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Research article

Optimization of aluminium recovery from water treatment sludge using Response Surface Methodology

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ABSTRACT

For decades, water treatment plants in Malaysia have widely employed aluminium-based coagulant for the removal of colloidal particles in surface water. This generates huge amount of by-product, known as sludge that is either reused for land applications or disposed to landfills. As sludge contains high concentration of aluminium, both can pose severe environmental issues. Therefore, this study explored the potential to recover aluminium from water treatment sludge using acid leaching process. The evaluation of aluminium recovery efficiency was conducted in two phases. The first phase used the one factor at a time (OFAT) approach to study the effects of acid concentration, solid to liquid ratio, temperature and heating time. Meanwhile, second phase emphasized on the optimization of aluminium recovery using Response Surface Methodology (RSM). OFAT results indicated that aluminium recovery increased with the rising temperature and heating time. Acid concentration and solid to liquid ratio, however, showed an initial increment followed by reduction of recovery with increasing concentration and ratio. Due to the solidification of sludge when acid concentration exceeded 4 M, this variable was fixed in the optimization study. RSM predicted that aluminium recovery can achieve 70.3% at optimal values of 4 M, 20.9%, 90 °C and 4.4 h of acid concentration, solid to liquid ratio, temperature and heating time, respectively. Experimental validation demonstrated a recovery of 68.8 ± 0.3%. The small discrepancy of 2.2 ± 0.4% between predicted and validated recovery suggests that RSM was a suitable tool in optimizing aluminium recovery conditions for water treatment sludge.

1. Introduction

Aluminium is the third most abundant element in the Earth's crust (Exley, 2009). It is often used as coagulants in water treatment process, usually in the form of alum, poly aluminium chloride, ferric aluminium sulphate and aluminium hydroxide chloride (Evuti and Lawal, 2011). During the treatment process, sludge equivalent to 4–7% of the total net of water produced was generated as by-product (Sun et al., 2015). Annually, a typical water treatment plant can yield approximately 100,000 tons of sludge comprising of silicon dioxide (SiO_2), aluminium oxide (Al_2O_3), iron oxide (Fe_2O_3) and a small fraction of other oxides (Ahmad et al., 2016a, 2016b). The composition of water treatment sludge together with the type of coagulant used from previous studies are compiled in Table S1 (Supplementary Materials). It can be observed that the types of coagulant used during the water treatment process possess some considerable effects on the sludge composition. For example, sludge treated with aluminium-based coagulants such as alum and poly aluminium chloride (PAC) will have higher aluminium content, whereas those treated with iron based-coagulants such as ferric chloride ($FeCl_3$) will contain more iron.

In common practice, water treatment sludge are either discharged into waterways or disposed to landfills (Geraldo et al., 2017). Malaysia employs similar practice, where water treatment sludge are dried in sludge lagoons prior its disposal to landfills that do not have adequate

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cover soil. Since alum is the widely applied coagulant, there is a high chance of aluminium leaching when in contact with acid rain. The free aluminium can cause significant environmental impacts which threaten the health of humans and animals (Teixeira et al., 2011). Thus, there is a need for aluminium recovery from the water treatment sludge in order to resolve its environmental impacts.

Generally, there are four methods to recover aluminium from the water treatment sludge, i.e. acid leaching, base leaching, ion exchanging and membranes (Ahmad et al., 2016a). Acid leaching is the preferred method due to its high efficiency and low cost as compared to the other methods (Huang et al., 2010; Chen et al., 2012). Some of the reaction mechanisms for acid and aluminium are shown in Table S2. According to literature, recovery of aluminium from treatment sludge using acid leaching process can range between 40 and 100% (Kyncl, 2008; Xu et al., 2009b; Chen et al., 2012). Several factors affecting the efficiency of acid leaching process include acid concentration, solid to liquid ratio, temperature and heating time (Jiménez et al., 2007; Nair and Ahammed, 2014; Jung et al., 2016). The evaluation of these factors to achieve optimum recovery condition is normally conducted by either traditional methods or the multivariate statistic methods (Bezerra et al., 2008). One factor at a time (OFAT) approach is the commonly employed traditional method (Ishikawa et al., 2007; Xu et al., 2009a; Chen et al., 2012). However, it ignores the interactions among the various variables and is time-consuming (Liyana-Pathirana and Shahidi, 2005). Thus, Design of experiments (DoE) approach, a statistically planned experiment, is introduced to provide an effective way to obtain the optimum conditions for responses with a limited number of experiments (Nair et al., 2014; Montgomery, 2017).

Amongst the multivariate statistic methods, Response Surface Methodology (RSM) is considered the most relevantly utilized for analytical optimization (Bezerra et al., 2008). It is also known as a collection of statistical and mathematical techniques which is useful for developing, improving and optimizing processes (Myers et al., 2016). RSM has been used in the studies of water treatment, but few focus on aluminium recovery from water treatment sludge (Nair and Ahammed, 2015; Jung and Ahn, 2016). Judging from the multiple factors involved in obtaining the optimum aluminium recovery conditions, RSM should be engaged to improve the performance of recovery and were proved to be a suitable tool for aluminium recovery prediction from water treatment sludge (Nair and Ahammed, 2014, 2017).

