

Preparation and characterization of a cost-effective red mud/polyaluminum chloride composite coagulant for enhanced phosphate removal from aqueous solutions



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ABSTRACT

Composite coagulant has raised great concerns due to its cost-effectiveness and outstanding coagulation performance. A novel composite red mud (RM)/polyaluminum chloride (PACl) coagulant (RMPACl) was prepared for effective removal of phosphate from aqueous solution. The RM obtained from alumina industrial waste was employed as an alternative alkali to adjust the basicity of the semi-product of polyaluminum chloride (ACl). The RMPACl was characterized by pH titration, Al(III) species distribution, TEM, XRD, FTIR, phosphate removal performance as well as flocs properties analysis. The results showed that RM can significantly enhance the basicity of ACl. The TEM and FTIR results indicated that RMPACl was composed of polymeric aluminum species and residual RM particles. In addition, the phosphate removal experiments demonstrated that the optimum dosage of RMPACl was 147.5 mg/l for the decontamination of phosphate with an initial concentration of 5 mg/l, which is much smaller than that of PACl. RMPACl with relatively smaller and porous flocs exhibited a faster settling time than PACl, because the additional RM played an important role in the phosphate removal. More importantly, the RMPACl performed better than PACl in the treatment of the synthetic and natural phosphate contaminated water. It is anticipated that RMPACl can be proposed as a cost-effective coagulant for the effective removal of phosphate from natural water.

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1. Introduction

Coagulation has been widely applied in water treatment due to its easy operation and cost-effectiveness. Among different coagulants, polyaluminum chloride (PACl) is considered to be a promising agent for the coagulation process, which exhibits excellent coagulation performance in previous reported studies [1,2]. Generally, two steps are involved in the traditional preparation of PACl: firstly, the semi-product of polyaluminum chloride (ACl) is prepared by digesting aluminum hydroxide with hydrochloric acid; various alkalis such as calcium aluminate, caustic soda, lime or aluminum slag are then added into ACl to adjust basicity usually ranging from 15% to 85% [3].

Although the pre-polymerized aluminum salts (PACl) show better coagulation performance in some cases in comparison with

conventional coagulants [4], their applications in water treatment are still limited due to the high cost of raw materials [5]. Therefore, it is urgently required to develop more cost-effective coagulants for water treatment.

Red mud (RM), composed of aluminum oxide, iron oxide, calcium oxide and silica, is a highly alkaline (with pH 10.0–12.5) waste generated in the production of alumina [6]. The manufacture of 1 ton of alumina generally results in the production of 1.0–1.5 ton of RM. As digested in the caustic solution of soda, RM carries high alkali; up to 2 tons of liquor at 5–20 g/L caustic (as Na₂CO₃) accompanied every ton of dry mud solids [7]. Each year, about 90 million tons of RM may be produced worldwide and 10–20 million tons of caustic RM must be disposed annually in China [8]. Usually, RM is discharged into storage impoundments where environmental pollutions are likely to occur due to the leakage of alkaline leachate into the ground water. Therefore, final disposal of RM is facing a huge challenge due to the high alkalinity and large amounts involved.

In previous studies, RM has been used for preparation of adsorbents [9–12], coagulants [13,14], and catalysts [15]. However, few

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studies have been reported on the utilization of the alkaline of RM. Since RM is an industrial waste with high alkali, it is considered to be cost-effective to employ RM in the preparation of coagulants. Additionally, it is reported that RM shows effective removal of phosphate from water [9]. In this study, RM composite coagulant (RMPACI) was prepared from RM and the semi-product of polyaluminum chloride (ACI). High alkaline industrial waste RM was utilized for basicity adjustment of the ACI. The characterizations of RMPACI were conducted using pH titration, Al(III) distribution, TEM, XRD, FTIR, and floc properties analysis. A jar test was conducted to evaluate the coagulation performance for phosphate removal.

2. Materials and methods

2.1. Materials

Industrial grade ACI (Al_2O_3 wt% = 13.6, basicity = 2.0%) and PACI (Al_2O_3 % = 10.0, basicity = 76.3%) were provided by Zhongke Tianze Flocculants Corporation, China. Red Mud (RM), which was a by-product of the Bayer process, was supplied by Shandong Aluminum Corporation, China. The chemical composition of RM was determined by acid digestion and ICP-OES analysis. The resulted chemical composition of RM shown in Table 1 demonstrated the main components of RM were Fe_2O_3 , Al_2O_3 , SiO_2 , CaO , and Na_2O . Other trace constituents were K_2O , MgO , ZnO , and TiO_2 (data not shown).

