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Production of porous aluminium and iron sulphated oxyhydroxides using industrial grade coagulants for optimised arsenic removal from groundwater



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Introduction

Arsenic contamination of groundwater is a critical situation for millions of people who rely on polluted wells for drinking water, everyday consumption and agricultural activities [1]. People exposed to arsenic rich groundwater involuntarily accumulate this poisonous compound internally, which can, in the long-term lead to the development of arsenicosis. One of the first symptoms of this disease is the development of skin cancer on the foot and hand; the cancer may eventually spread to internal organs resulting in organ failure [2]. It is estimated that 2.5 million, the number of people exposed to arsenic contaminated water, may develop arsenicosis in the next 50 years [3].

Several different methods have been used to remove dissolved arsenic from groundwater. Membrane techniques, flocculation– coagulation, co-precipitation with dissolved metal oxides and adsorption techniques are considered the most widely applied methods [4]. Adsorption technique is the main technique applied in removing arsenic from groundwater because of its simple

ABSTRACT

The optimisation of Fe and Al oxyhydroxide materials produced using industrial grade coagulants is presented in this work. The effects of synthesis pH and post-synthesis washing procedure onto the arsenic adsorption capacity of the materials were investigated. It was shown that the materials produced at higher pH were more efficient in removing As(V), especially after cleaning procedure. The materials produced at lower pH were less efficient in removing As(V) but the higher presence of sulphate groups in the materials produced at lower pH enhanced As(III) adsorption. Most performing materials can remove up to 84.7 mg $As(V) g^{-1}$ or 77.9 mg $As(III) g^{-1}$.

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maintenance, low running cost and above all, the good understanding and familiarity of the technology by the local population, suppliers and engineering companies [5].

In order to increase the efficiency and lifetime of adsorption units, many researchers have developed innovative materials with enhanced adsorption capacity [6]. The materials developed to remove arsenic fall usually into: (i) a low cost option. (ii) a simple activation technic of low cost materials or the synthetic (iii) highly efficient sorbent option. Example of low cost materials includes the use of natural ores like Laterite, Bauxite or Ilmenite [7–9]; or the use of biosorbents like chitosan, chitin or biomass [10,11] as well as the use of industrial by-products like red mud or acid treated ore [12,13]. The main activation strategies that have been applied to materials for As removal include thermal activation [14] and chemical doping or activation [15]. The development of highly porous and/or reactive synthetic sorbents tailored towards As adsorption is also very often reported. Such as the production of iron and aluminium coated Organised Mesoporous Silica or highly porous zirconium oxides [16] and [17].

One of the major limitations in applying the developed materials from laboratory scale to on field application is related to both the cost of production of the highly efficient materials and the issue of securing the chemicals supply for these specific materials.

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The present study focuses on the development of engineered materials with relatively low cost and enhanced removal capacity by using commercial coagulants. It is the first report regarding the optimisation of iron and aluminium oxyhydroxides using commercial coagulants received directly from suppliers working in the wastewater treatment plant (WWTP) industry market. The main parameters governing the various materials' properties have been identified as the pH of synthesis and the post-cleaning process applied to the sorbent, and are thus investigated in terms of impact on the As removal capacity of the material. Maximum adsorption capacity of the materials and the effect of the pH on the adsorption of arsenic are discussed as well as the adsorption pathways followed for As removal by these porous aluminium and iron sulphated oxyhydroxides.

Materials and methods

Chemicals

Any chemicals that were used during the production of mesoporous iron and aluminium oxides were of reagent grade or higher, except for the iron and aluminium sulphate solutions which were received from an industrial supplier and are described below. The remaining chemicals used in the arsenic adsorption experiments were purchased from Sigma Aldrich and were of reagent grade or higher.

Materials production

Commercial coagulants

The metal hydroxides adsorbents were thus produced using widely available commercial coagulants using a simple procedure involving the hydroxylation of metal sulphate solution by sodium hydroxide. The coagulants used in this study were received directly from a local producer, Clinty Chemicals Limited, located in Ballymena, Northern Ireland.

Three different commercial coagulant solutions were received: an iron(III) sulphate solution, commonly referred to as ferric, an aluminium sulphate solution, often called alum, and a mixed solution of ferric and alum. The iron(III) and aluminium sulphate solution is usually called to FAS, for ferric aluminium sulphate.

Ferric sulphate is produced by dissolving iron(II) sulphate in water at 70 °C and then adding nitric acid which forces the oxidation of iron(II) to iron(III). Aluminium sulphate is produced by dissolving aluminium hydroxide in sulphuric acid at 70 °C. FAS is produced by mixing both ferric and alum solutions in the correct proportion. The process temperature is achieved by a combination

ALUM

of diluting acid solution with water (endothermic reaction) and by the use of steam generated on-site. The production process of coagulants is achieved on-site by a batchwise process.

Synthesis procedures

Alum, FAS and diluted ferric solutions were used to produce corresponding metal oxyhydroxides. H_2O_2 was used to increase the ORP (Oxidative Reduction Potential) of the solution which was already fairly high (from 200 to 500 mV). H_2O_2 was added until bubbling was spotted indicating that no more H_2O_2 was consumed. The pH was then gradually increased from starting pH (around 1) to synthesis pH by the addition of a concentrated NaOH solute on (30% w/w) under heavy stirring by means of an overhead stirrer. The first set of experiments was carried out with 200 mL of coagulant, and seven different synthesis pHs were tested.

The resulting slurry was allowed to rest for 16 h to cool down and separated at room temperature and was then filtered under vacuum. The recovered solid was repeatedly washed with distilled water and filtered a second time. The resulting cleaned slurry was dried at 120 °C for 16 h. A selection of the remaining materials was also cleaned after drying. The cleaning process involved using 5 g of dried material held in a stainless steel bucket and cleaned by ultrapure water in a 1 L beaker stirred by a magnetic stirrer. Fig. 1 illustrates the synthesis procedure followed.

All materials produced were dried at 60 °C for 24 h, crushed down and sieved to a particle size lower than 75 μ m. The resulting powder which was then used in adsorption experiments.

Materials nomenclature

pH 4 to 10

Washing

In the current study, 42 different materials were produced. To refer to the different category of materials produced, the following names are used:

- AlOx: refers to any oxyhydroxides material produced using alum.
- FeOx: refers to any oxyhydroxides material produced using ferric.
- FASOx: refers to any oxyhydroxides material produced using FAS.
- FAS(OH)s includes AlOx, FeOx and FASOx materials.

Otherwise the materials are named in function of the coagulant used, the pH of synthesis and the cleaning process used. Names are built as follows:

"Coagulant_Name"-"pH_synthesis"-"Washing_after_drying".

FAS-4-N

H₂O₂ and NaOH

Alu

FERRI

FAS

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