maximum removal for COD was 85%, and the remained COD was 11,400 mg/L. At 45 °C, residual COD decreased from 11,400 to 7450 mg/L (although COD removal efficiency increased only from 85% to about 90%). This was because at high temperature, both hydrolysis of metal ions in the PSFA and collision of particles in wastewater were accelerated, which facilitated the flocculation process.

In conclusion, the optimum flocculants dosage was chosen as 7/50 mL wastewater (equivalent to 30.33 g PSFA(s) per litre wastewater), since at this dosage a similar level of colour removal efficiency was achieved as compared to higher doses and the residual COD was relatively low. It should be noticed that after treated by PSFA (at room temperature), the effluents still had 20 times colour and about 11,400 mg/L COD, which far surpassed the discharge standards (see Table 1). When lime or NaOH was added to adjust filtration effluents' pH to 7 or above, residual colour disappeared and nearly 100% colour removal was achieved while the COD value remained the same. Thus we conclude that the residual colour may be caused by iron ions. Owing to low pH value (<4), metal ions in the PSFA could not be further hydrolyzed and transformed into hydroxide precipitate. And the remaining COD is believed to be a result of the formation of complex organic compounds known as soluble microbial products which could not remove by coagulation–flocculation method.

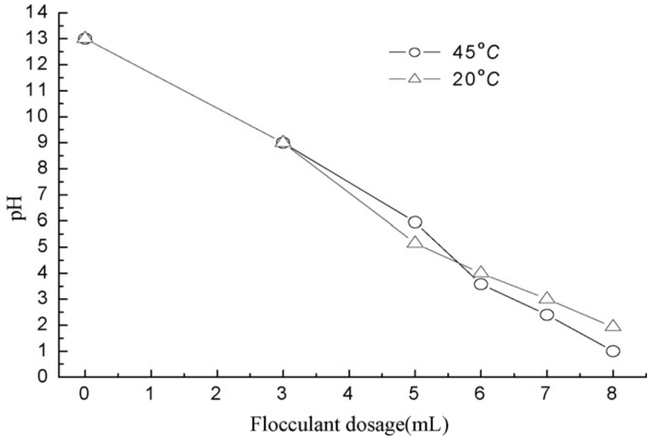

Fig. 2. pH change versus flocculant dose of PSFA.

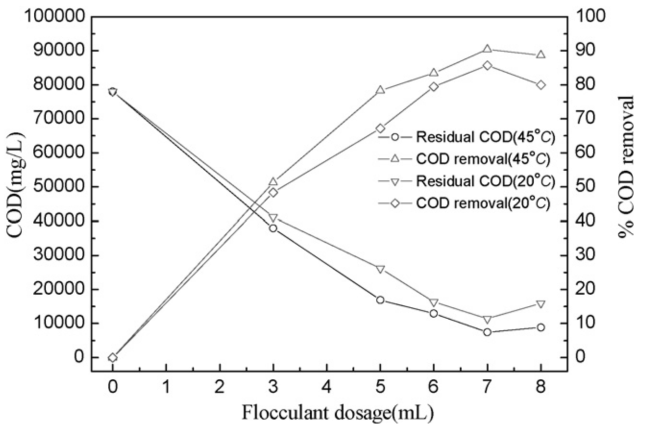

Fig. 4. COD removal versus flocculants dose of PSFA.

Table 2
Rate of moisture content and sludge production using PSFA as coagulant.

Items	Sludge moisture content (%)	Sludge yield (g/L wastewater)
Value	58.5	40.02

4.2. Moisture content and sludge yield

The sludge produced in the physical–chemical treatment is due to the amount of organic matter and total solids in suspension that are removed and the compounds formed from the coagulant used, since practically almost all of the latter will form part of the sludge solids [12].

Table 2 listed the rate of moisture content of sludge (or filter residue) treated by PSFA and total sludge yield.

