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Characterization and performance of poly-silicic-cation coagulant in treatment of recycled paper wastewater

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ABSTRACT

Tertiary treatment of pulp and paper wastewater has become increasingly pressing since stricter legislations were issued by the central government and some provincial governments in China. Coagulation process after biological treatment has been regarded as a techno-economically feasible technology for the high quality effluent. In this paper, a study on preparation and characterization of poly-silicic-cation coagulants (PSiCs) with different Si/(Al + Fe) molar ratios is described. X-ray diffraction, infrared spectra, ultraviolet/visible absorption scanning, and microscopic imaging were used as measures for characterization. Coagulant performance was evaluated using it in papermaking wastewater treatment. The results showed that new complex compounds had formed in the PSiC polymers and the optimal Si/(Al + Fe) molar ratio for the coagulant polymerization was about 0.9, at which the polymerization degree was highest. The content of ionic polymerized bonds and branch-like PSiC units would decrease when the molar ratio was lower or higher than 0.9. The highest efficiency appeared when $PSiC_{0.9}$ was used in the wastewater treatment, rather than $PSiC_{0.6}$ and $PSiC_{1.5}$. The results also showed that concentrations of turbidity, COD, color, and residual aluminum in the PSiC treated effluent were lower than that from coagulation tests using traditional coagulants.

Keywords: Recycled paper wastewater; Poly-silicic-cation coagulant; Coagulation; Characterization of morphology and structure

1. Introduction

Pulp and paper wastewater accounted for 23% of the total COD in all the industries in China in 2012 [1]. The wastewater is also rich in toxic suspended solids and recalcitrant compounds like fatty acids, wood extractives, chlorinated organics, tannins, and lignin and their derivatives [2]. More stringent standards for the wastewater and water pollutant discharge in pulp and paper industry were issued by the central government of China in 2008. For the recycled paper production, which accounted for 65% of the total paper production in China in 2012, the limit value of wastewater discharge has been decreased from 60 to 20 m^3 for one ton of paper production and the allowed COD

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concentration decreased to 80 mg/L according to the new standards [3]. Some provincial government even issued much more stringent standards, e.g. COD discharge limitation for the recycled paper wastewater in Shaanxi Province has been only 50 mg/L since 2011 [4]. In this situation, high efficient tertiary treatment after biological process becomes essential for this kind of wastewater.

Coagulation is normally employed in the tertiary treatment in the case of pulp and paper wastewater treatment. Pokhrel and Viraraghavan [5] have investigated the current technologies for the tertiary treatment of pulp and paper wastewaters. Their study involved coagulation, chemical oxidation, ozonation, adsorption, and membrane filtration techniques for the removal of residual COD, color, chlorinated phenolic compounds, and adsorable organic halides. They concluded that the optimization combination of coagulation and biological treatment process would provide a long-term solution for treatment of effluent from pulp and paper mill. Coagulation with conventional alum, ferric chloride, ferric sulfate, and lime has been widely studied [6,7]. However, as the environmental regulations become increasingly stringent, more efficient and safer approaches for treatment of paper mill wastewater should be developed.

Polymeric coagulants or polyelectrolytes, especially those with high molecular weight, are widely used in recent years, since they are able to produce large, dense, compact, and strong flocs, which can precipitate well, and also able to reduce the sludge volume [8]. Composite polysilicate coagulants (PSCCs), e.g. poly-zinc-silicate-sulfate, poly-aluminum-silicate-chloride, and poly-aluminum-silicate-sulfate developed in 1990s on the basis of poly-silicic acid (PS), aluminum, and iron-containing coagulants, are inorganic polymers with high molecular weight [9–11]. PSCCs are conventionally prepared via composite polymerization and copolymerization. The former means the polymerization between PS and hydroxylated metal salts, and the later means the hydroxylation of the mixture of metal salts and PS [12]. Synchronous polymerization was developed to polymerize the silicate and the hydroxylated metal salts, and to reduce costs [13,14]. However, coagulation optimization has mainly been performed by means of "trial and error" owing to the high complexity of the coagulation process and the variety of available polyelectrolytes [15]. The effectiveness and cost of coagulation depends on the type and concentration of coagulant, solution pH, ionic strength, and the characteristics of the organic residues in the effluent. A study focused on how the characteristics of PSCCs affect the coagulation performance may bring about the improvement of PSCCs preparation process and better choice of coagulants for users.

