



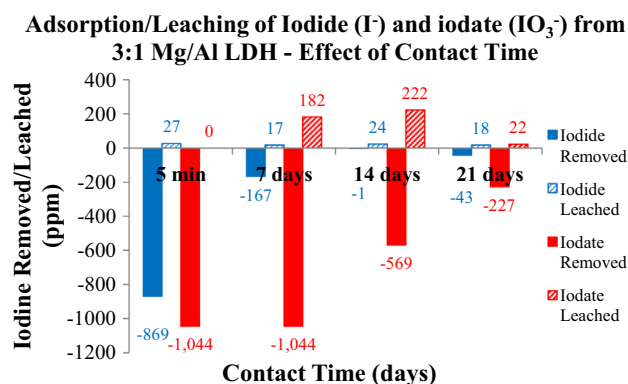
Regular Article

Leaching of iodide (I^-) and iodate (IO_3^-) anions from synthetic layered double hydroxide materials

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GRAPHICAL ABSTRACT



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ABSTRACT

Several studies have previously demonstrated that layered double hydroxides (LDHs) show considerable potential for the adsorption of radioiodine from aqueous solution; however, few studies have demonstrated that these materials are able to store radioactive ^{131}I for an acceptable period. The leaching of iodide (I^-) and iodate (IO_3^-) from Mg/Al LDHs has been carried out. Contact time appeared to be a more significant variable for the leaching of iodate (IO_3^-) compared to that of iodide (I^-). Experimental results are fitted to the pseudo second order model, suggesting that diffusion is likely to be the rate-limiting step. The presence of carbonate in the leaching solution appeared to significantly increase the leaching of iodide (I^-) as did the presence of chloride to a lesser extent. The maximum amount of iodate (IO_3^-) leached using ultrapure water as the leaching solution was 21% of the iodate (IO_3^-) originally present. The corresponding result for iodide (I^-) was even lower at 3%.

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1. Introduction

Iodine is an essential element for healthy thyroid development where it is required for the production of a number of hormones. The recommended daily intake of iodine for optimal health is

between 80 and 150 mg/day though many people around the world suffer from iodine deficiency [1–3]. While iodine is not toxic to humans in low doses, several radioactive isotopes (or radioisotopes) of iodine exist which may be taken up by the body and concentrate in the thyroid resulting in a possible detriment to human health [1–3]. The primary iodine radioisotope of concern is the ^{131}I isotope, which is a major by-product of uranium fission and is important consideration for nuclear energy industries and

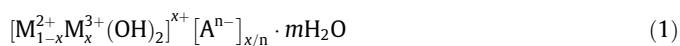
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regulators [1,2]. The ^{131}I isotope is also used extensively in medical applications resulting in wastes that need to be properly stored and disposed of to ensure public safety.

It is clear that the unexpected leaching of radioactive iodine species from an adsorbent could result in a serious hazard; however, as the ^{131}I isotope has a comparatively short radioactive half-life of around 8 days it may only prove necessary to immobilise the ^{131}I species for a period of weeks or months. Once the radioactivity of the material has decayed to a level that is no longer dangerous, the remaining iodine could be removed and the adsorbent reused or recycled. Layered double hydroxides (LDHs) are possible candidate materials for this application as their thermally activated products typically have a high uptake capacity towards anionic species and the reformed LDHs may readily exchange anions under the correct conditions [4–6].

LDHs are a group of anionic clay materials that have been investigated as adsorbents for the removal of various anionic pollutants from aqueous solution. LDHs (also known as hydrotalcite like materials) consist of a layered brucite ($\text{Mg}(\text{OH})_2$) like structure in which a portion of the divalent cations (M^{2+}) has been substituted with a trivalent cation (M^{3+}). The result of this substitution is a layered structure which carries a net positive charge that must be balanced by exchangeable anions intercalated into the interlayer regions [7–9]. LDHs are represented by the general formula:



where M^{2+} is the divalent cation, M^{3+} is the trivalent cation, A^{n-} is the exchangeable anion (usually carbonate, chloride or nitrate) and x is usually $0.2 < x < 0.33$ [7–9]. The structure and composition of LDH materials have been reported in numerous publications [4–6], while the nomenclature and composition of the LDH supergroup have been reviewed more recently by Mills et al. [10]. Calcination of a LDH at 300–500 °C results in the decomposition of the LDH structure and the elimination of interlayer anions and water. The resulting mixed metal oxide (often referred to as a thermally activated LDH) may undergo spontaneous reformation when exposed to a solution containing suitable anions. This reformation (or memory) effect is often utilised to improve the anion uptake capacity of LDHs [4–6]. Reformation involves the reconstruction of an LDH when the mixed metal oxides are exposed to water. Anions in the solution such as iodide (I^-) or iodate (IO_3^-) are intercalated into the interlayer region of the LDH to replace the charge balancing anions lost during thermal activation.

