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Research article

Comprehensive reuse of drinking water treatment residuals in coagulation and adsorption processes

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ABSTRACT

While drinking water treatment residuals (DWTRs) inevitably lead to serious problems due to their huge amount of generation and limitation of landfill sites, their unique properties of containing Al or Fe contents make it possible to reuse them as a beneficial material for coagulant recovery and adsorbent. Hence, in the present study, to comprehensively handle and recycle DWTRs, coagulant recovery from DWTRs and reuse of coagulant recovered residuals (CRs) were investigated. In the first step, coagulant recovery from DWTRs was conducted using response surface methodology (RSM) for statistical optimization of independent variables (pH, solid content, and reaction time) on response variable (Al recovery). As a result, a highly acceptable Al recovery of 97.5 ± 0.4% was recorded, which corresponds to 99.5% of the predicted Al recovery. Comparison study of recovered and commercial coagulant from textile wastewater treatment indicated that recovered coagulant has reasonable potential for use in wastewater treatment, in which the performance efficiencies were 68.5 ± 2.1% COD, 97.2 ± 1.9% turbidity, and 64.3 ± 1.0% color removals at 50 mg Al/L. Subsequently, in a similar manner, RSM was also applied to optimize coagulation conditions (Al dosage, initial pH, and reaction time) for the maximization of real cotton textile wastewater treatment in terms of COD, turbidity, and color removal. Overall performance revealed that the initial pH had a remarkable effect on the removal performance compared to the effects of other independent variables. This is mainly due to the transformation of metal species form with increasing or decreasing pH conditions. Finally, a feasibility test of CRs as adsorbent for phosphate adsorption from aqueous solution was conducted. Adsorption equilibrium of phosphate at different temperatures (10–30 °C) and initial levels of pH (3–11) indicated that the main mechanisms of phosphate adsorption onto CRs are endothermic and chemical precipitation; the surfaces are energetically heterogeneous for adsorbing phosphate.

1. Introduction

The chemical coagulation technique using aluminum (Al)-based reagents such as aluminum sulfate (alum), aluminum chloride, and polyaluminum chloride (PAC) is the most common process in conventional drinking water treatment plants to enhance drinking water quality (Ghafari et al., 2014; Nair and Ahammed, 2014). Even though high purity drinking water can be obtained by following a process of coagulation, the enormous quantities of sediment sludge, referred to as drinking water treatment residuals (DWTRs) or waterworks residuals, are generated (Okuda et al., 2014). It has been estimated that drinking water treatment plants produce 1200 tons of DWTRs daily in the Republic of Korea (ME, 2013). In addition, the available literature has reported that several million tons of DWTRs might be produced annually all around world (Evuti and Lawal, 2011). Besides this, due to the scarcity of landfill sites, coupled with an overall reinforcement of environmental restriction, the disposal of sludge has become more problematic and expensive. Rising concerns over environmental pollution and the effective control of DWTRs have spawned considerable research into alternative management strategies.

In this regard, due to the amphoteric nature of DWTRs, their reuse as a coagulant recovery under acidic conditions in the range of pH 1–3 has been considered a potential approach because the

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process could effectively recover coagulant at a rate of over 70%, along with enhancing the dewaterability and reducing the sludge volume (Panswad and Chamnan, 1992; Huang et al., 2010). If successful coagulant recovery from DWTRs can be accomplished, such that this recovered coagulant can be reused as a coagulation reagent for wastewater treatment, this would lead to a reduction of commercial coagulant usage as well as to a volume/solid reduction, thereby undoubtedly leading to environmental and economic benefits (Prakash and Sengupta, 2003). However, there are two major drawbacks that must be considered in any implementation of this strategy. First, in strong acidic conditions, some other metallic components in DWTRs, such as manganese, zinc, and lead could be also extracted; the levels of these impurities may make it impossible to justify the reuse of these materials in drinking water treatment plants because trihalomethane, which is classified as a carcinogenic compound, will be generated during the chlorination process (Evuti and Lawal, 2011). Even though coagulant recovery followed by membrane processes has been applied to enhance the purity of recovered coagulant, in a global viewpoint, direct reuse of recovered coagulant for industrial wastewater treatment seems to be more suitable. Second, there is a weakness in the acid based Al recovery process because coagulant-recovered residuals (CRs) are generally characterized as having low pH (<2.0); thus, if directly disposed of in a landfill, their acidified condition can lead directly to their damaging the environment. In addition, further economic advantages can be expected if it is possible to exploit an alternative route for CRs (Keeley et al., 2012). Hence, in order to comprehensively manage DWTRs, the application of CRs as adsorbent for anionic target pollutants such as phosphates can also be considered.