In this study, poly-silicic-cation coagulants (PSiCs) with different Si/(Al + Fe) molar ratios were prepared by the method of synchronous polymerization, using industrial waste pyrite slag and waste sulfuric acids as raw materials, and characterized by means of X-ray diffraction (XRD), infrared spectra (IR), ultraviolet/visible absorption (UVA) scanning, and microscopic imaging. The performance of end products were studied and evaluated through the treatment of biologically treated wastewater from a recycled paper mill.

2. Materials and methods

2.1. Materials

The industrial wastes included pyrite slag and waste sulfuric acids which were used for the preparation of the coagulants were obtained from Meixian Sulfuric Acid Plant of Shaanxi Province and Xi'an Modern Chemistry Research Institute, respectively.

Effluent out of secondary settler of a wastewater treatment system of Wanlong Paper Mill at Xi'an, which had been biologically treated, was used for coagulation performance test.

2.2. Preparation of PSiC

PSiCs were prepared by the following steps:

(1) *Extraction of the metals from pyrite slag*: 40 g pyrite slag was added into a beaker containing 150 ml waste sulfuric acid with concentration of 50%, then stirred, heated, and kept orderly at a temperature of 80–90°C for 2 h to extract useful metals from the slag. The final mixture was paper filtered. The filtrate was the metal salt solution, which contains about 1.12 mol/L Fe^{3+}, 0.12 mol/L $Al^{3+,}$ and trace amount of the other metals.

(2) *Polymerization of PSiCs*: 49 ml sodium silicate solution was blended with various amounts of the metal salt solution, and then titrated with sodium hydroxide, making the pH be 1.8. After aged at 25°C for 1 d, the final mixture was the PSiC solution.

2.3. Characterization of PSiCs

The PSiC solution was diluted 400 times, and then scanned by a TU-1810 spectrophotometer (Puxi, China). The liquid samples were primarily dried and then analyzed with a D/MAX-RB X-ray diffractometer

Table 1
Qualities of paper mill aerobic effluent

Items	Paper mill aerobic effluent
Turbidity (NTU)	70
COD (mg/L)	120
Chroma (times)	16
pH	7.2

(Rigaku, Japan) for crystalline phases. Then the solid coagulants were measured by a KBr pressed disc with a Tensor37 IR spectrophotometer (Bruker, Germany). The solution samples were dripped onto glass slides and dried at room temperature, and then observed and photographed by a microscopic imaging system (the Fifth Factory of Optical Instruments, China), which consisted of a XSP (2XC) electron microscopy, a complementary metal-oxide semiconductor, and a computer.

2.4. Coagulation performance

The biologically treated effluent (Table 1) used for the study of PSiCs coagulation performance was obtained from the secondary settler of a recycled paper mill (Xi'an Wanlong Paper Mill, China), where a deinking pulping process was used to produce white coating paperboard.

A coagulation tester (ZR4-6, Zhongrun Co. Ltd., China) with six cups, agitators for each cup and a programmer was used for the coagulation performance test. The coagulant solution was added to the wastewater samples, which were then stirred successively at 150 rpm for 2 min and at 30 rpm for 10 min, precipitated for 30 min. The supernatant below the surface 3 cm was collected as sample for further analysis. The turbidity, COD, color, and residual aluminum of the sample were determined by HI93703 (HANNA, Italy) turbidimeter, back titration (GB11984-89, China), colorimetric dilution (GB11903-89, China), and spectrophotometry (GB/T 8538-1995, China), respectively.

3. Results and discussion

3.1. XRD analysis

Fig. 1 illustrated the XRD spectra of the prepared PSiCs with three different Si/(Al + Fe) molar ratios. The primary compounds in PSiCs are shown in Table 2. Clearly, the spectra of diffractive crystals, such as $Fe_2(SO_4)_3$, Fe_2O_3, Fe_3O_4, $Al_2(SO_4)_3$, Al_2O_3, and SiO_2 were not observed in PSiCs, implying that Fe, Al, and Si had been polymerized rather than remained as a simple mixture, which agreed with the suggestions of Tong Sun [16].

