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Characteristics and aluminum reuse of textile sludge incineration residues after acidification

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Abstract

The chemical composition and aluminum speciation of sludge incineration residue (SIR) were determined. Cementation of aluminum from sulfuric acid solution using SIR was studied. The results showed that acid-soluble inorganic aluminum was the predominant component in the sludge, and the total leached aluminum increased from 62.2% to 92.9% after incineration. Sulfuric acid dosage and reaction time were found to affect aluminum recovery positively. Conversely, the increase in temperature significantly inhibited recovery reactions. The optimized leaching condition was 1.66 g sulfuric acid per gram of SIR with a reaction time of 3 hr at 20°C, resulting in the highest aluminum leaching rate of 96.7%. Compared to commercial aluminum sulfate solution coagulants, the leaching solution demonstrated higher COD_{Cr}, turbidity and color removal efficiency for textile wastewater.

Key words: textile sludge; sludge incineration residue; aluminum reuse; acidification

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Introduction

The textile industry consumes large quantities of water and produces polluted wastewater. The regular bio-processing units, chemical reagents are used both in influent and effluent processing to maintain the steady and effective operations of the textile wastewater treatment system. This lead to large amounts of physicochemical and biological sludge (Lubello et al., 2009; Lin and Peng, 1996; Pala and Tokat, 2002; Zodi et al., 2009). For approximately every million tons of textile wastewater 25 m^3 of sludge is generated, and two-thirds of which is physicochemical in nature. In addition large quantities of organic waste, mixed sludge contain a large amount of iron and aluminum ions from added coagulants and flocculants, which inevitably leads to serious secondary pollution if it is not properly treated. Some studies have focused on sludge application to land, but it has been found to result in adverse environmental impacts. Both fresh and stabilized textile sludges had toxicity effects on bacteria, algae, daphnids, fish, earthworms, and higher plants (Rosa et al., 2007a, 2007b).

The current trend of research and development in textile sludge treatment is to make full use of the active ingredients in sludge to achieve sludge minimization and resource recovery. So far, sludge incineration has been applied in the brick industry and in the production of biogas for power generation. Murakami et al., (2009) studied the combustion characteristics of sewage sludge for energy recovery. Physicochemical and engineering properties of a composite textile sludge sample and their suitability for structural and non-structural applications and partial replacement of cement has also been evaluated (Balasubramanian et al., 2006). It has been found that it is possible to use textile sludge as substitute for cement, with a maximum up to 30% from manufacturing non-structural building materials sources. However, the inorganic salts and toxic metals in the sludge could pose a potential threat to the residents.

Some researchers have indicated that dried sludge could be used as adsorbent in wastewater treatment, but the dried sludge contain organic ingredients which may pollute the treated wastewater (Ozmihci and Kargi, 2006; Dhaouadi and M'Henni, 2008; Li et al., 2005). With the development of sludge incineration, more attention has been paid to the metals in incineration residues, particularly the partitioning and fate of heavy metals (Toledo et al., 2005; Corella and Toledo, 2000; Taib et al., 1999; Balogh and Nollet, 2008; Tang et al., 2008; Karamalidis and Voudrias, 2007). Although many potential applications of sludge incineration residues have been proposed, it has only been

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reused for agricultural land and cement production (Wong et al., 2000; Kikuchi, 2001; Zhang et al., 2001; Autret et al., 2007). There are very few reports on metal recovery from incineration residues of textile sludges and the use of its leaching solution as wastewater treatment reagents for enhanced textile wastewater treatment.

In the present study, we investigated the chemical composition and aluminum speciation in SIR and optimized aluminum leaching conditions from the residue by sulfuric acid. The leaching solution was tested for its coagulation effects on textile wastewater treatment.

1 Materials and methods

1.1 Material and methods

Samples were collected from a textile wastewater treatment plant, which generates about one ton of sludge daily. The combined biological and physiochemical treatment processes are used with aluminum sulfate as the major coagulant. The collected sludge samples were air dried, grounded and sifted through a 100 mesh sieve. An aliquot of the smashed and sifted sample was incinerated at 600°C.