In general, the characteristics of sludge may affect the whole wastewater treatment cost. Large volume of sludge is hard to transport. Higher moisture rate sludge may also increase the transportation cost and cause problem in subsequently disposal procedure such as incineration because more energy is need to evaporate the moisture when incinerate the sludge. The sludge moisture content in Table 2 showed that the sludge was suitable to incineration and did not need dehydration by plate-frame pressure filtration, which should make the final disposal sludge easier.

4.3. Proposed mechanism for waste removal

There exist few established conclusions as to why IPFs efficiency is superior to the traditional coagulants. The actions and mechanisms of IPFs are not fully understood [13]. It is generally thought that the traditional coagulants and IPFs perform differently due to their features of chemical speciation [13,14].

For alum, hydrolysis species like monomers and $Al(OH)_{3(am)}$ with low positive charge may adhere onto the particle surface [15] to destabilize the waste suspension and then sweep the destabilized particles away by amorphous precipitates.

According to the flocs structure and the flocs settling (or floating) speed differences between PSFA and $Al_2(SO_4)_3$, we believe that the coagulation mechanism of PSFA is probably involving a hydrolysis–polymerization process as follows: first, the metal ions in the PSFA hydrolyze to form polynuclear such as $Al_{13}O_4(OH)_{24}{}^{7+}$ (also reported to range from 3^+ to 7^+, normally simplified as Al_{13})[13]. Then, hydrolysis products are adsorbed onto the waste particles' surface through charge neutralization, chemical bridging and sweep-flocculation to destabilize the waste suspension. Here, the formation of polynuclear is thought to be the main cause to produce flocs of larger size and faster floating velocity. This process can be very complex thus further investigations need to be conducted.

4.4. Comparison between PSFA and $Al_2(SO_4)_3$

To further demonstrate the feasibility of PSFA in treating UF concentrated banknote printing wastewater, a comparison was carried out between PSFA and $Al_2(SO_4)_3$, which was a commonly used coagulant and was currently used by Nanchang banknote printing factory. For $Al_2(SO_4)_3$ as coagulant, lab, experiments had determined that the optimum dosage was 8 mL coagulant per 50 mL wastewater (Equivalent to 36.00 g $Al_2(SO_4)_3$(s) per litre wastewater). Before dosage, the initial pH of wastewater was adjusted to pH 8–9 with H_2SO_4. Table 3 showed the treatment result under optimum dosage by $Al_2(SO_4)_3$. Practical results used in the factory were also showed for comparison. Items in Table 3 demonstrated that lab experimental results were in good agreement with practical results. At optical conditions, $Al_2(SO_4)_3$ has given nearly 97% colour reduction and 69% COD reduction at pH 7.01 and the residual colour and COD were 30 times and 25,200 mg/L, respectively.

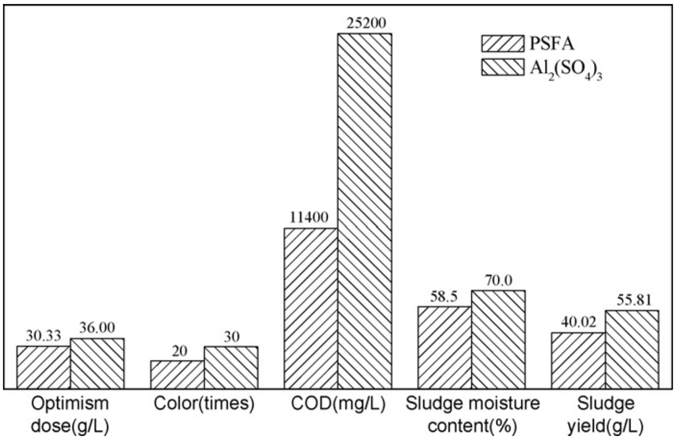

Fig. 5. Comparison of best results obtained using PSFA and $Al_2(SO_4)_3$.