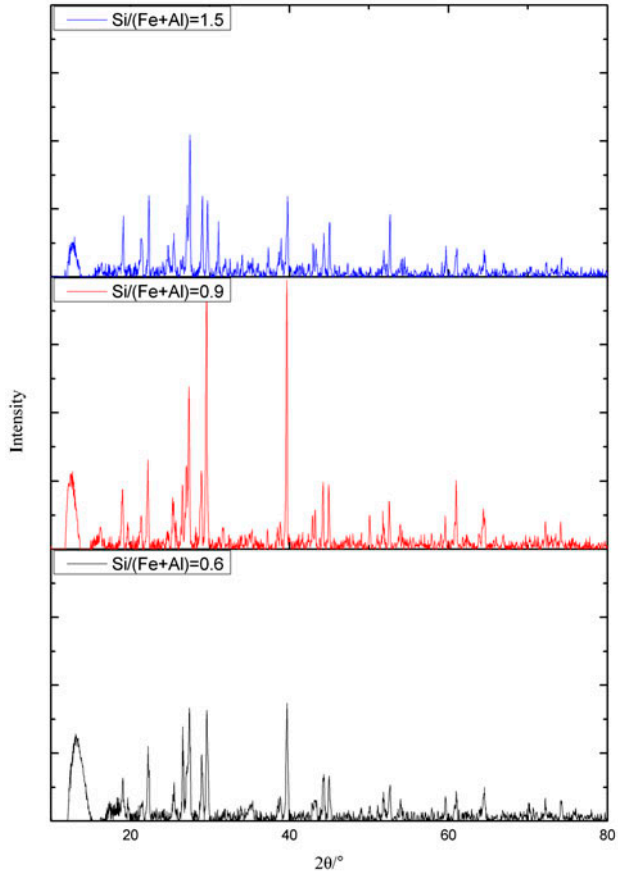

Fig. 1. XRD spectra of PSiC with Si/(Al + Fe) molar ratios of (a) 0.6, (b) 0.9, and (c) 1.5.

Table 2
Primary compounds in PSiC at different Si/(Al + Fe) molar ratios

	Si/(Al + Fe) molar ratios		
Compound	0.6	0.9	1.5
$Mg_2Al_2SiO_5(OH)_4$	√	–	–
$Fe_3Si_2O_5(OH)_4$	√	–	–
$Fe_4Al_4Si_2O_{10}(OH)_8$	–	√	–
AlOOH	–	√	–
$Al_2Si_2O_5(OH)_4$	√	√	–
$Fe_3Fe\ SiO_4(OH)_5$	–	√	√
$CaSO_4$	–	√	–

The XRD spectra also showed some identical and different peaks, suggesting that the same and distinctive compounds were formed in each PSiC. These results implied that Si/(Al + Fe) ratio has some effect on the polymerization and conformation of PSiCs.

Besides, polymerization of Fe, Al, and Si was more complex in PSiC with Si/(Al + Fe) ratio of 0.9 (marked as $PSiC_{0.9}$) than PSiCs with other molar ratios. PSiC with Si/(Al + Fe) ratio of 1.5 ($PSiC_{1.5}$) only contained $Fe_3Fe\ SiO_4(OH)_5$.

3.2. Analysis of IR spectra

The IR spectra of the three solid PSiCs samples are shown in Fig. 2. All spectra exhibited two kinds of bonds at 3,500–3,300 and 1,641–1,638 cm^{-1}, which were assigned to the stretching vibration of –OH [17,18]. The peaks at 2,500–3,100 cm^{-1} were attributed to the vibration of intramolecular chelation –OH [17]. All the three PSiCs showed peaks at 2,605 and 2,489 cm^{-1}, meaning that they all contained intramolecular chelation –OH. Furthermore, the peaks at 1,152–1,179 cm^{-1} corresponded to the asymmetric stretching vibration of Fe–O–Fe or Al–O–Al [16]. The peaks at 1,044 cm^{-1} and around 970 cm^{-1} were attributed to the symmetrical stretching vibration of Si–O–Al and Si–O–Fe, respectively [15,19]. The above bonds in the PSiCs would hydrolyze inevitably to produce hydroxy complex with a single core or multicore silicon, as well as polysilicon fragment with Fe or Al, therefore, they expressed strong ability of charge neutralization, adsorption, and capture during coagulation procedure [19]. The peak at 795–799 cm^{-1} assigned to the connection of Si–O–Si tetrahedron [20] did not present in the spectra of all the three PSiCs. These results suggested that PSiCs had a reticular formation, which was consistent with the morphological analysis. In addition, the peaks at 450–650 cm^{-1} were related to the bending vibration of Fe–OH, Al–OH, Si–O, Fe–O, and Al–O [20]. The intensity of the above peaks primarily increased with the increase in Si/(Al + Fe) ratio, and then decreased when the ratio was more than 0.9. Overall, the IR results implied that Si/(Al + Fe) ratio had an important effect on the structure of the PSiCs.