1.2 Aluminum speciation in textile sludge

Dried sludge and its incineration residues were divided into four 2 g samples and placed in 250 mL flasks. A 100-mL of 1.0 mol/L KCl, 1.0 mol/L NH_4Ac, 1.0 mol/L H_2SO_4 and 1 mol/L NaOH solutions, were respectively added to each flask. The flasks were shaken for 60 min at 20°C and left to stand for 30 min. It was then subjected to vacuum filtration with 0.45 μm membrane. Exchangeable Al^{3+} absorbed on sludge particles were extracted with 1.0 mol/L KCl while exchangeable Al^{3+}, soluble $Al(OH)_3{}^0$, aluminum fulvate, $Al(OH)^{2+}$, $Al(OH)_2{}^+$, and aluminum polymer adsorbed in colloids were extracted by 1.0 mol/L NH_4Ac. In addition acid-soluble inorganic aluminum, such as $Al(OH)_3$, Al^{3+}, $Al(OH)^{2+}$, $Al(OH)_2{}^+$ and Al_2O_3, except for aluminum humate were extracted with 1.0 mol/L H_2SO_4. On the other hand, all inorganic aluminum, as well as aluminum humate were extracted by NaOH (Van Benschoten and Edwards, 1990). The Al speciation was carried out according to the Van Benschoten and Edzwald method (Schintu et al., 2000).

1.3 Analyses

Aluminum speciation concentration was measured by spectrophotometer according to the national standard GB/T5750.6-2006 (Chrome azurol S colorimetric analysis) (MOHC, 2006). Chemical oxgen demand (COD), volatile suspended solid (VSS), mixed liquor suspended solid (MLSS), colority and turbidity were measured following APHA standard methods (APHA, 1998).

1.4 Sludge acid leaching

The effects of sulfuric acid dosage, reaction time, and temperature on aluminum leaching efficiency were studied. Sulphuric acid dosages of 1.08, 1.23, 1.37, 1.51, 1.66 and 1.80 g were respectively added to per gram sludge incineration residues, with the temperature fixed at 20°C. Temperatures of 20, 40, 60, and 80°C were used with the sulfuric acid dosage 1.66 g. The reaction time was varied between 0.5 to 5 hr.

1.5 Wastewater treatment test

Textile wastewater was taken from a cotton mill in Shanghai, China and was treated by a lab-scale modified anaerobic/anoxic/aerobic (MAAO) process. The MAAO system consisted of an anoxic reactor, an anaerobic reactor, an aerobic reactor and first settling reactor and second settling reactor. The sludge was returned from the bottom of the first and second settling reactor to the anaerobic reactor and anoxic reactor. The recirculation ratios for both were 1. The whole system was placed in a temperature controlled room at 25°C. A variable amount of acid leaching solutions from previous step was mixed with 500 mL water sample in beakers and was stirred at 120 r/min for 1 min followed by 30 r/min for 30 min, and was left to stand for 30 min. Turbidity, COD_{Cr} and colority of the samples before and after the treatments were measured.

2 Result and discussion

2.1 Sludge chemical composition and elemental analysis

Metallic element composition was analyzed by ICP-AES as shown in Table 1. The sludge mainly contains Al, Fe, Mn and Ca, in which Al accounts for more than three-quarters of the total metal content, up to 157 mg/g. Toxic and harmful elements such as As, Cr, Ca and Hg were not found. After incineration, all heavy metal contents were enriched except for Pb. The reason for the enrichment is probably because of the significant reduction in the mass of the sludge due to the incineration of its organic contents. The exception observes for Pb may be due to its volatility at high temperature and requires a more in-depth study. Al was enriched by 1.6 times reaching a final concentration of

Table 1 Contents of metal elements in the sludge

Elements	Al (mg/g)	Ca (mg/g)	Fe (mg/g)	Mg (mg/g)	Zn (mg/g)	Cu (mg/g)	Ni (mg/g)
Before incineration	157	12.0	20.1	1.43	0.77	0.57	0.09
After incineration	248	44.1	41.9	4.49	1.99	0.66	0.14
Elements	Pb (mg/g)	Mn (mg/g)	As (mg/g)	Cr (mg/g)	Cd (mg/g)	Sn (mg/g)	Hg (mg/g)
Before incineration	0.08	0.12	ND	ND	ND	ND	ND
After incineration	ND	0.37	ND	ND	ND	ND	ND

ND: not detected.

