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Phosphorus removal by acid mine drainage sludge from secondary effluents of municipal wastewater treatment plants

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ABSTRACT

Acid mine drainage (AMD) sludge, a waste product from coal mine water treatment, was used in this study as an adsorbent to develop a cost-effective treatment approach to phosphorus removal from municipal secondary effluents. Batch tests were carried out to study the effects of pH, temperature, concentration, and contact time for phosphorus removal from wastewater. Batch tests were followed by continuous flow tests using a continuous stirred tank reactor (CSTR). Adsorption of orthophosphate onto AMD sludge particles followed the *Freundlich* isotherm model with an adsorption capacity ranging from 9.89 to 31.97 mg/g when the final effluent concentration increased from 0.21 to 13.61 mg P/L. P adsorption was found to be a rather rapid process and neutral or acidic pH enhanced phosphorus removal. Based on a thermodynamic assessment, P adsorption by AMD sludge was found to be endothermic; consequently, an increase in temperature could also favor phosphorus adsorption. Results from batch tests showed that leaching of metals common to AMD sludges was not likely to be a major issue of concern over the typical pH range (6–8) of secondary wastewater effluents. CSTR tests with three types of water (synthetic wastewater, river water, and municipal secondary effluent) illustrated that P adsorption by AMD sludge was relatively independent of the presence of other ionic species. In treating municipal secondary effluent, a phosphorus removal efficiency in excess of 98% was obtained. Results of this study indicated that it was very promising to utilize AMD sludge for phosphorus removal from secondary effluents and may be relevant to future efforts focused on the control of eutrophication in surface waters.

1. Introduction

The release of phosphorus (P) from municipal wastewater treatment effluents into the environment is one major cause of eutrophication in receiving water bodies (Oleszkiewicz and Barnard, 2006; Edwards and Withers, 2007). In order to mitigate the growth of aquatic plants and phytoplanktonic algae, environmental agencies are required to develop phosphorus total maximum daily load (TMDL) for many watersheds (Havens and Schelske, 2001; Walker, 2003). In addition to TMDL requirements, some regulations can require the phosphorus concentration in wastewater effluents to be as low as 50 μg/L (Genz et al., 2004). Phosphorus exists in municipal wastewaters in different forms, including total phosphorus, soluble phosphorus, and particulate phosphorus. In general, primary and secondary treatment is

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effective in removing particulate phosphorus. Phosphorus in the secondary effluents is mostly soluble and is present as orthophosphate (Duenas et al., 2003). Furthermore, only orthophosphate is consumed for phytoplankton growth causing eutrophication in surface water bodies (Selig et al., 2002). Consequently, tertiary treatment of secondary municipal effluents to remove orthophosphate has become increasingly necessary to meet environmental regulations worldwide.

There are a wide range of technologies available to remove phosphorus from wastewaters, such as chemical precipitation, biological treatment, adsorption processes, constructed wetlands, and a number of wastewater-and-sludge-based methods (Morse et al., 1998; de-Bashan and Bashan, 2004), which typically require considerable capital investment and maintenance costs for infrastructure and reagents. Thus, it is desirable to develop cost-effective processes for phosphorus removal from secondary wastewater effluents. In particular, P adsorption using iron oxide/hydroxide and aluminum hydroxide has been recently studied and found to be successful.