3.3. Analysis of UVA spectra

The UVA spectra of the three PSiCs are shown in Fig. 3. As mentioned in Section 2.3, the coagulant sample was diluted 400 times with tap water before measured. The peak at 280–320 nm was contributed to Fe^{3+} [20], and the absorption peak at 230 nm was produced by the $Al(OH)_4^-$ electronic transition from the highest (occupied) to the lowest (unoccupied) electron orbit [21]. $Al(OH)_4^-$ is thought to be the precursor of Al_{13}, which has high molecular weight and positive charge and is the most effective component of polyaluminum coagulants [22]. Fe^{3+} or Al^{3+} produced by hydrolysis caused the intensity decrease of characteristic peaks or even disappear. Generally, single ion can hydrolyze easily in oligomer, but ion hydrolysis is difficult for high polymers. The complexation state of

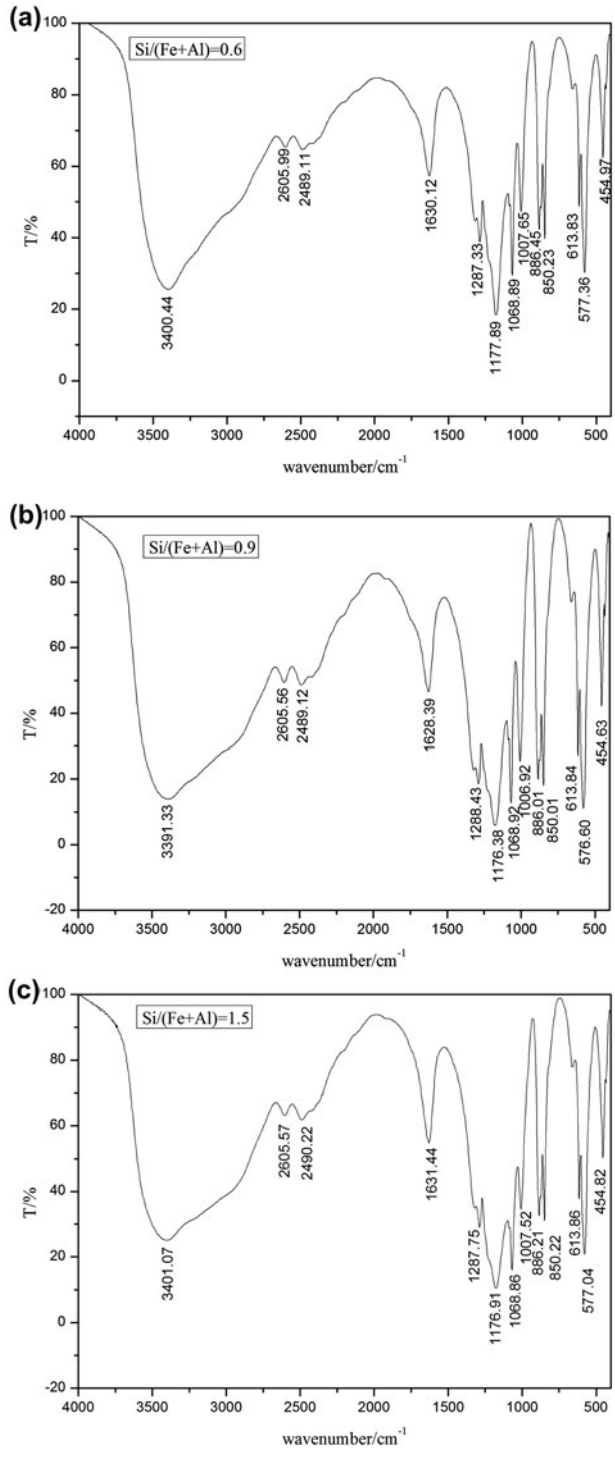

Fig. 2. IR spectra of PSiC with different Si/(Al + Fe) molar ratios of (a) 0.6, (b) 0.9, and (c) 1.5.

