



Removal efficiency of fluoride by novel Mg-Cr-Cl layered double hydroxide by batch process from water

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Abstract

The fluoride ion removal from aqueous solution using synthesized Mg-Cr-Cl layered double hydroxide has been reported. Mg-Cr-Cl was characterized by X-ray powder diffraction, Fourier-transform infrared, thermo-gravimetric analysis, differential thermal analysis, and scanning electron microscope. Adsorption experiments were carried out in batch mode as a function of adsorption dosages, contact time, pH, and initial fluoride concentration to get optimum adsorption capacity. The adsorption kinetic study showed that the adsorption process followed first order kinetics. The fluoride removal was 88.5% and 77.4% at pH 7 with an adsorbent dose of 0.6 g/100 mL solution and initial fluoride concentration of 10 mg/L and 100 mg/L, respectively. The equilibrium was established at 40 min. Adsorption experiment data were fitted well with Langmuir isotherm with $R^2 = 0.9924$. Thermodynamic constants were also measured and concluded that the adsorption process was spontaneous and endothermic in nature. The removal percentage decreased slowly with increasing pH. This process is suitable for industrial effluents. The regeneration of the material is not possible.

Key words: layered double hydroxide (LDH); fluoride; adsorbents

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Introduction

In a class of anionic clays materials, layered double hydroxides (LDHs) have general formula $[M^{II}_{1-x}M^{III}_x(OH)_2]^{x+}(A^{n-})_{x/n} \cdot yH_2O$, where M^{II} and M^{III} is a divalent and trivalent metal cations respectively, and A^{n-} , generally represented as a valence anion. The structure of hydroxide layers of LDH may be derived from mineral brucite or $Mg(OH)_2$ octahedral layers in which M^{2+} ions are partially exchanged by M^{3+} ions yielding positively charged $[M^{II}_{1-x}M^{III}_x(OH)_2]^{x+}$ layers. These layers are balanced by A^{n-} anions sandwiched between the layers. The interlayer space is occupied by both anions and water molecules (Cavani et al., 1991; Rajamathi et al., 2001; Del Arco et al., 1999).

In recent years LDHs are widely used as host material to trap anionic pollutants from solutions thus having a good potential in remediation of environmental contaminants (Chitrakar et al., 2008). LDHs show high anion exchange capacity and incorporate a wide variety of anions into the inter-layer (Khan et al., 2002; Kim et al., 2005; Carlino, 1997). By virtue of this advantage, they have the potential

to scavenge hazardous anionic contaminants present in wastewater (Foss et al., 2000; Houri et al., 1998; Tezuka et al., 2004). LDH structures can trap contaminants such as nonionic organic pollutants (Dékány et al., 1997), phenols (Yapar and Yilmaz, 2004), radioactive ^{131}I (Kang et al., 1999; Fetter et al., 1997), chromate, and selenate/selenite (You et al., 2001) nitrate (Tezuka et al., 2004), phosphate (Daou et al., 2007), and arsenate(V) ions (Yang et al., 2006). They have also received considerable attention in field of medicines, nano scale reactions, in separation technology, bio-molecule immobilization processes, especially used as adsorbents, catalysts, catalytic precursors, electrochemical etc. (Lucredio and Assaf, 2006; Kantam et al., 2006). However, the adsorption process by using LDH for different inorganic anions, particularly F^- , has received less attention and is not well understood.

Fluoride is an essential mineral for the growth of dental and bones in mammals, but excessive intake of it through foods and drinks cause's dental and skeletal fluorosis. Groundwater contaminated with excess fluoride mainly in the concentration range of > 1.5 mg/L is hazardous to health; the permissible limit of fluoride in drinking water is 1.5 mg/L (WHO, 1984). The fluoride toxicity