2.2. Preparation of RMPACI

A series of RM composite coagulants were prepared in order to find out the effect of RM on basicity adjustment. Basicity value (B) was determined according to the AWWA Standard for PACI, described as follows [16]:

$$B = \frac{[\text{OH}]}{3[\text{Al}]} \times 100(\%) \quad (1)$$

The predetermined amount of RM was added into the ACI solution under vigorous stirring. The chosen RM/ACI ratios in terms of weight-RM/volume-ACI (w/v) were 0.1, 0.25, 0.3, 0.4 and 0.5 until irreversible precipitation occurred resulting in viscous and hard handling products. The preparation was carried out in a flask within the water bath at 80°C for 2 h. Thereafter, RMPACI samples with different basicity values were obtained and were found to be stable for several weeks.

2.3. Characterization

pH titration and zeta potential of RMPACI were carried out by using Auto-potentiometric titrator (Metrohm 716). Residual RM remained in RMPACI was separated by centrifugation, washed and dried in an oven. The specific surface area of residual RM was determined by an accelerated surface area and porosimetry (Micromeritics ASAP 2000).

The Al(III) species distribution was measured by a timed complexation spectroscopy method using a spectrophotometer (HACH DR-5000). The main Al(III) species can be categorized to Al(III) monomeric species (denoted as Al_a), Al(III) polymeric species (denoted as Al_b) and Al(III) colloidal and precipitated species (denoted as Al_c). This procedure had been applied to identify the

nature of the coagulant species, and qualify the amount of metal complex formed. Details about the procedure can be referred to Zhou et al. [17].

The morphology and structure of RMPACI were studied by using transmission electron microscopy (TEM) (PHILIPS TECNOL 20), Fourier transform infrared spectroscopy (FTIR) (Bruker Optics Tensor 27) and X-ray diffraction (XRD) (Shimadzu XRD-6000). The flocs were collected from the coagulation process during the formation stage, and dried at 50°C with a freeze drier (Boyikang FD-1A-50). The morphology of the flocs was examined by a Hitachi S-3000N scanning electron microscopy (SEM).

2.4. Coagulation experiments

Coagulation experiments were performed using synthetic water containing phosphate (5 mg/l) and natural eutrophic water. Standard jar tests were conducted using a six-jar tester (JTY-6 Laboratory Stirrer). Coagulation performance of RMPACI for phosphate removal, including the effects of coagulant dosage, alkalinity, and settling time, were investigated by using synthetic phosphate contaminated water and compared with that of conventional coagulant PACI. In addition, the practical coagulation performance of both coagulants was also evaluated using eutrophic Xiangxi river water from the three gorges reservoir area. 1L phosphate contaminated solutions were prepared for batch experiments under different conditions. All the experiments were conducted in duplicate. Details about the experimental procedures can be referred to our previous study, i.e., Ni et al [18].

Phosphate concentration of the sample was measured using the ascorbic acid method with a spectrophotometer (HACH DR-5000), according to American Public Health Association (APHA) standard methods [19]. The phosphate removal efficiency η (%) was calculated as below:

$$\eta = \frac{(C_i - C_e)}{C_i} \times 100(\%) \quad (2)$$

where C_i is the initial phosphate concentration (mg/l), C_e is the final phosphate concentration of the supernatant samples (mg/l).

2.5. Determination of floc properties

Dynamic floc size was monitored using a Malvern Mastersizer 2000 (Malvern Co., United Kingdom) as the coagulation proceeded [20]. The schematic diagram of the online monitoring system for dynamic floc size was shown in Fig. 1. Flocs in the suspension were monitored by drawing water flow through the sample cell of the instrument and back into the jar by a peristaltic pump, which was located downstream of the instrument to prevent disturbing the flocs prior to measurement. Size measurements were taken every

Table 1
Chemical composition of RM (%).

Components	Al_2O_3	Fe_2O_3	SiO_2	Na_2O	$\text{CaO} + \text{MgO}$
wt%	37.6	29.2	21.3	4.2	6.3

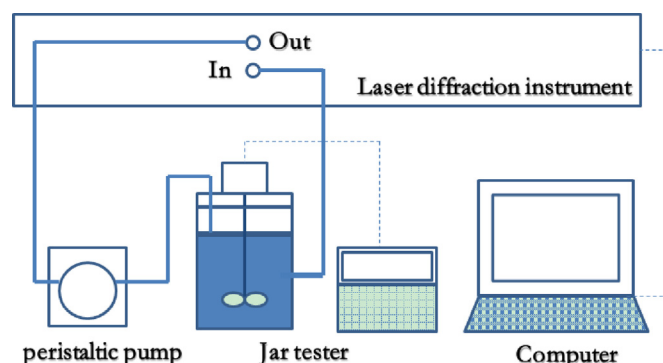


Fig. 1. Flowchart of laser light scattering online floc size monitoring.

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