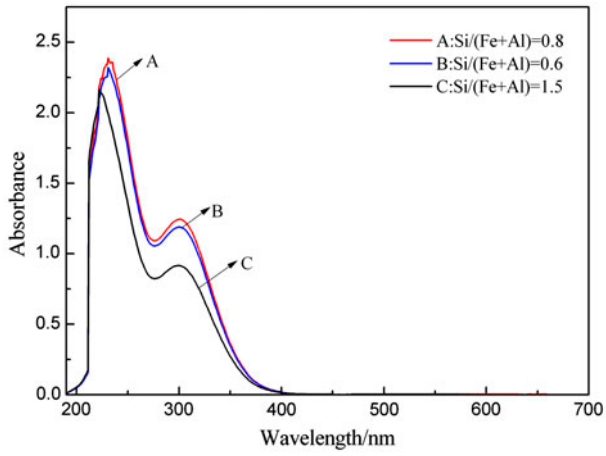

Fig. 3. UVA scanning spectra of PSiC with different Si/(Al + Fe) molar ratios.

Fe^{3+} and Al^{3+} in PSiC can be evaluated qualitatively according to the intensity change of the characteristic peaks in UVA.

Fig. 3 indicated that the intensity of the characteristic peaks would increase with the increasing Si/(Al + Fe) ratio, but would decrease when the ratio was more than 0.9. There were mutual acceleration between Fe–O–Fe and Si–O–Fe, and mutual retardation between Si–O–Fe and Si–O–Si [20]. Therefore, the formation rate of Fe–O–Fe was high and that of Si–O–Si was low, that was why certain amount of metal ions could retard the process of PS gelation [20]. At low Si/(Al + Fe) ratio, Si–O–Fe–O–Fe–O–Si was formed owing to the mutual acceleration, which resulted in slow Fe^{3+} release in the hydrolysis. However, at lower pH, the transference of polymerization–dissociation equilibrium shifted from complexation to dissociation [23], and then the unstable bonds Fe–OH–Fe (or Fe–O–Fe) might break [24]. On the contrary, at high Si/(Al + Fe) ratio, only a small amount of Fe^{3+} participated owing to the mutual retardation between Si–O–Fe and Si–O–Si, and Si–O–Fe–O–Si–O–Si was the primary bond in PSiC.

Al^{3+}, compared with Fe^{3+}, had a similar intensity variation as the characteristic absorption peak (Fig. 3). In Si/(Al + Fe) with molar ratio of 0.9, Al^{3+} was mainly combined with the end-radicals OH or O in PSiC. Since Si–O–Al was stable, Si–O–Al–O–Al–O–Si was prone to polymerization rather than dissociation, and the species would increase in size, which would make the absorbance increase [20]. PSiC with Si/(Al + Fe) molar ratio of 0.6 ($PSiC_{0.6}$) contained more metal salt, which was an acid liquid. So the pH was low and Si–O–Al–O–Al–O–Si may be broken into Si–O–Al due to the transference of polymerization–dissociation equilibrium from complexation to dissociation. However, at high Si/(Al + Fe) molar ratio, Si–O–Si–O–Si–O–Si was the main reaction mode because of the lack of Al^{3+}, and thus the peak of Al^{3+} was low. Therefore, the polymerization between Si and Al can be expressed as Si–O–Al–O–Si–O–Si. Overall, the UVA results supported the statement that PSiC was a complex compound of Si, Fe, and Al rather than a simple mixture.

3.4. Morphological analysis

For the morphological analysis, PSiC solution samples were dripped onto glass slides and then coagulant clusters would continuously grow according to their external structures [25]. The polymerization structures of Fe, Al, and Si were observed and photographed by the microscopic imaging system.

