

Characteristics and mechanisms of phosphate adsorption on dewatered alum sludge

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Abstract

The adsorption characteristics of phosphate adsorption on the dewatered alum sludge were identified as a function of pH and ion strengths in solution. In addition, adsorption mechanisms were investigated by conducting batch tests on both the hydrolysis and P-adsorption process of the alum sludge, and making a comparative analysis to gain newer insights into understanding the adsorption process. Results show that the adsorption capacity decreased from 3.5 to 0.7 mg P/g sludge when the solution pH was increased from 4.3 to 9.0, indicating that adsorption capacity is largely dependent upon the pH of the system. The results of the competitive adsorption between phosphate and typical anions found in wastewater, such as SO_4^{2-} and Cl^- , onto alum sludge reveal that alum sludge can selectively adsorb phosphate ions. The insignificant effect of SO_4^{2-} and Cl^- on P-adsorption capacity indicates that phosphate adsorption is through a kind of inner-sphere complex reaction. During the adsorption process, the decrease of phosphate concentration in solution accompanied with an increase in pH values and concentrations of SO_4^{2-} , Cl^- and TOC (total organic carbon) suggests that phosphate replaced the functional groups from the surface of alum sludge which infers that ligand exchange is the dominating mechanism for phosphate removal. At the same time, the simultaneous decreases in PO_4^{3-} and total aluminium concentration in solution indicate that chemical reaction and precipitation are other mechanisms of phosphate removal.

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1. Introduction

Alum sludge is an inescapable by-product of the processing of drinking water in water treatment works where aluminium salt is used as the coagulant. Aluminium sulphate is arguably the most widely used coagulant in drinking water treatment. When aluminium sulphate is added to raw water, it dissociates into Al^{3+} and SO_4^{2-} . The Al^{3+} ions are immediately surrounded by water molecules and hexaquoaluminium ($[\text{Al}(\text{H}_2\text{O})_6]^{3+}$) is rapidly formed. The hexaquoaluminium formed then undergoes series of rapid hydrolytic reactions to form charged polymeric or oligomeric hydroxocomplexes of various structures. Such hydrolytic products include $[\text{Al}(\text{H}_2\text{O})_5\text{OH}]^{2+}$, $[\text{Al}(\text{H}_2\text{O})_4(\text{OH})_2]^+$, $[\text{Al}_6(\text{OH})_{15}]^{3+}$, $[\text{Al}_8(\text{OH})_{20}]^{4+}$ and $\text{Al}(\text{OH})_3(\text{s})$, etc. [1]. During coagulation in

water treatment process, these complexes adsorb and modify the surface charge of the colloidal particles, e.g. natural organic matter (NOM) such as humic and fulvic acid, microorganisms such as bacteria, protozoa and algae, and inorganic substances such as fine soil particles [2]. Thereafter, in the ensuing treatment units including flocculation, sedimentation and filtration, the colloids in the raw water are removed and transferred to the sludge phase together with the hydrolytic aluminium species.

In recent years, the management of alum sludge has become a significant issue in environmental engineering due to the enormous quantities generated and the associated disposal costs and constraints. In Ireland alone, a double-fold increase in alum sludge generation has been forecast by the end of the next decade, from a current estimate of 15,000–18,000 t/Pa of the dewatered solids. However, as a sustainable approach to mitigate these effects, current trends have indicated a progressive drive towards alum sludge reuse as beneficial material. Such beneficial reuses include the use of alum sludge as an adsorbent for phosphorus removal from wastewaters. In particular, previ-

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ous works done by the authors showed that the Irish dewatered alum sludge has a latent adsorption capacity, and it can be utilized as a “low-cost” phosphorus sorption medium in wastewater treatment [3,4]. In other instances, conjunctive evidences from literature have also shown that alum sludge can help remove phosphorus in wastewater. This is attributed to the abundant aluminum ions in the alum sludge, which enhance the processes of adsorption and chemical precipitation that help to remove such pollutants from wastewater [5–7]. However, an in-depth understanding of the mechanisms and characteristics of phosphorus adsorption by the alum sludge is crucial to its effective utilization as an adsorbent material.

Therefore, the aims of this study are: (1) to investigate the phosphate adsorption mechanisms of a dewatered alum sludge, (2) to identify the factors that affect the phosphate adsorption capacity of the alum sludge, and (3) to examine the structural identity of the phosphate impregnated alum sludge (PIAS).

