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Technical note

A simple technology for arsenic removal from drinking water using hydrotalcite

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Abstract

The use of a synthetically prepared clay material, hydrotalcite (HT), for the removal of arsenite (As(III)) and arsenate (As(V)) from drinking water is described. Percolation through HT of water containing 500–1000 μg/L As (levels often found in As-contaminated well water) produced leachate with As levels well below 10 μg/L. The technology could be coupled to that used in less-developed regions for removing organisms from drinking water, viz. leaching through porous pots and filter candles. The 'spent' HT is easily converted into valuable phosphatic fertilizer that would have an insignificant effect on soil arsenic levels, thereby reducing the overall cost of manufacture and distribution.

Keywords: Arsenic removal; Drinking water; Hydrotalcite; Phosphate fertilizer

1. Introduction

The presence of arsenic in drinking water, at levels shown to be a significant health hazard, is receiving increasing attention worldwide (Nordstrom, 2002). Levels of arsenic in excess of the World Health Organization recommended limit of 10 μg/L can occur naturally in water bodies used as sources of drinking water, but the problem is exacerbated by the sinking of wells in areas of arsenic-rich geological strata. Values in the range 10–1000 μg/L have been reported and the UN Synthesis Report on Arsenic in Drinking Water (www.who.int/water_sanitation_health/dwq/arsenic3) provides an excellent overview of the extent of the problem and of technologies being evaluated for its solution.

This paper canvasses the use of a synthetic clay mineral, hydrotalcite (HT), as a material for removing arsenic from water at household or communal level in less-developed regions. Hydrotalcite is essentially an aluminium-substituted brucite ($Mg(OH)_2$) mineral and forms when a proportion of the magnesium ions in the structure is replaced by aluminium, leading to an excess of structural positive charge. The positively charged sheets align, with interlayer spacings being occupied by *anions* to maintain electrical neutrality. This process is the reverse of the adsorption reactions involving smectitic clays, where structural negative charge is balanced by interlayer *cations*. Hydrotalcite occurs naturally in specimen amounts under favourable conditions, but can be easily synthesised using inexpensive raw materials such as bauxite and magnesite (Gillman and Noble, 2005).

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The term 'hydrotalcite' is generally reserved for the form wherein carbonate ions occupy the interlayer spacings. When other interlayer anions such as nitrate, chloride and phosphate are dominant, the materials are referred to as 'hydrotalcite-like compounds'. In this paper, the general term 'hydrotalcite' (HT) will be applied irrespective of the charge-balancing anion and an anion prefix used to describe a particular form, e.g. NO_3–HT and Cl–HT for nitrate and chloride hydrotalcite, respectively.

Hydrotalcite specificity for anions follows the order $CO_3 > PO_4 > SO_4 > Cl > NO_3$ (Tamagawa, 2003) and, in the absence of carbonate, a chloride or nitrate HT will readily adsorb phosphate from solution as was demonstrated by Ookubu et al. (1993). The similarity between the structure of the arsenic anions arsenite and arsenate to that of phosphate anions indicates that HT could be used to effectively remove arsenic from water. This paper therefore presents results of laboratory experiments comparing the capacity of several HT materials to adsorb arsenic, followed by prototype technology studies that might be adaptable for use in regions referred to above.

2. Adsorption of As(III) by Cl–HT, NO_3–HT and CO_3–HT

2.1. Materials and methods

Arsenic measurements were made with a Varian UltraMass 700 ICP-MS, externally calibrated using a commercially available standard solution, and also an In internal standard to correct for instrument drift and matrix effects. The limit of detection for As under the conditions used was 0.42 µg/L.

To obtain an approximate 500 µg/L As(III) stock solution, a saturated arsenic trioxide (As_2O_3) solution that was approximately 10,000 mg/L As was diluted by a factor of 20,000. The actual As(III) concentration was determined by ICP-MS to be 432 µg/L.

The synthesis of HT is easily effected by co-precipitation of a Mg/Al double hydroxide. The ratio of Mg/Al should be held between 0.25 and 0.33, and pH must be higher than that at which either Mg or Al hydroxide would separately precipitate. In the three syntheses described below, pH was maintained at or above 9.5. XRD analysis was carried out on the three HT compounds to confirm that true HT materials had been formed.

An NO_3–HT slurry was prepared by adding 500 mL of a mixed nitrate solution (1 M $Mg(NO_3)_2$, 0.5 M $Al(NO_3)_3$) via a peristaltic pump at 450 mL/min to 250 mL of concentrated ammonia (25%) with vigorous stirring. The final pH of the HT precipitate was 9.5. Following a number of centrifuge-washings to the point of dispersion to virtually remove all ammonium nitrate, the NO_3–HT 'cake' (moisture content 87%) was stored in a sealed container.

A CO_3–HT slurry was prepared by bubbling CO_2 through an NO_3–HT slurry (see above) for 2 h. After centrifuge-washing to the point of dispersion to remove residual nitrate, the CO_3–HT 'cake" (moisture content 89%) was stored in a sealed container.

A Cl–HT solid was prepared using the method of Ookubu et al. (1993). A 350 mL solution of mixed chloride (0.86 M $MgCl_2$, 0.4 M $AlCl_3$) and, separately, a 2.5 M NaOH solution were pumped into a beaker under vigorously stirred conditions, with adjustment of pump rates to maintain pH of the resultant slurry at about 9.5. The amount of alkali consumed was 420 mL. The slurry was aged at 80 °C for 8 h, followed by centrifuge-washing to the point of dispersion and drying of the 'cake' at 80 °C.

In separate experiments, 0.1, 0.5, 1.0 and 2.5 g (o.d. basis) of NO_3–HT cake or Cl–HT powder were added to 500 mL aliquots of the stock As(III) solution that initially contained 432 µg/LAs. In another experiment, 2.5 g (o.d. basis) of CO_3–HT cake was added to 500 mL of As stock solution. The suspensions were stirred in an open beaker for 1 h and a small sample of supernatant solution then taken for As analysis. After a further 17 h stirring, another supernatant solution sample was taken. The samples were passed through a 0.45 µ membrane filter prior to As determination.

2.2. Results and discussion

XRD patterns of the three synthesised hydrotalcite clays are presented in Fig. 1. Basal spacings calculated from the 003 reflections (near 10° 2Θ) were 0.89 nm, 0.77 nm and 0.78 nm for NO_3–HT, CO_3–HT and Cl–HT, respectively, values that are in very good agreement with published basal spacings. The sharpness of the 003 and 006 peaks demonstrates the layered structure of these materials.

The concentrations of As(III) remaining in solution following contact with NO_3–HT and Cl–HT, over a range of HT addition rates, and for two contact times, are summarized in Table 1. (Addition of HT in carbonate form did not alter solution As concentration and CO_3–HT will not be further discussed.)

When added in sufficient quantity, both NO_3–HT and Cl–HT reduced As(III) concentrations from those