Fig. 4 showed the morphologies of the PS and the PSiC samples. The PS sample contained spherical or spheroidal particles, and the PSiC had some branch-like or even network structures. Those unique structures were horizontally composed of small irregular units, which were fit for the adsorption and aggregation of fine particles. The poly-aluminum-ferric-silicate-sulfate had a chain-net structure, that probably because Fe, Al, and Si had polymerized, and the chain became thick and the hole net got larger with the increase in silicon dose. As shown in Fig. 4, PSiC was nearly circular at low Si/(Al + Fe) molar ratio, and the branch length increased with the increase in silicon dose. That was because, with low metal salt content, Si/(Al + Fe) ratio was also low, branch-like PSiC units became large as cross-copolymerization among Fe, Al, and Si, while at high metal salt content, the three-dimensional polymer of silicate will loosely gather into chain and mesh aggregates. The results agreed with Sun [16]. However, the gelation of the PS would weaken its sweeping abilities. The rate of gelation process was proportional to the silicon dose in the PS [20]. Therefore, in selection of Si/(Al + Fe) ratio for coagulants, both coagulation performance and stability should be considered.

3.5. Coagulation performance

The $PSiC_{0.9}$ was used for the coagulation treatment tests of the biologically treated effluent from a recycled paper mill. Polymeric aluminum chloride (PAC), which was widely regarded as a high-quality coagulant, was used as reference. The results of the coagulation tests are shown in Figs. 5–7. The tests with $PSiC_{0.9}$ showed excellent removal efficiencies of COD, color and turbidity, which were 67, 95, and 99%,

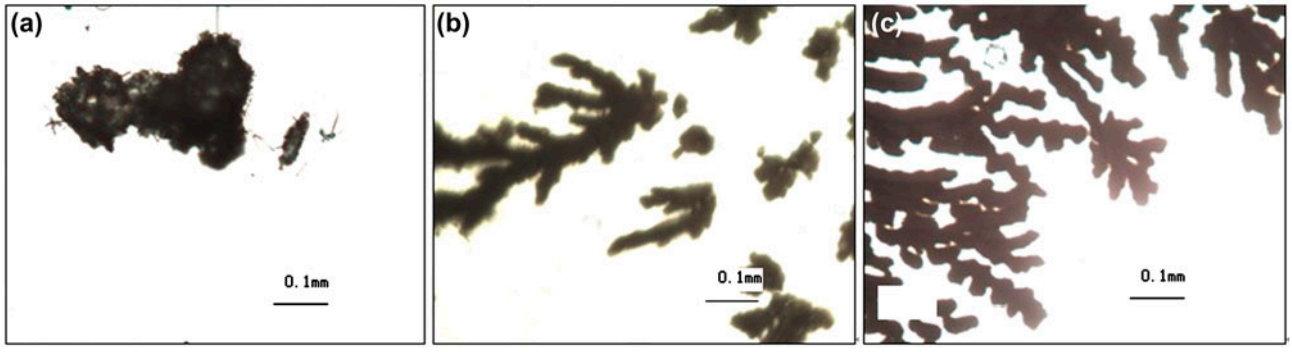

Fig. 4. Morphology of PSiC with Si/(Al + Fe) molar ratios of (a) 0.6, (b) 0.9, and (c) 1.5.

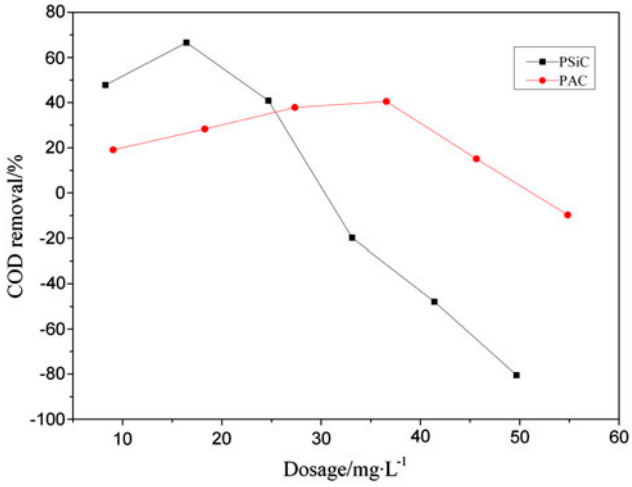

Fig. 5. COD removal of PAC and $PSiC_{0.9}$ with different dosage in the treatment of biologically treated effluent from a recycled paper mill.

