



Evaluation of a novel composite inorganic coagulant prepared by red mud for phosphate removal

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ABSTRACT

Red mud is a waste produced in very large quantities by the aluminum industry. This study relates to a process of transformation of red mud, by treatment in hydrochloric pickle liquor of bauxite, into a novel composite inorganic coagulant that is usable for phosphate removal. The optimum conditions for preparing the composite coagulant with red mud and hydrochloric pickle liquor of bauxite were studied. The effect of various factors such as pH, ionic strength and water temperature on performance of the prepared coagulant was investigated. The coagulation performance of the composite coagulant by treating biologically pre-treated municipal sewage and eutrophic water was evaluated. The leaching potential of heavy metals from the coagulant was also investigated. The results showed that the prepared coagulant could reduce the phosphate in values lower than 0.02 mg/L. Compared with commercial polyaluminum chloride coagulant (PACl), it could promote phosphate removal efficiency from 4.9% to 10.4%. Overall, it is suggested that the composite coagulant is an efficient coagulant and it can be possibly considered as a promising option for tertiary treatment of wastewaters and removing nutrients in eutrophic water, especially for the removal of phosphate. It exhibits better coagulation performance than commercial polyaluminum chloride coagulant.

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1. Introduction

As known, phosphate in wastewaters is regarded as a major nutrient for growth of both vegetable plants and microorganisms. But excess amount of phosphate concentration in the effluent discharge accelerates eutrophication that affects many natural water bodies [1–4]. Eutrophication of the water bodies is one of the most important environmental problems. Eutrophication can lead to abundant development of aquatic plants, growth of algae and disturb the balance of organisms present in the water [5]. This affects water quality through the depletion of oxygen level because of high biological oxygen demands, and acidification. It in turn can have harmful effects on fish and other aquatic life, microorganism's growth as well as it causes reductions in biodiversity. Consequently, the removal of phosphate in domestic and industrial discharges is absolutely necessary to avoid any kind of problems.

Phosphorus usually occurs in wastewater and surface water in the form of organic phosphates and inorganic phosphates. The

treatment of excessive phosphate can be achieved by several physical, chemical and biological methods [6–10]. Physical methods for water treatment are either too expensive, or not useful with regard to phosphate elimination from water [11,12]. The biological method is low cost but the removal efficiency usually does not exceed 30%, which means that remaining phosphate should be removed by other techniques [13]. Chemical removal techniques are widely accepted methods of phosphate removal, especially coagulation process. Coagulation process is highly efficient in removing phosphate. It is regarded as an important option for the tertiary treatment of wastewater, intended for prevention of eutrophication. However, a typical drawback of coagulation process is the high cost associated with the use of metal salts [14]. Therefore, low cost and easily available materials such as fly ash, alum sludge, red mud and other waste materials were used as raw materials for preparation of coagulant to remove phosphate from wastewater [15].

Red mud is a residue of the aluminum industry rejected during the alkaline extraction of alumina from bauxite by the Bayer or sintering process. The quantity of red mud is almost equal to the primary aluminum production: alkaline digestion of 2.5 t of bauxite affords 1 t alumina and about 1.5 t of red mud [16]. Therefore, an average Al_2O_3 productivity of $10 \times 10^6 \text{ t year}^{-1}$ involves a mass of by-products of $15 \times 10^6 \text{ t year}^{-1}$ discharged as slurry retaining variable water contents in China [17,18]. Though being washed and considered as an inert solid waste, red mud remain strongly alkaline and are highly

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corrosive. The high alkaline content of red mud and its enormous quantities caused significant ecological problems and considerable negative environmental effects.

Many efforts are being made globally to find suitable uses for red mud. Some researches focused on the application of red mud in wastewater treatment. Red mud can be used as an adsorbent to remove pollutants and toxic materials from water, such as Cu^{2+} [19], Cr^{3+} [20], $\text{C}_6\text{H}_5\text{OH}$ [21], Zn^{2+} [22], F^- [23], $\text{C}_{16}\text{H}_{18}\text{N}_3\text{CISIUPAC}$ [24] and PO_4^{3-} [25].

Red mud is rich in calcium, aluminum, iron and silicon, which are essential raw materials for the production of water and wastewater treatment coagulants. Some team of researchers studied the capacity of untreated red mud to coagulant phosphate [26]. However, the removal efficiency of phosphate obtained during these experiments was limited, only about 20%. These studies thus confirm the need for a novel method to transform red mud into an effective coagulant for phosphate removal.

Hydrochloric pickle liquor of bauxite mainly consists of AlCl_3 and HCl . It is usually used as a semi-manufactured material for production of polyaluminum chloride (PACl). In general, some alkalis such as calcium aluminate, caustic soda, lime and aluminum slag react with hydrochloric pickle liquor of bauxite to prepare PACl. If we replace these alkalis with red mud, it may not only reduce the cost for preparing coagulant, but also provide a feasible way to recycle the alkalis in the red mud. Meanwhile, the dissolved metals of red mud in hydrochloric pickle liquor of bauxite could enhance the coagulation efficiency of the coagulant.