Table 2 Amount of aluminum species in sludge

Leaching solution	$Al(OH)^{2+}$, $Al(OH)^{+}$	$Al(OH)_3$, Al_2O_3	Al-HA
From original sludge			
Amount of Al (mg/g)	6.8	94.8	50.8
Percentage of total Al (%)	4.5	62.2	33.3
From incineration residue			
Amount of Al (mg/g)	12.9	227.6	4.4
Percentage of total Al (%)	5.3	92.9	1.8

248 mg/g after incineration.

2.2 Aluminum speciation in the sludge and its incineration residue

The amount of Al in various forms, leached from the sludge and its incineration residue, are listed in Table 2.

Before incineration, $Al(OH)_3$ in the original sludge was 94.8 mg/g, which was accounted for over 50% of total Al in the sludge. It was followed by aluminum humate, accounting for 33.3% of the total Al leached, as 50.8 mg/g. The amount of $Al(OH)^{2+}$ and $Al(OH)_2{}^{+}$ was as low as 6.8 mg/g. The aluminum humate content which was 4.4 mg/g after incineration was significantly reduced. Acid-soluble inorganic Al, mainly in the form of Al_2O_3, increased by 92.9% of the total leached Al and reached 227.6 mg/g. $Al(OH)^{2+}$ and $Al(OH)_2{}^{+}$ concentration was still low, only 12.9 mg/g. The differences are presumably due to the incineration process, in which the sludge was reduced about 88% of total mass at 600°C so that Al was significantly enriched in the residue, and aluminum humate was decomposed under such high temperature.

2.3 Acid leaching operation condition optimization

2.3.1 Effects of acid dosage on aluminum leaching

A series of convincing experimental results tend to prove that sulfuric acid is an effective and less costly acid for metal leaching (Dash et al., 2008; Poulin et al., 2008). Therefore, sulfuric acid was used for leaching Al. The effects of sulfuric acid dosage on Al leaching from textile SIR is illustrated in Fig. 1.

As shown in Fig. 1, the leaching rate of Al increased with increasing acid dosage. The theoretical acid dosage is 1.37 g/g SIR, according to the amount of Al in Table 1. However, in experiment this amount of acid leached only around 75% of the total Al content in the sample, indicating that there are other proton consumers in the incineration residue and that a higher sulfuric acid dosage is required. When the acid dosage was increased to 1.66 g/g, the leaching rate was 96.7%. The Al leaching rate was only about 1% higher than when the acid dosage was increased to 1.80 g/g. The high acidity of leaching solution would give a drop to pH value of water when it was applied to wastewater treatment process and require more basic reagents in the following process, resulting in a waste, thus the optimal dosage of acid was 1.66 g.

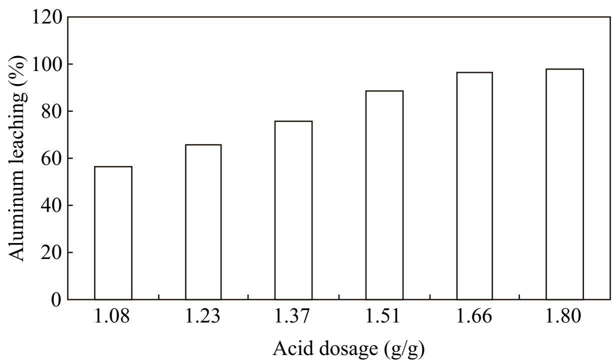

Fig. 1 Effects of sulfuric acid dosage on Al leaching.

2.3.2 Effects of reaction temperature and time on aluminum leaching

The reaction temperature and time are two important factors during the acid leaching process.

As shown in Fig. 2, the Al leaching rate decreased from 96.7% to 81.5% as the reaction temperature rase from 20 to 80°C. It was also observed that the solid-liquid mixture became more viscous as the reaction temperature increasing, leading to difficulties in solid-liquid separation, thus some of the leached aluminum ions were trapped in the filter paper and the measured values of Al decreased. Raising the reaction temperature could also increase the cost for reaction. As the result the reaction was optimized to carry out at 20°C.