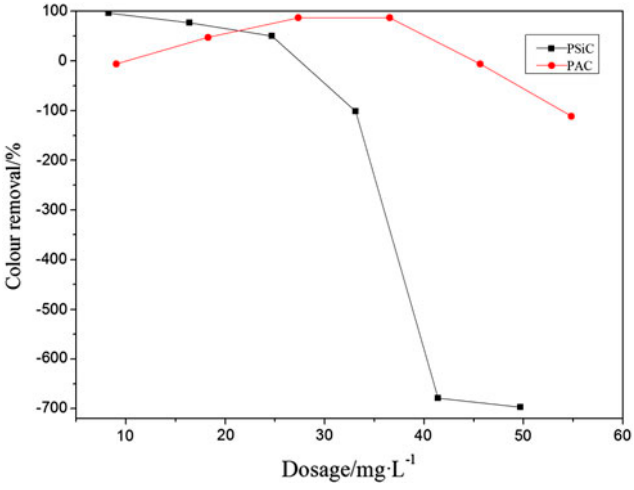

Fig. 6. Color removal of PAC and $PSiC_{0.9}$ with different dosage in the treatment of biologically treated effluent from a recycled paper mill.

respectively, and were much better than that in the tests with PAC. According to the popular coagulation theory, such results may imply that PSiC had stronger neutralization, adsorption, and capture capacities than the traditional coagulant. The paper mill wastewater treated with PSiC could meet the most stringent discharge standard in China. At the same time, the dosage of $PSiC_{0.9}$ was much lower than that of PAC. It means that, when $PSiC_{0.9}$ was used, higher pollutant removal efficiency could be achieved with a lower cost than that with the traditional coagulant.

The dosages of PSiCs with different Si/(Al + Fe) molar ratios and PAC were 16 mg/L (for Al + Fe) and 36 mg/L, respectively. The wastewater sampling volume in jar test was 250 ml.

The tests with the same pH and similar basicity showed that the coagulation performances of all the three PSiCs were quite different (see Fig. 7), which suggested that the coagulation efficiency determined by the charge neutralization, adsorption bridging, and sweeping ability was affected by the molar ratio of Si/(Al + Fe). The removal efficiencies of turbidity and COD increased with the increase of Si/(Al + Fe) molar ratio, but decreased when the silicon dose was too high. $PSiC_{0.9}$ has better coagulation performance than $PSiC_{0.6}$ and $PSiC_{1.5}$. The results were in consonance with the above-mentioned IR and morphological analyses. Moreover, the fact was the content of –OH and branch-like units in $PSiC_{0.6}$ was lower than in $PSiC_{0.9}$, which suggested that polymerization of Fe, Al, and Si was enhanced with the increase of silicon dose. Therefore, the charge neutralization and adsorption bridging abilities of $PSiC_{0.9}$ were better, and thus its pollutant removal efficiency was reasonably higher than that of $PSiC_{0.6}$. In addition, more low-molar polymers but not high polymers were formed when the Si/(Al + Fe) ratio was 1.5, which was considered to be the reason for the decrease of destabilization, adsorption bridging abilities, and pollutant removal efficiency. So, all the

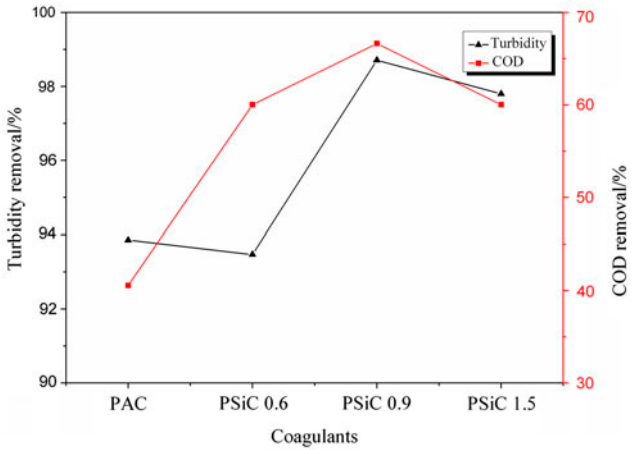

Fig. 7. Coagulation performance of PAC and PSiCs with different Si/(Al + Fe) molar ratios in the treatment of biologically treated effluent from a recycled paper mill.

coagulation performances were consistent with the above revealed characterization of PSiCs.

