

Hydrated cement: A promising adsorbent for the removal of fluoride from aqueous solution

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Abstract

The present study was carried out to investigate the potential of cement hydrated at various time intervals for the removal of excess F^- from aqueous solution by using batch adsorption studies. The influence of different adsorption parameters, viz. effect of adsorbent dose, initial concentration, pH, interfering ions and contact time were studied for their optimization. It was observed that the adsorbent exhibited reasonably significant F^- removal over a wide range of pH. The presence of carbonate and bicarbonate ions in aqueous solution were found to affect the F^- removal indicating that these anions compete with the sorption of F^- on adsorbent. The equilibrium adsorption data were fitted well for both the Freundlich and Langmuir isotherms and the adsorption capacities were calculated. Comparative studies for F^- removal in simulated and field water show relatively higher F^- removal in simulated water. XRD and SEM patterns of the hydrated cement were recorded to get better insight into the mechanism of adsorption process. From the experimental results, it may be concluded that HC was an efficient and economical adsorbent for F^- removal.

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1. Introduction

It is well documented that trace elements are essential and beneficial to human health in minute concentrations, as they play a significant role in many metabolic processes and act as cofactors. However, exceeding their permissible intake is known to be toxic and has adverse effects on general body metabolism. One such trace element, which is ubiquitously distributed in soil, earth and water is fluoride [1]. Fluoride (F^-) in drinking water may be beneficial or detrimental depending on its concentration and total amount ingested [2]. Fluoride is beneficial especially to young children (below 8 years of age) when present within permissible limits of 0.5–1.5 mg/L for the calcinations of dental enamel and bone formation [3]. Concentrations higher than this not only affects teeth and skeleton but also cause several neurological damages in severe cases [4].

Higher concentrations of F^- in groundwater is a global problem, occurring in many continents and affecting millions of

people. According to a UNICEF report, fluorosis is endemic in at least 25 countries across the globe. In India alone, excess of F^- in drinking water is prevalent in 150 districts of 17 states of the country [5]. According to the Department of Drinking Water Supply under the Ministry of Rural Development, India, rural supply is, to a large extent dependent on groundwater (85%). Hence, it becomes necessary to reduce the F^- concentration within permissible limit of 1.5 mg/L according to Indian standards. The limit also varies among countries and the age of the people exposed [5].

Various treatment procedures have been reported for the removal of excess F^- from water. These can be broadly classified into three categories namely, precipitation, adsorption and membrane based. Precipitation involves the addition of soluble chemicals to water. Fluoride is removed either by precipitation, co-precipitation or adsorption onto the formed precipitate. Adsorption involves the passage of contaminated water through an adsorbent bed, where F^- is removed by physical, ion-exchange or surface chemical reaction with adsorbent [6]. Other defluoridation methods include membrane processes, nanofiltration, electrodialysis, etc. These processes are effective and can remove F^- to a suitable level but they are expensive and require

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frequent regeneration of beads or membrane and cleaning of the scaling and fouling [7]. Among them adsorption is still widely accepted pollution removal technique because of its ease of operation and cost-effectiveness. Recently, researchers have devoted their study on different types of low-cost but effective materials [7–10].

Portland cement, a low-cost fine-powdered building material usually consists of four main components such as tricalcium silicate, dicalcium silicate, tricalcium aluminate, and a tetracalcium aluminoferrite. Portland cement was mixed with water to get hydrated cement. The main advantage of using hydrated cement for F^- removal over other chemical treatment methods is that it does not produce sludge, abundant availability and low-cost material. So, the main objective of this study was to investigate the F^- removal potential of hydrated cement under different empirical conditions by batch adsorption studies.

2. Materials and methods

2.1. Materials

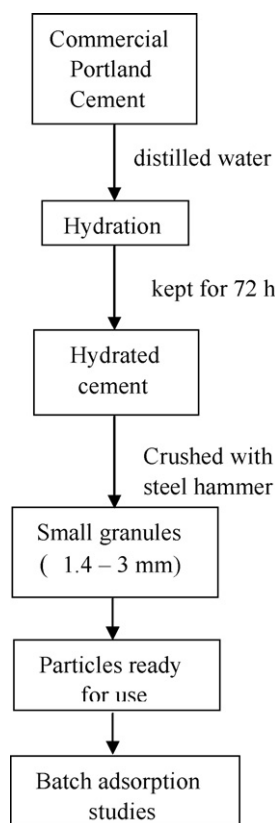
Cement used for this study was obtained from local commercial sources. All other chemicals used in the present study were of analytical grade purchased from E-Merck India Ltd., Mumbai, India. A stock solution of F^- and Ca^{2+} was prepared by dissolving known weight of sodium fluoride and calcium chloride in distilled water and desired working F^- and Ca^{2+} solution was prepared from stock solution by appropriate dilution.

2.2. Preparation of adsorbent

Preliminary studies were carried out by using cement hydrated for various time intervals of 24–120 h, respectively. Among them, cement hydrated for 72 h showed better results and it was used for further study. The detailed procedure for its preparation is as follows. About 1000 g (1 kg) of the commercially available Portland cement was taken in a vessel and the required amount of the distilled water (500 mL) was added to it. The pH of the cement in distilled water was about 9. Then, it was kept for 72 h at room temperature for hydration. After hydration/air-drying, the hydrated cement which is usually obtained in the form of clinkers was broken into small granules of ~ 1.4 –3 mm size. The entire procedure for preparation of hydrated cement (HC) was shown in Scheme 1.

2.3. Batch adsorption studies

Hundred milliliters of the desired F^- solution was taken into a 250 ml of Tarson conical flask and known weight of the adsorbent was added to it and then shaken (150 rpm) on a horizontal rotary shaker (Model No.CIS-24, Remi Instruments, Mumbai, India) for 24 h in order to attain equilibrium. The conical flasks were removed from the shaker and then allowed to stand for 2 min for settling the adsorbent. All the batch adsorption studies were conducted at room temperature ($30 \pm 2^\circ C$). Similar procedure was followed to determine the optimum conditions and to study the effect of initial concen-



Scheme 1. Schematic diagram for the preparation of hydrated cement as an adsorbent.

tration, pH, adsorbent dose, interfering ions, etc. The specific amount of F^- adsorbed was calculated from the following equation:

$$q_e = \frac{C_0 - C_e}{W} \times V \quad (1)$$

where q_e is the adsorbate loading (mg/g) in the solid (HC) at equilibrium; C_0 , C_e are the initial and equilibrium concentrations of F^- (mg/L) respectively; V the volume of the aqueous solution (L) and W is the mass (g) of adsorbent used in the experiments.

The effect of pH on F^- removal was studied by adjusting the pH of solution using 0.1N HCl and NaOH solution. In case of each sample, pH was recorded by using Orion Model 920A⁺, pH meter (Thermo Electron Corporation, Waltham, MA).

2.4. Methods of analysis

After attaining equilibrium, the experimental samples were filtered through Whatman No. 42 filter paper and filtrate was analyzed for residual F^- concentration by using fluoride ion selective electrode (Orion number 9409 on a Sargent Welch pH/activity meter model PAX 900). Heavy metals if any released from the adsorbent after equilibrium studies were estimated by inductively coupled plasma-atomic emission spectroscopy (ICP-AES, Model OPTIMA-4100DV) method. Similar procedure was used for experiments on effect of anions, initial concentration, pH, etc.

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