Therefore, this study aimed to investigate the recovery efficiency of aluminium from water treatment sludge using acid leaching process. Optimization of the recovery factors was conducted in two phases, namely OFAT (Phase 1) and RSM (Phase 2). Phase 1 examines the individual effect of each factor on aluminium recovery, while Phase 2 predicts the optimal condition of each factor as well as the potential aluminium recovery. To determine the effectiveness of RSM as an optimization tool, the predicted conditions were experimentally validated.

2. Materials and methods

2.1. Sample collection and preparation

The water treatment sludge was obtained from of a local water treatment plant located at Kota Tinggi, Johor, Malaysia. The water treatment sludge was collected at the sludge lagoon of the water treatment plant and stored in plastic containers. The samples were kept on ice during transportation and stored at 4 °C in the refrigerator before being used in experiments. The collected water treatment sludge was dried in an oven at a temperature of 105 ± 5 °C for 24 h to remove its moisture content. The dried sludge was then crushed using mortar and pestle followed by sieving with a wire mesh sieve of 150 μm to obtain powdered sludge. Aluminium in water treatment sludge is known to present in the form of kaolin (Awab et al., 2012; Fungaro and Silva, 2014; Bashar et al., 2016). Kaolin has been known for its high chemical inertness, which makes it not suitable to be used as a chemical reactant (Aderemi and Oludipe, 2000; Edomwonyi-Otu et al., 2010). However, literature have shown that conducting calcination process on kaolin can convert this chemically inert mineral into meta-kaolin, a mineral that reacts easier and is more active than kaolin (Ilić et al., 2010; Hosseini et al., 2011). Therefore, water treatment sludge was calcined at 700 °C for 30 min in furnace before being used in the leaching process. The conversion of kaolin into meta-kaolin is shown in Equation (1) below:

$$Al_2O_3{\cdot}2SiO_2{\cdot}2H_2O \rightarrow Al_2O_3{\cdot}2SiO_2 + H_2O \quad (1)$$

2.2. Sample characterization

The conversion of kaolin to meta-kaolin in collected water treatment sludge was verified with X-ray Diffraction (XRD) (Rigaku, Canada). Meanwhile, Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) (Agilent 710 Series, US) was selected for the identification of calcined sludge composition. Prior to ICP-OES analysis, sampled sludge was digested using microwave acid digestion in accordance to US-EPA 3051 SW-846 (2007). 1 g of sludge was mixed with hydrochloric acid and nitric acid at a ratio of 1:4. The mixture was then heated at 200 °C and 45 bar in the microwave (Milestone, Italy). Other parameters comprising of pH, Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Suspended Solids (SS) and Volatile Suspended Solids (VSS) were analysed according to APHA (2005) standard method. The sludge used in this study has a pH of 7.4, with 107.1 mg/L, 196.3, 12.6 g/L and 2.1 g/L BOD, COD, suspended solids and volatile suspended solids, respectively.

2.3. Acid leaching process for aluminium recovery

Sulphuric acid (QReC, New Zealand) was chosen as the leaching agent due to its availability and low cost (Park et al., 2007). 50 mL of sulphuric acid was added to sludge samples in a 250 mL conical flask and refluxed using a hot plate and Liebig condenser. The experimental range for each factor is specified in Section 2.4. At the end of the leaching process, the mixture was cooled down and centrifuged at 5000 rpm to separate the solids and leaching solution. The leaching solution was analysed using ICP-OES for aluminium analysis. All experiments were carried out under atmospheric pressure. Aluminium recovery was computed using Equation (2) in the following (Nair and Ahammed, 2014).

$$Al_{\%} = \frac{Al_{LC}}{Al_{WTS}} \times 100\% \quad (2)$$

where Al% is the percentage of aluminium recovery, Al_{LC} and Al_{WTS} are the amounts of aluminium in the leaching solution and water treatment sludge, respectively.

2.4. Optimization of aluminium recovery

To optimize the aluminium recovery factors, experiments were executed in two phases. Phase 1 determined the effects of acid concentration (2–8 M), solid to liquid ratio (10–25%), temperature (70–130 °C) and heating time (2–8 h) on the recovery of aluminium. The experiments were conducted by varying a single factor while keeping the values of all other factors fixed at a specific set of conditions. The subsequent Phase 2 focused on the elucidation of interactions between different factors involved during aluminium recovery using RSM with Central Composite Design (CCD). Aluminium recovery (Y) was chosen as the response variable while solid to liquid ratio (X_1), temperature (X_2) and heating time (X_3) were chosen as the independent variables. Experimental data obtained were fitted to the second-order polynomial model as shown in Equation (3) (Zhang et al., 2011; Jung and Ahn, 2016).