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in groundwater has imposed a serious problem to meet the requirements of drinking water in many countries like India, China, Sri Lanka, West Indies, Spain, Holland, Italy, Norway and Mexico. Therefore, several technologies have been developed to reduce the amount of fluoride to lower levels from groundwater and other source of waters, by addition of chemicals to cause precipitation-coagulation (Hu et al., 2005), ion-exchange (Castel et al., 2000), adsorption (Fan et al., 2003), membrane processes (Pontie et al., 2003), electrolytic de-fluoridation (Mameri et al., 2001), electro dialysis (Hichour et al., 2000), etc. In ion exchange method the percentage of removal is decreased in presence of other ions and alkalinity. Regeneration of resin is a problem because it leads to fluoride rich waste; this process is expensive because of the cost of resin, regeneration and waste disposal. In the removal by membrane process, all the ions present in water are removed, even though few ions are very much essential for living organism. Further remineralization is required when the treated water are use especially for drinking water. The process is cost bearing as the water becomes acidic and needs pH correction; lot of water gets wasted as brine. The electro dialysis method is not feasible for large amount of contaminants and is expensive due to cost of membrane, regular replacement of membrane. Commonly adopted Nalgonda technique for removal of fluoride is based on precipitation and coagulation, where precipitation is 18% to 33% and forms soluble aluminum fluoride complex ion, which itself is toxic (Apparao et al., 1990). Due to use of aluminum sulfate as coagulant, the concentration of aluminum and sulfate increases, and this process is not automatic, which requires a regular attention and maintenance, this makes cost expensive of the process. Considering the limitations and difficulties of the above processes we emphasize adsorption treatment which is cost-effective and it can remove fluoride up to 90%.

Adsorption of fluoride on activated alumina (Hao and Huang, 1986), zeolite (Onyango et al., 2004), hydroxypapatite (Sundaram et al., 2008), alum, carbon (Li et al., 2003), bone char (Montoya et al., 2007), fly ash (Chaturvedi et al., 1990), red mud (Cengeloglu et al., 2002), lime, clay, aluminum sulfate, magnesite, dolomite etc. has already been reported. Adsorptions with LDHs have benefitted water purification technologies in many ways; however recently Mg-Al-LDHs has reported by Xin-Yao for the use of for removal of Cr(VI) and As(V) from water, they synthesized the material through simple solvothermal method. Further they used these materials for the removal of trivalent and pentavalent arsenic from water (Yu et al., 2012). The principal objective of present work is to study the fluoride uptake behavior of Mg-Cr-Cl layered double hydroxide. Mg-Cr-Cl LDH was prepared by co-precipitation synthesis routes, and was characterized by X-ray powder diffraction (XRD), Fourier-transform infrared (FT-IR), thermo-gravimetric analysis

(TGA), differential thermal analysis (DTA), and scanning electron microscope (SEM). Further removal mechanism of fluoride from drinking water has been evaluated by taking different relevant parameters into account.

1 Experimental

1.1 Reagents and chemicals

All chemicals used in this study were of analytical grade obtained from E. Merck. All synthesis was done using distilled water. Stock solutions of fluoride were prepared by dissolving 2.21 g of sodium fluoride in 1 L distilled water. The solutions of different concentration were prepared by diluting the freshly prepared stock solution by distilled water. The beaker, funnel, measuring cylinder, volumetric flask and conical flask used were of Borosil. Only PVC wares were used for handling fluoride solution. All PVC wares were washed in dilute HNO₃ acid bath and rinsed thoroughly with distilled water prior to use.

1.2 Synthesis of Mg-Cr-Cl layered double hydroxide

The synthesis of Mg-Cr-Cl LDH was done by the standard process: Two solutions A, B were prepared separately. Solution A was prepared having equal volume of MgCl₂ and CrCl₃ (with Mg²⁺/Cr³⁺ molar ratios of 3:1). The solution A was added drop wise by a peristaltic pump to an alkaline solution B containing equal volume of (2 mol/L NaOH, 1 mol/L NaCl) under vigorous stirring. The light blue suspension developed was further stirred for 18 hr at 80°C maintaining the pH at 10.0 ± 0.5 by subsequent addition of NaOH. Then the solid was recovered by filtration, washed several times with deionized water to remove excess of hydroxide present, and dried overnight at 40°C. The resulting solids were obtained which is used in characterization study and adsorption experiments.

1.3 Characterization methods

FT-IR of the samples were recorded using PerkinElmer FT-IR spectrophotometer SPECTRUM RX-I with 16 scans and over the wave number range 400–4000 cm⁻¹. The sample was pressed into KBr pellets with sample to KBr ratio as 1:50, at a pressure of 2 tons. The thermal behavior of the LDH was determined from thermo-gravimetric analysis and differential scanning calorimetry (TGA/DSC) analysis carried out using NETZSCH STA 409C. The sample of 30 mg was used and alumina was used as reference. Scanning electron micrographs of the sample was obtained by JEOL JSM-6480LV scanning electron microscope. The sample was coated with platinum for 30 sec at a current of 50 mA before the SEM micrograph was obtained. Powder XRD patterns of the samples were recorded using PHILLIPS X'PERT X-ray diffractometer. Scanning was carried out using CuK α radiation (wavelength 1.5418 Å) at a scan rate of 1°/min and was analyzed using software provided with the instrument.

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