Arsenate removal from water using sand–red mud columns

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Abstract

This study describes experiments in which sorption filters, filled with chemically modified red mud (Bauxsol) or activated Bauxsol (AB) coated sand, are used to remove As(V) (arsenate) from water. Bauxsol-coated sand (BCS) and AB-coated sand (ABCS) are prepared by mixing Bauxsol or AB with wet sand and drying. Samples of the BCS and ABCS are also used in batch experiments to obtain isotherm data. The observed adsorption data fit the Langmuir model well, with adsorption maxima of 3.32 and 1.64 $mg\,g^{-1}$ at pH values of 4.5 and 7.1, respectively for BCS; and of 2.14 $mg\,g^{-1}$ for ABCS at a pH of 7.1. Test results show that higher arsenate adsorption capacities can be achieved for both BCS and ABCS when using the columns compared to results for batch experiments; the difference is greater for BCS. Additional batch tests, carried out for 21 days using BCS to explain the observed discrepancy, show that the equilibrium time previously used in batch experiments was too short because adsorption continued for at least 21 days and reached 87% after 21 days compared to only 35% obtained after 4 h. Fixed bed column tests, used to investigate the effects of flow rate and initial arsenate concentration indicate that the process is sensitive to both parameters, with lower flow rates (longer effective residence times in the columns) and initial arsenate concentrations providing better column performance. An examination of the combined effect of potential competing anions (i.e. silicate, phosphate, sulphate and bicarbonate) on the column performance showed that the presence of these anions in tap water slightly decreases arsenate removal. Each breakthrough curve is compared to the Thomas model, and it is found that the model may be applied to estimate the arsenate sorption capacity in columns filled with BCS and ABCS. The data obtained from both batch and column studies indicate that BCS and ABCS filtration could be effectively used to remove arsenate from water, with the latter being more efficient.

1. Introduction

Arsenic (As) is a known carcinogen that poses serious health risks to humans, e.g. chronic exposure to arsenic from food, air, soil, and drinking water can induce cancers of the skin and internal organs, and can also cause unwanted cardiovascular and neurological effects (Tseng, 1977; Mandal et al., 1998). Exposure to arsenic usually occurs through the ingestion of arsenic present in drinking water and food (Le, 2002). Arsenic may be present at elevated concentrations in surface and subsurface waters as a consequence of the natural weathering of arsenic bearing minerals (especially arsenopyrite), particularly where the pumping of groundwater allows increased oxygen penetration into

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the soil, where it can accelerate the oxidative decomposition of arsenic containing sulphide minerals and produce acid that may leach arsenic from other minerals. Arsenic concentrations in surface and ground waters can also be increased as a consequence of the anthropogenic discharge of arsenic waste from petroleum refining, glass melting, and smelting of metal sulphide ores (e.g. of lead, copper, zinc, gold and silver) or from the use of arsenic containing pesticides and fertilizers (Bothe and Brown, 1999). Consequently, there is increasing public concern about arsenic contamination in water and its effects on human health (Karim, 2000).

Arsenic can occur in natural waters both in organic and inorganic forms, but laboratory scale treatment technologies are generally tested to remove inorganic arsenic from water; as the inorganic arsenic species (arsenite, As(III) and arsenate, As(V)) are the major arsenic species in natural waters. Arsenite is the chemically dominant form in reducing environments e.g. anoxic groundwater; whereas, arsenate is the dominant form in oxidising conditions. Given the likely dominance of arsenate in potentially potable water, and the clearly evident need to remove arsenate from such water in several countries this study examines only arsenate removal; arsenite, if present, can be readily oxidised by conventional means for removal as arsenate. Furthermore, previous studies using activated Bauxsol (AB) (Genç-Fuhrman et al., 2004b) have shown that arsenite should be oxidised to arsenate for optimal arsenic removal.

Most people affected by arsenic contamination worldwide are poor, live in small communities and rely on tube wells as a source of drinking water; e.g. in West Bengal and Bangladesh (Thirunavukkarasu et al., 2003). They therefore need an appropriate method that is cheap, simple to use and adaptable to either a household or a village scale. One such method could involve the use of a fairly cheap and readily available material, such as red mud, which is an insoluble residue generated at alumina refineries using the Bayer process. Seawater-neutralised red mud (Bauxsol) and AB have previously been investigated for arsenic removal from water (Genç et al., 2003; Genç and Tjell, 2003; Genç-Fuhrman et al., 2004a, b), and these studies have shown that they can be effectively used for arsenic removal primarily due to their high aluminum and iron oxide/hydroxide content. In practise, an additional advantage of Bauxsol and AB over other chemical methods is that no toxic sludge is produced as a result of arsenic removal and the spent Bauxsol or AB will satisfy the toxicity characteristics leaching procedure (TCLP) criteria for classification as an inert waste (Genç et al., 2003; Genç-Fuhrman et al., 2004a); such a classification substantially reduces costs and risks associated with the management of the spent sorbent.

As a result of the promising results obtained in batch tests, this study was developed to investigate the possibility of using Bauxsol and AB under continuous flow conditions. With our knowledge only Ho et al. (1991) tested the use of sand and red mud (raw material of Bauxsol and AB) in columns to remove bacteria and viruses, but did not investigate the sorption of inorganic contaminants like arsenic. Consequently, this study is designed to: (i) develop new sorbents from Bauxsol and AB (ordinary Bauxsol and AB particles are too fine to use in column tests) that are suitable for use in column tests, (ii) conduct batch tests to examine arsenate adsorption using the new sorbents, and (iii) conduct column studies to investigate the arsenate uptake characteristics of the new sorbents under different flow rates, inflow arsenate concentrations, and in the presence of common anions in the influent water.

2. Materials and experimental methods

2.1. Experimental solutions

Deionised water is used in the preparation of standard solutions and for dilution of the samples. High purity sodium hydrogen arsenate heptahydrate ($Na_2HAsO_4 \cdot 7H_2O$) purchased from Sigma, is used to prepare a $1\,g\,L^{-1}$ arsenate stock solution by dissolving 4.164 g of the powder in 1 L deionised water. Secondary stock solutions of 100 and $10\,mg\,L^{-1}$ arsenate were prepared weekly from the $1\,g\,L^{-1}$ stock solution, and the required working standards were prepared daily from the secondary stock solutions. Laboratory tap water from the city of Lyngby, Denmark is spiked with the required quantities of arsenate (in case of C2 also with phosphate, silicate, sulphate and bicarbonate) and used in the batch and column studies. Major physicochemical characteristics of the used tap water are summarised in Table 1. Examination of the possible influence of anions in the water used stock solutions containing $1\,g\,L^{-1}$ of phosphate, silicate, sulphate, and $10\,g\,L^{-1}$ bicarbonate that were prepared using NaH_2PO_4, $Na_2SiO_3 \cdot 5H_2O$, Na_2SO_4, and $NaHCO_3$, respectively. All glassware and sample bottles were soaked in 6–10% nitric acid (HNO_3) for at least 12 h, and rinsed with deionised water four times before use. All chemicals used in this study are of analytical grade and are used without additional purification.

2.2. Bauxsol-coated sand (BCS) and activated Bauxsol-coated sand (ABCS)

Bauxsol is prepared by treating the caustic (pH > 13) red mud from an alumina refinery with seawater (it may also be prepared by using other magnesium- and calcium-rich brines, or a mixture of magnesium chloride