2. Materials and methods

2.1. Preparation of test materials

Dewatered alum sludge cake (moisture content 72–75%) was collected from an industrial filter press of the sludge dewatering unit of a Water Treatment Works in Southwest Dublin, Ireland where aluminium sulphate is used as coagulant. After collection, the sludge cakes were air-dried and the moisture content decreased to 10.2% at the time of being used. The air-dried sludge was then ground and sieved to provide the test adsorbent with diameter <0.063 mm.

Artificial wastewater was synthesized by dissolving pre-weighed potassium dihydrogen phosphate (KH_2PO_4) in distilled water. The solution was then incubated in the laboratory at $20 \pm 2^\circ\text{C}$, and adjusted to different pH (using sulphuric acid (0.01 M) and sodium hydroxide (0.1 M)). Solutions were kept airtight to prevent CO_2 from affecting solution pH.

Chloride stock solution (6.0 M) and sulphate stock solution (3.0 M) were prepared by dissolving pre-weighed amount of NaCl and Na_2SO_4 , respectively, in distilled water.

2.2. Characterization of the alum sludge

Elemental, physical and chemical analyses of the dewatered alum sludge were carried out using Inductively Coupled Plasma-Atomic Emission Spectrometer (ICP-AES, IRIS Advantage), TOC-V CSH (Shimadzu) and Fourier Transform-Infrared (FT-IR, EQUINOX-55). The morphological structure of the dewatered alum sludge was examined by X-ray diffractometer (XRD, D/max-3C) and Scanning Electron Microscope (SEM, JSM-6700F).

2.3. Adsorption characteristics tests

2.3.1. Adsorption capacity

Batch tests were conducted by adding pre-determined amount of the sludge to a pH controlled distilled water (pH 4.3–9) and measuring the zeta potential. Sulphuric acid (0.01 M) and

sodium hydroxide (0.1 M) were used to control the pH of the adsorption system to designed values (pH 4.3–9) to investigate the effect of pH on adsorption behaviour/capacity and the amount of acid/alkaline required to maintain the pH was calculated from the concentration and volume added, including a correction for the dilution effect. Thereafter, standard aliquots of the phosphate stock solution was introduced, giving a resultant phosphate concentration of 5 mg P/l, which simulated the P level in typical municipal wastewater [8]. The mixtures were then mechanically agitated for promoting adsorption over a 48 h pre-determined equilibrium time [3]. After adsorption, equilibrated samples were filtered using 0.45 μm millipore filter paper (Millipore) and analysed for phosphate concentration. Adsorption capacities were evaluated from a linearized form of the Langmuir adsorption isotherm:

$$\frac{C_e}{q} = \frac{1}{Q_0} C_e + \frac{1}{Q_0 b} \quad (1)$$

where q is the mass of phosphorus adsorbed per unit mass of sludge; C_e the equilibrium concentration of phosphorus (mg P/l) in suspension after adsorption; Q_0 the maximum adsorption capacity (mg P/g sludge) and b is a constant related to the energy of the adsorption–desorption process with unit of inverse of concentration C_e . By plotting C_e versus C_e/q , a straight line with slope $1/Q_0$ is obtained and the Q_0 can be calculated.

Phosphate concentrations and pH values were obtained using a Hach spectrophotometer (DR/2400) and a pH meter (WTW, pH 325), respectively. The zeta potential of the alum sludge was measured at different pH using a zeta Potential Analyser (ZC-2000). All the adsorption tests were repeated twice and the average value of measurements was reported.

2.3.2. Anion selectivity of alum sludge and structural identity of PIAS

In order to investigate the selective adsorption of Cl^- , SO_4^{2-} and PO_4^{3-} by the alum sludge, pre-determined amount of the sludge was added to a pH modified distilled water (pH 4.3–9) followed by the introduction of Cl^- and SO_4^{2-} at concentrations ranging from 300 to 1000 mg/l. The total volume of the mixture was noted. Thereafter, 10 mg P/l of equal volume as the initial mixture volume was added giving a resultant P concentration of 5 mg P/l. The final mixture was then mechanically agitated for 48 h equilibrium time and residual phosphorus concentration determined as in Section 2.3.1. The structural identity of the PIAS was studied by investigating the effect of the ionic strength on the equilibrium of the surface complex formed.

2.4. Adsorption mechanism

2.4.1. Ligand exchange

To examine the possibility of ligand exchange in the adsorption mechanism, samples of alum sludge (5 g/l) were hydrolyzed in distilled water (initial pH 7.0 ± 0.02) in an airtight container and left for 28 days (final pH 5.98). Thereafter, phosphate concentration in the hydrolysed suspended solution was brought up to 5 mg P/l and phosphate adsorption by the hydrolyzed

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