Recently our research group published a literature review which included the application of LDHs for the removal of iodine from aqueous solution, which has heavily influenced the direction of the work reported in this article [11]. While Layered double hydroxides (LDHs) and related materials have been widely investigated as sorbents for the removal of various toxic anions from water (including iodine species), the possibility of leaching of the adsorbate from the LDH sorbent is usually overlooked. In this article we report on some experiments designed to assess the amount of leaching of radioiodine from LDH materials under ordinary atmospheric and temperature conditions. Significant leaching of iodine species from LDHs materials over the first 3–4 weeks would pose a significant hazard, making LDHs unsuitable for the removal of radioiodine from aqueous solution. As this experiment is concerned with leaching, variables affecting the adsorption of iodide (I^-) or iodate (IO_3^-) will not be discussed here, but are presented in other publications.

2. Experimental reagents

All reagents used in the experiments described in this article were AR grade except for the potassium iodate (KIO_3) used to pre-

pare the stock solutions, which was LR grade. Chemicals were used as received with no additional purification or pre-treatment unless otherwise stated.

3. Experimental procedure

3.1. Synthesis of the Mg/Al LDH adsorbent

The main adsorbent used in this study was the 3:1 Mg/Al LDH, the preparation of which has previously been described by our group in [12]. A large batch of the 3:1 Mg/Al LDH (ideal formula: $\text{Mg}_6\text{Al}_2(\text{OH})_{16}(\text{CO}_3)\cdot 4\text{H}_2\text{O}$) was synthesised by a straightforward co-precipitation method from a mixed metal solution containing aluminium chloride hexahydrate (603.6 g) and magnesium chloride hexahydrate (1524.7 g) were dissolved in deionised water (10,000 mL). A caustic solution containing sodium hydroxide (800.2 g) and sodium carbonate (1690.9 g) in deionised water (10,000 mL) was used to precipitate the LDH. The pH of the solution was monitored throughout the reaction with additional sodium hydroxide was added to ensure the pH remained above 10.0. The LDH was collected by vacuum filtration and with a 0.1 M sodium carbonate solution to exchange any possible interlayer chloride. Further washing of the LDH in ultrapure water was required to remove a sodium chloride synthesis by-product. The LDH was dried in an oven under air and ground to a uniform consistency with the aid of a mortar and pestle.

3.2. Characterisation of LDH adsorbents

Characterisation of the 3:1 Mg/Al LDH adsorbent materials used in this study has previously been reported by our group in [12–15].

3.3. Preparation of Mg/Al LDHs with adsorbed iodine species

In order to perform leaching experiments it is necessary to obtain an LDH material that contains adsorbed iodine species. One possible option would be to attempt the direct synthesis of a LDH with interlayer iodine species; however, this may be difficult due to the low affinity LDHs typically exhibited for iodide (I^-) and iodate (IO_3^-) when compared to other competing anions that may be present during synthesis (including carbonate, nitrate, chloride and hydroxide). Additionally, LDHs may undergo some irreversible decomposition during thermal activation (particularly at high temperatures) which would not be replicated using this approach. A second approach is to perform an independent adsorption step to simulate the kind of sorption that would be employed in the actual application.

Selected layered double hydroxides were thermally activated by calcination under air in a muffle furnace. Samples were weighed into open ceramic crucibles, which were then placed in the furnace. Once the furnace reached the desired temperature heating was stopped and the sample was allowed to remain in the furnace for 1 h as it slowly cooled. At the conclusion of that hour, the thermally activated LDHs were removed from the furnace and allowed to cool further before storage under vacuum until needed for characterisation or adsorption experiments.

LDH samples containing adsorbed iodine species were prepared by adding the required mass (usually 0.50 g) of LDH adsorbent to 20 mL of iodide (I^-)/iodate (IO_3^-) solution (usually with an approximate concentration of 1000 ppm) pipetted into 50 mL centrifuge tubes, which were used as reaction vessels for the experiment. The capped centrifuge tubes were capped placed on a rotary suspension mixer for one week ensuring that adsorption was complete. After the adsorption step was complete the resulting mixture was separated by centrifuging with the LDH adsorbent residue

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