The objective of this work was to study the feasibility of a new coagulant prepared by red mud and hydrochloric pickle liquor of bauxite for phosphate removal from wastewater. Firstly, the phosphate removal efficiency of the new coagulant was evaluated to optimize the preparation conditions such as reaction time, reaction temperature and liquid/solid ratio. Secondly, the influence of temperature, pH and salt concentration on the removal efficiency of phosphate was investigated in order to determine optimal operating conditions. Coagulation experiments were also conducted for the evaluation of the new coagulant capability to treat biologically pre-treated municipal wastewater and eutrophic water, comparing with the coagulation efficiency of conventional coagulant PACl. Finally, the leaching potential of heavy metals from the coagulant was evaluated. The initial results of laboratory scale experiments will be useful for the development of a novel, stabilized and highly effective coagulant.

2. Materials and methods

2.1. Materials

Red mud used in this study was provided from Shandong Aluminum Corporation, Shandong, China. Before experiments were performed, the red mud was first homogenized and dried. Then the dried red mud samples were crushed in a mortar. The chemical analysis of raw red mud is given in Table 1 [6], where it can be seen

that red mud is primarily composed of Ca, Si, Fe and Al oxides. Other minor elements such as Mg, K, Ti and Na are also precipitated as solid phases. Additional details on the characteristics of red mud are given elsewhere [27]. Before preparing the novel coagulant, red mud placed in porcelain were calcined at various temperatures (300, 500, 700, 900 °C) for 2 h in a 48000 electric muffle furnace (Keda Scientific Co., Ltd). The power was stirred at 30 min intervals during the heat treatment.

Hydrochloric pickle liquor of bauxite was obtained from Tianjin Dagang Flocculant Corporation, Tianjin, China. It was industrial grade. The chemical analysis of hydrochloric pickle liquor of bauxite showed that it contains 13.6% Al_2O_3 . The hydrochloric pickle liquor of bauxite is highly acidic and the pH of hydrochloric pickle liquor of bauxite is 1.7.

For comparison reasons, commercially available PACl containing 17% Al_2O_3 obtained from Tianjin Dagang Flocculant Corporation, Tianjin, China was also examined.

The analysis of phosphate was carried out using the ascorbic acid method with a HACH DR/4000U spectrophotometer, according to APHA standard methods [28]. A pH meter (Orion) was used to measure the pH of solutions in experiments. All chemicals used in the experiments were analytic grade and all experiments were conducted in duplicate and the average values were used for analysis.

2.2. Preparation of coagulation reagents

Firstly, an aliquot of 200 ml of pickle liquor was added to a three-necked flask and placed in an electric heat constant temperature water bath at atmospheric pressures. Secondly, varied amounts of red mud were introduced into the three-necked flask according to the weight/volume ratio of red mud/hydrochloric pickle liquor of bauxite (W/V, 5:100 to 45:100). Reaction temperatures were varied from 25 °C to 95 °C and reaction time were varied from 1 h to 5 h for optimization purposes. The mixture was stirred to improve the chemistry reaction time. Finally, the mixture was cooled at room temperature and the coagulants were prepared well.

2.3. Performance of the produced coagulant

Coagulation tests were performed by using synthetic water, actual municipal sewage and eutrophic water. All coagulation tests were conducted using a six-jar tester (FlocculatorJTY-6, Beijing Daiyuan Co., Beijing, China). Phosphate was the key factor we considered in testing the performance of the novel coagulant, so we just considered the phosphate removal efficiency of the coagulant in the treatment of synthetic water to get the optimum preparation conditions. The synthetic water was prepared by diluting a concentration 1 g P L^{-1} in PO_4^{3-} to the experimental concentration by adding tap water of very low hardness. The pH value and phosphate of the synthetic water were 7.0 and 1.0 mg/L. For treating the synthetic water, the coagulant dose was kept constant at 50 mg/L. The actual municipal sewage was taken from secondary effluent of Beixiaohe sewage treatment plant (Beijing, China). The original pH and phosphate of the municipal sewage were 7.5 and 3.20 mg/L. The actual eutrophic water was taken from a lake in Beijing Zoo, Beijing, China. The original pH and phosphate of the eutrophic water were 7.6 and 0.12 mg/L. The jar-test experimental conditions were: initial rapid mixing at 300 rpm for 2 min, followed by slow mixing at 100 rpm for 20 min and final sedimentation for 30 min. At the end of these experiments about 50 ml of sample was withdraw 5 cm below the liquid surface for phosphate measurement.

In order to estimate the leaching potential of heavy metals from the new coagulant, the treated wastewater was separated off using $0.45 \mu\text{m}$ filter membrane and analysis for heavy metals by Inductively Coupled Plasma Mass Spectrometer (Plasma Quad 3).

Table 1

Average composition of red mud used (% by wt.).

Constituent	% (w/w)
SiO_2	18.08
Fe_2O_3	15.30
Al_2O_3	8.26
CaO	38.02
MgO	1.63
Na_2O	3.55
K_2O	0.36
TiO_2	4.51

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