Fig. 5 compared the best results obtained by using PSFA and $Al_2(SO_4)_3$. From this figure, it was evident that PSFA showed an advantage in treatment of banknote printing ultrafiltration concentrated wastewater over $Al_2(SO_4)_3$. Since it required less amounts of coagulants to achieve the optimism results and effluents treated by PSFA contained 11,400 mg/L COD, less than half of that treated by $Al_2(SO_4)_3$. Obviously, the PSFA performed much better than alum in removing COD. No significant difference in colour removal efficiency could be observed between the two kinds of coagulants (or flocculants). PSFA also generated lower moisture content sludge and the sludge production was also found to less than that of $Al_2(SO_4)_3$. Sludge production accounted for 25–65% of the total operating cost of a secondary treatment [12], so PSFA had an advantage in handling and treating the sludge (filter residue) than $Al_2(SO_4)_3$.

In the coagulation–flocculation process, the settling (floating) speed of the sludge formed is important since this will influence the overall cost and efficiency [16]. The settling (floating) performance of between PSFA and $Al_2(SO_4)_3$ was further compared with column settling (floating) test.

Fig. 6 showed the different setting/floating performances between PSFA and $Al_2(SO_4)_3$. In the case of PSFA as flocculant, it was interesting to notice that the sludge floated in less than 1 min (Fig. 7), so the solid–liquid separation time required for the dye removal was very short, which was important for practical wastewater treatment applications; while in the case of $Al_2(SO_4)_3$, the sludge settled very slowly (Fig. 8), the final gravity settling stage lasted for more than 10 h. The sediment volume (denoting by the height of the sludge, since the cross section area of the columns

Table 3
Treatment result under optimum dosage using $Al_2(SO_4)_3$.

No.	Optimism dosage (g/L)	H_2SO_4[a] (g)	Colour (times)	COD (mg/L)	pH	Sludge moisture content (%)	Sludge yield (g/L)
1	36.00	3	30	25200	7.01	70	55.81
2[b]	37.50	3	30	25900	7.09	68	–

[a] For pH adjustment use.

[b] Data provided by Nanchang banknote printing factory.

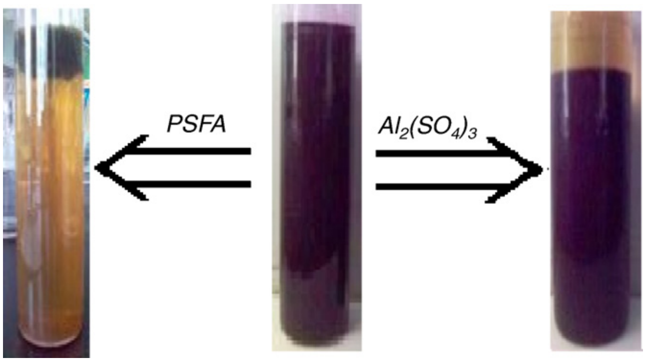

Fig. 6. Photographic response of different treatment result between PSFA (at the settling time of 1 min) and $Al_2(SO_4)_3$ (after 10 h settlement) at optimum dose.

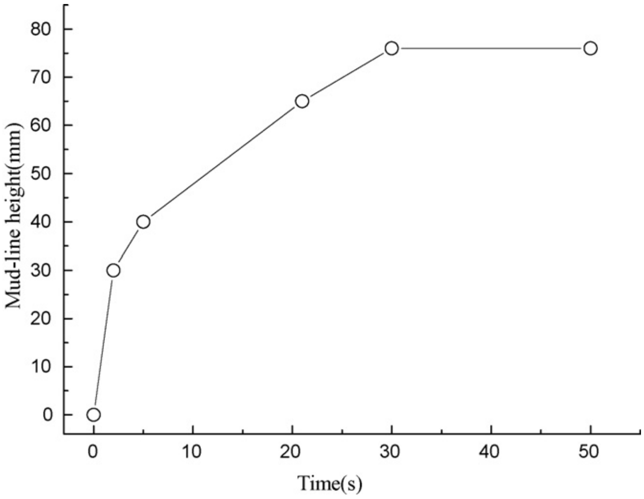

Fig. 7. Sludge settling/floating time of PSFA.

used in the coagulation–flocculation test procedure is the same) of the flocculated sludge of the wastewater treated with PSFA was 1 cm, less than that of $Al_2(SO_4)_3$ (about 5 cm).