In Fig. 2, at 20°C, the Al leaching rate at the beginning increased sharply as reaction time increased. However, after 3 hr the reaction was approaching balance with only a slight increase in Al leaching rate. Taking into account the recovery cost, the reaction time was optimized as 3 hr.

2.4 Comparison of SIR leaching solution with commercial aluminum sulfate solution

Under the optimized conditions, the addition of H_2SO_4 at 0.92 mL/g of sludge incineration residue, and maintaining the reaction at 20°C for 3 hr, the highest Al leaching rate 96.7% was achieved. A comparison of leaching solution and the commercial aluminum sulfate solution is listed in Table 3.

The leaching solution had an Al content of 7.6%, iron content of 0.53% and a pH of 3.65, however, there was no organic content due to the incineration at 600°C. While the Al content was slightly lower than the industrial quality standards on aqueous aluminum sulfate, the iron content was slightly higher than the standard value. But the iron salt and calcium salt are also good coagulants and will form floccules during the coagulation process to precipitate together with sludge, so they do not affect the effluent quality. Therefore, this leaching solution could be used as a coagulant in wastewater treatment.

Table 3 Comparison between leaching solution and commercial aluminum sulfate

Items	Al_2O_3	Fe	Organic matter	pH
Leaching solution	7.6%	0.53%	0	3.65
Commercial solution aluminum sulfate	7.8%	0.25%	–	3.00

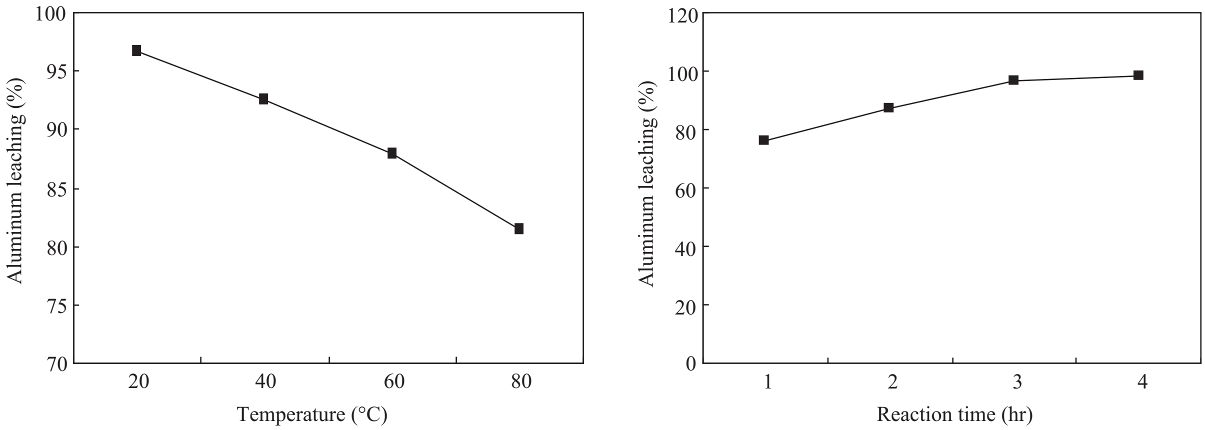

Fig. 2 Effects of reaction temperature (a) and reaction time (b) on Al leaching.

2.5 Application of leaching solution as coagulant

2.5.1 Effects of coagulant dosage on textile wastewater

Wastewater samples were obtained from a textile mill in Shanghai, containing active, sulfide, acidic and other dyes. The wastewater had COD_{Cr} value of 698.4 mg/L, turbidity value of 50.4 NTU and pH of 9.22. The acid leaching solution from previous steps was reused as a Al coagulant and the results are shown in Fig. 3.

As shown in Fig. 3, the removal rate of pollutants by leaching solution is similar to that of commercial aluminum sulfate. When the leaching solution as coagulant increased from 0.5 to 2 mL/L, COD_{Cr} significantly decreased from 698.4 to 356.9 mg/L, with the highest COD_{Cr} removal percentage of 48.9%. The leaching solution is also efficient for the wastewater turbidity removal in the tested dosage range. The turbidity removal rate reached 92.6% when the dosage was 2 mL/L. Al and Fe may play a major role in removing these pollutants by concrete effects. Some of dyes, notably disperse dyes could react with these leaching solution to formed deposits. The results also showed that leaching solution could replace the usual chemicals, such as alum sulphate, used as coagulation agents in the field of wastewater treatment.