Kang et al. (2003) evaluated the adsorption of orthophosphate by iron oxide particles (ferrihydrite, goethite, and hematite) and found that ferrihydrite was most effective in removing phosphorus to a very low concentration. For ferrihydrite, the *Freundlich* isotherm parameters were $K = 19.4$ mg P/g and $n = 0.293$. Genz et al. (2004) studied the advanced phosphorus removal from membrane bioreactor effluents using activated aluminum oxide and granular ferric hydroxide (GFH). Both adsorbents were proven to be suitable for potential application in fixed bed adsorbers, while GFH showed better affinity and adsorption capacity at low phosphorus concentrations. The adsorption capacity of GFH ranged from 7.9 to 12.3 mg P/g, when equilibrium P concentrations increased from 0.1 to 0.3 mg P/L. Recovered ochre (i.e., $Fe(OH)_3$ or $FeO \cdot OH$-rich sludge) was tested for phosphorus removal from wastewater (Heal et al., 2004). It was found that ochre had a high adsorption capacity due to its high content of iron oxide/hydroxide (maximum adsorption capacity 17.8–21.5 mg P/g), and its potential to treat sewage effluent and agricultural runoff was assessed. Adler and Sibrell (2003) investigated the use of flocs from acid mine water neutralization to reduce the loss of soluble phosphorus from agricultural and animal wastewater, and found iron and aluminum-rich flocs were effective in controlling the release of soluble phosphorus from soil and manure. The adsorption capacities of acid mine drainage (AMD) flocs ranged from 10 to 20 mg P/g at an equilibrium concentration of 1 mg P/L. Galarneau and Gehr (1997) studied the capacity of aluminum hydroxide as an adsorbent to remove various forms of phosphorus. At a dose of 8 mol Al/mol P, the removal efficiency was 95%, 95%, and 40% for orthophosphate, condensed phosphate, and organic phosphate, respectively. Spent alum sludge, a byproduct from water treatment (mostly $Al(OH)_3$), was also demonstrated to be effective in the removal of phosphorus from wastewater (Yang et al., 2006). The maximum adsorption capacities ranged from 0.7 to 3.5 mg P/g when pH varied from 9.0 to 4.3. Furthermore, quartz particles coated with iron and aluminum oxides were evaluated as a filter media to remove phosphorus from wastewater (Arias et al., 2006). The maximum adsorption capacity was 4.4 mg/g of Fe-oxide-coated particles.

AMD, produced at both active and abandoned mines, is generally characterized by low pH and often high concentrations of dissolved metals (Fe, Al, Mn, and trace metals such as Pb, Cu, and Zn), resulting in environmental problems across the globe (Johnson and Hallberg, 2005). Typical AMD treatment processes rely on the removal of dissolved metals via precipitation. As a residual of this treatment approach, large volumes of sludge are produced. These sludges are a mixture of metal oxides/hydroxides and solids content is very low (1–5%). When compounded by the low economic value of the waste sludge, substantial difficulties in dewatering, and the high cost of off-site hauling, the ultimate disposal of AMD sludge is problematic (Ackman, 1982; Dempsey et al., 2001; Viadero et al., 2006). In an AMD site treating a flow of 100–400 gal/min (400–1500 L/min) in West Virginia, USA, approximately 30,000 m^3 of sludge was generated annually (Wei et al., 2005). Consequently, a major challenge in AMD treatment is the management of large volumes of loose sludge (Ackman, 1982; Kuyucak, 1998; Wei et al., 2005). The waste sludge is usually transported by pipeline and/or truck, and is typically disposed of through one of the following methods: deep mine disposal, retained-in-pond disposal, or disposal at coal refuse areas (Ackman, 1982; Matlock et al., 2002). However, the chemical composition of AMD sludge is mostly amorphous micron- and submicron-sized metal oxide/hydroxide particles. AMD sludge generally has a high specific surface area and numerous functional groups which are chemically active in an aqueous environment (Kirby et al., 1999; Cornell and Schwertmann, 2003; Wei and Viadero, 2007).

To date, very little work has been reported in the peer-reviewed literature on the beneficial use of AMD sludge for phosphorus removal. However, given the wide range of studies successfully reporting P removal from wastewaters using Fe and/or Al hydroxide sludges, it was hypothesized that the AMD sludge containing a mixture of iron and aluminum hydroxide precipitates would be a suitable medium for the adsorption of dissolved orthophosphate from solution. The goal of this study was to develop a potentially low-cost, sustainable treatment approach to phosphorus removal from secondary wastewater effluents in which a waste product from the mining industry is used for further benefit. Batch tests were carried out to study the adsorption kinetics, isotherms, and adsorptive thermodynamics in conjunction with studies on the effects of pH, temperature, concentration, and contact time for phosphorus removal from wastewater. Batch tests were followed by continuous flow tests using a continuous stirred tank reactor (CSTR). Based on the outcomes of these studies, the applications and implications of using AMD sludge as a tertiary process in municipal wastewater treatment were discussed.

2. Materials and methods

2.1. Adsorbent

AMD sludge was used as adsorbent in this study. The sludge was collected from a bond-forfeited treatment site in north central West Virginia, USA, where AMD was actively treated with hydrogen peroxide and anhydrous ammonia. The sludge