On the basis of the above mentioned issues, this study mainly comprises three parts: (I) coagulant recovery, (II) application of recovered coagulant, and (III) application of CRs as adsorbent. In the research in part (I), even though coagulant can also be recovered efficiently from DWTRs under alkaline conditions over pH 12, coagulant recovery was conducted under only acidic conditions because it was possible to generate very high sludge volume after alkaline-based extraction (Li et al., 2005). Besides this, most studies of coagulant recovery have been performed using the conventional optimization method; however, the use of this method may have ignored interactive effects of key parameters on the process. Therefore, a statistical and mathematical optimization technique, namely response surface methodology (RSM), was used for coagulant recovery due to the fact that influential parameters, alone or in combination, can have effects on the process efficiency. Subsequently, the research in part (II) was conducted toward the application of recovered coagulant. In this part, real cotton textile wastewater was selected as the target wastewater. Because of the very considerable volume of wastewater generation and its unique properties such as its containing a wide variety of dyes, its non-biodegradability, and its toxic characteristics, textile wastewater can have a detrimental impact on aquatic ecosystems and can cause problems at biological wastewater treatment plants; as a result, such wastewater can be cost effectively removed by help of coagulants (Georgious et al., 2004; Pala and Tokat, 2002; Verma et al., 2012). For the optimization of the coagulation process, the operational parameters (i.e., Al dosage, initial pH, and reaction time) were statistically optimized using RSM. Finally, as described above, effectively dealing with CRs is one of the great challenges for any comprehensive management strategy. We hypothesized that, though the proportion of Al content will decrease due to coagulant recovery in acidic conditions, the attaching of ionized Al (Al^{3+}) on the surface of the CRs, or the use of remaining Al content in DWTRs, may provide an additional way to remove phosphate from aqueous solution. Therefore, a feasibility study on the direct reuse of CRs as adsorbent for phosphate removal from aqueous solution was also carried out. We present the results of the adsorption equilibrium test, which show that CRs exhibited reasonable performance as phosphate adsorbents.

2. Materials and methods

2.1. Drinking water treatment residuals (DWTRs) and cotton textile wastewater

Using PAC as the coagulant, the dewatered DWTR samples used in this study were collected from a drinking water treatment plant in Seoul, Republic of Korea, whose influent water source is the Han River. The collected DWTRs were dried again in an oven at 105 °C for 24 h to remove retained water content in the sample. Subsequently, they were ground into powder using a knife-milling Waring commercial blender; then, the fraction passing through a 425 μm sieve was collected. Concentrations of relevant elements detected in the DWTRs are summarized in Table 1. It can be seen that the DWTRs contain predominantly high levels of Si, Al, and Fe contents of 37.3%, 33.0%, and 15.2%, respectively, and relatively small amounts of K, Ca, S, P, and other elements. The percentage of metal oxide forms indicates that the elements were present in their highest oxidation state (Zheng et al., 2004).

Cotton textile wastewater was collected from the wastewater storage tank of a local cotton textile wastewater treatment plant in Seoul, Republic of Korea, and then directly stored at below 4 °C in a dark condition to avoid any change in physico-chemical characteristics before use. As listed in Table 2, the cotton textile wastewater had a COD concentration of 685 ± 50 mg/L, pH of 8.1 ± 0.2, and turbidity of 92.6 ± 2.8 NTU. The absorbance of the wastewater was monitored at a maximum wavelength (λ_{max}) of 226 nm and the wastewater had an Abs_{226} of 1.42 ± 0.03 (5 times diluted value).

2.2. Experimental procedure

2.2.1. Preliminary test I: determination of key parameter ranges for Al recovery

In the preliminary test, two batch processes were conducted to determine the minimum and maximum ranges of the pH (test I) and the reaction time (test II) for the next statistical optimization test using RSM. In this study, on the basis of the pre-experimental comparison result, which used 1 mol of each H_2SO_4 and hydrochloric acid at pH 1.5 condition, as acid chemical reagents, only sulfuric acid (H_2SO_4) was considered due to its higher Al recovery performance (supplementary data, Fig. S1). In both batch tests, a certain amount of powdered DWTRs were added to 500 mL (in a vitreum cylindrical reactor) of deionized distilled water (solid content was fixed at 1.5% w/w). The mixture was continuously stirred at 150 rpm (with reaction time of 30 min for batch test I, while this value was 5–50 min for batch test II); then, the mixture was filtered immediately to avoid any further reaction and filtrate was analyzed for calculation of Al recovery. To maintain specified conditions, the pH was adjusted using 1 mol of H_2SO_4 and 1 mol of NaOH. All batch tests were performed at room temperature (25 ± 1.5 °C). In order to minimize random errors, each batch experiment was conducted in parallel and the average values (in

Table 1
Chemical property (XRF) of DWTRs.

Metal (% dry weight)	Si	Al	Fe	K	Ca	S	P	Etc.[a]
	37.3	33.0	15.2	3.7	2.4	2.0	1.3	5.1
Oxide (% dry weight)	SiO_2	Al_2O_3	Fe_2O_3	K_2O	CaO	SO_3	P_2O_5	Etc.[a]
	42.0	39.3	8.3	1.9	1.4	2.3	1.3	3.5

[a] The total sum of below 1.0% elements.