When $PSiC_{0.9}$, PAC, and aluminum sulfate (AS) were used in another coagulation test, residual color and aluminum in the treated effluent were determined. The results clearly showed that $PSiC_{0.9}$ had higher color removal efficiency than PAC and AS (Fig. 8). These results implied that, to some extent, $PSiC_{0.9}$ had higher lignin removal efficiency than PAC and AS as the quinonoid chromophoric group in lignin, especially ortho-quinone lignin-like structure was the main source of color [26]. It was also reasonable to say that $PSiC_{0.9}$ had better effect on COD removal, for lignin had been considered to be non-degradable organic and it was the major part of residual COD in

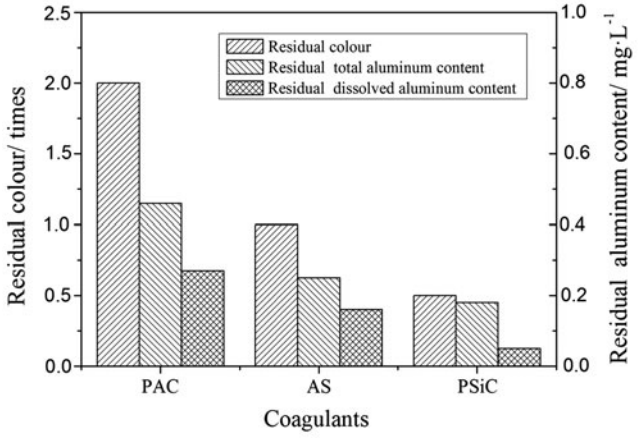

Fig. 8. Residual color and aluminum content in the effluent from secondary settler of a recycled paper mill treated with $PSiC_{0.9}$, PAC, and AS.

biologically treated pulp and paper wastewaters [5]. The underlying cause of the better coagulation efficiency could be assumed that PSiC with metal ions was not only able to improve neutralization capacity, but also able to capture the finer colloidal particles by its ability of adsorption and network capture derived from its net-like structure.

The total residual aluminum and dissolved aluminum content in the coagulation treated effluent have been investigated too, as aluminum has biological toxicity to the micro-organisms in environment and in wastewater treatment system [27]. After treated by $PSiC_{0.9}$, the total residual and dissolved aluminum concentration in the effluent were measured to be 0.18 and 0.05 mg/L, respectively, which were far lower than those in the effluents treated with PAC (0.46 and 0.27 mg/L) and AS (0.25 and 0.16 mg/L) (Fig. 8). Such a result strongly suggests that $PSiC_{0.9}$ is safer than the traditional ones, which might be a large advantage of the new coagulant since varieties of Al salts have been widely used for tertiary wastewater treatment and caused serious Al pollution [28].

4. Conclusions

A new inorganic polymer coagulant PSiCs with different Si/(Al + Fe) molar ratios were prepared by industrial wastes with the method of synchronous polymerization. It has been observed that ions can polymerize with some new complex compounds in the PSiCs. The results of characterization analysis of the new coagulant showed that the polymerization structure, complexation degree, and compound crystals of the coagulants were pretty affected by silicon dose. The peak intensities of –OH, Fe–O–Fe, Al–O–Al, Si–O–Fe, Si–O–Al, Fe–OH, and Al–OH as well as the contents of high polymers decreased when silicon dose was too low or too high. PSiC with Si/(Al + Fe) ratio of 0.9 mainly contained Si–O–Fe–O–Fe–O–Si and Si–O–Al–O–Al–O–Si, and formed net-like structure, which was considered to be the reason for better performance of coagulation. In PSiCs with the other molar ratios like 0.6 or 1.5, only a small amount of Fe^{3+} and Al^{3+} participated in the complexation with PS. Morphological analysis also showed that the amount of irregular PSiC units increased with the increase in silicon dose.

The results of coagulation test with biologically treated effluent from recycled paper mill showed that the new coagulant had an excellent efficiency for the removal of turbidity, COD, and color, which were better in comparison with the widely used traditional coagulants like PAC and AS. Such coagulation performances were consistent with the analysis of PSiC

characteristics. The PSiC coagulant with Si/(Al + Fe) ratio of 0.9 was the optimal molar ratio for preparation of PSiC, since it produce more Si–O–Fe–O–Fe–O–Si, Si–O–Al–O–Al–O–Si and good polymerization structure, and thus a high complexation degree was obtained. Moreover, the PSiC allowed a lower dose in the tertiary treatment and produced an effluent with much lower residual aluminum content.

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