The difference in sludge production and sludge settling (or floating) speed was due to the different sludge structure and coagulation mechanism between PSFA and $Al_2(SO_4)_3$. Visual inspection of the sludge (flocs) indicated the formation of small, loose and low density sludge for $Al_2(SO_4)_3$; on the other hand sludge formed using PSFA as flocculant was larger and porous. This could be explained by different features of chemical speciation between traditional coagulants and IPFs [14]. For the traditional coagulants such as alum, the precipitation of aluminum hydroxide acting as charge neutralization species [9], the sludge thus formed had poor sediment behaviours. Whereas for inorganic polymeric flocculants such as PSFA, their coagulant mechanisms were believed to the combination effects of adsorption, chemical bridging, charge neutralization and sweep-flocculation [16,17]. It is believed that destabilization by bridging occurs when segments of a polymer chain absorbed on more than one particle, thereby linking the particles together [18], the sludge formed by this way was larger and porous and were more suitable for solid–liquid separation in sedimentation (floatation).

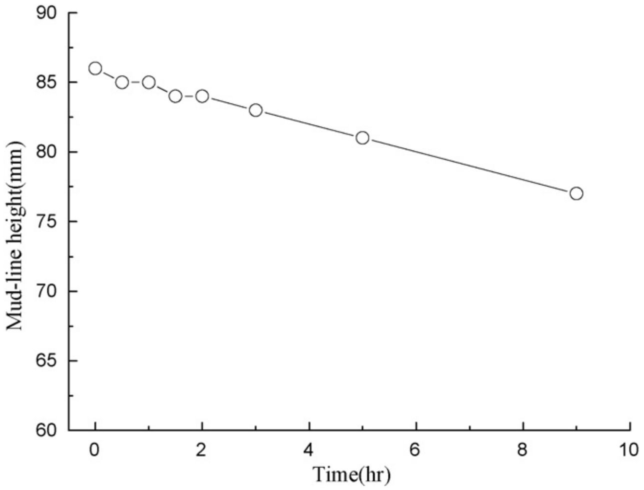

Fig. 8. Sludge settling/floating time of $Al_2(SO_4)_3$.

All in all, taking into account the coagulation–flocculation performance between PSFA and $Al_2(SO_4)_3$, the most attracting parts of PSFA were: (i) lower COD and colour contained effluents; (ii) less quantity and volume of sludge; (iii) better dewatering behaviour and solid–liquid separation flocs; (iv) providing a possibility to eliminate the high labour intensity plate-frame pressure procedure and replace it by ordinary filtration, thereby, this method generally offered lower cost of operation and maintenance as compared to the current used method. Thus, treatment of UF concentrated banknote printing wastewater with PFSA could be considered as competitive to that of $Al_2(SO_4)_3$.

Other advantages include easy dissolved flocculant and no pH adjustment before coagulation. A disadvantage of PSFA was that it might cause more pH depression of the wastewater. As a result, the effluents had very low pH value.

5. Conclusions

The study showed the using of PSFA as a floculant to treat banknote printing ultrafiltration concentrated wastewater was technically feasible and economically viable. The main conclusions from this work were:

(1) Experiments revealed that maximal colour removal efficiency of 98% and COD removal efficiency of 85% could be achieved at the optimal dose of 30.33 g/L. The effluents treated by PSFA had lower residual colour and the COD than that of $Al_2(SO_4)_3$.
(2) In the case of PSFA as flocculant, the sludge floated in less than 1 min and the solid–liquid separation time required for the dye removal was very short compared with that of $Al_2(SO_4)_3$. The sediment sludge volume and the sludge quantity treated by PSFA were less than that of $Al_2(SO_4)_3$. PSFA also produced lower moisture content sludge. All this led to the possibility of eliminating the high labour intensity plate-frame pressure procedure; hence this method generally offers a lower cost of operation and maintenance choice as compared to $Al_2(SO_4)_3$ serve as coagulant.

However, coagulation using PFSA is not good enough to make the treated wastewater meet the requirement of integrated wastewater discharge standard (Second grade of GB 8978-1996, China); therefore, the effluents have to be treated by subsequent treatment units before discharging.

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