2.5.2 Effects of coagulant dosage on the MAAO process effluent

The wastewater sample from the textile mill in Shanghai was treated in a laboratory scale MAAO system and the effluent had a COD_{Cr} of 55.44 mg/L, pH of 8.26 and turbidity of 1.7 NTU. The prepared coagulant was evaluated with this effluent and the results are shown in Fig. 4.

As illustrated in Fig. 4, the COD_{Cr} removal rate increased and then decreased with the coagulant dosage increased from 0.2 to 1.2 mL/L, peaked at 0.4 mL/L with the maximum COD_{Cr} removal rate of 48.48%, where COD_{Cr} value decreased from the original 55.04 to 28.56 mg/L while pH decreased from 8.26 to 6.89. Within the same dosage range, the turbidity decreased gradually when the coagulant dosage increased, then followed by a slight increase. When then dosage increased to 0.4 mL/L, the effluent turbidity decreased to 0.6 NTU from original 1.7 NTU, with a removal rate as high as 64.7%. After the coagulant treatment, modified AAO process effluent met all the standards on COD_{Cr}, turbidity and coloration in "Urban Wastewater Reuse-Industrial Water Quality" (GB/T19923-2005) of China.

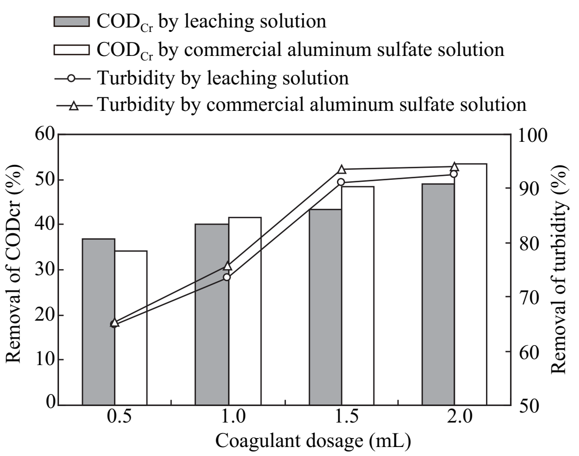

Fig. 3 Comparison between leaching solution and commercial aluminum sulfate on pollutants removal rate.

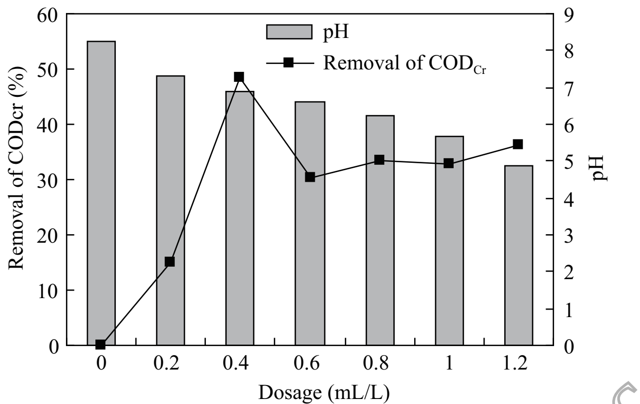

Fig. 4 Effects of coagulant dosage on the modified AAO process effluent.

3 Conclusions

This study demonstrates that Al leached in the tested SIR significantly increased to 248 mg/g, as 1.6 times of its original in content in the sludge with leaching condition of 1.66 g sulfuric acid per gram SIR and a reaction time of 3 hr at 20°C. The acid-soluble inorganic Al is the predominant component in the sludge, it increased from 62.2% to 92.9% of the total leached Al after incineration. Meanwhile, the Al humate content reduced from 33.3% to 1.8%. Under the optimized conditions, by adding sulfuric acid at 1.66 g/g SIR, maintaining the reaction at 20°C for 3 hr, the highest aluminum leaching rate was achieved at 96.7%. The Al content is similar with the industrial quality standards on aqueous aluminum sulfate in China. The leaching solution of SIR demonstrated significant effects on treating the textile wastewater as well as the modified AAO process effluent, indicating the potential to be used as a textile wastewater treatment agent for pre- and post-processing.

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