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ORIGINAL ARTICLE

Removal of fluoride by aluminum impregnated coconut fiber from synthetic fluoride solution and natural water

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Abstract Aluminum impregnated coconut fiber ash (AICFA) was used for removal of fluoride from synthetic fluoride solution. The AICFA showed high specific area and strong affinity toward fluoride. Synthesized AICFA was characterized by pH_{ZPC}, FTIR, SEM and XRD studies. Adsorption kinetics indicated that the adsorption equilibrium was reached within 60 min and the adsorption process followed the pseudo-second-order kinetic model better. The Langmuir isotherm model could fit the experimental data well. Thermodynamic parameters such as Gibbs free energy (ΔG°), enthalpy (ΔH°) and entropy (ΔS°) change of sorption were also evaluated which indicated that the adsorption process was spontaneous, feasible and exothermic in nature. Furthermore, the coexisting anions had significant effect on fluoride adsorption. Moreover, desorption study with AICFA showed that nearly 98% of fluoride could be leached out at pH 12. Further, the reusable properties of the material supported the possibility of its use commercially.

1. Introduction

Toxicity of hazardous ions, such as fluoride, is of interest for public health [1,2]. It is the strongest electronegative elements and in gaseous form is a very powerful oxidizing agent [3]. The natural abundance of fluoride ranges from 0.065% to 0.09% by weight in the earth's crust and it exists naturally as fluoride ions (F^-) which is extremely reactive [3]. The main source of fluoride for humans is drinking water contaminated by geological sources [4,5]. Fluoride at micro-molar level is considered as an effective anabolic agent because it promotes cell proliferations, whereas in millimolar concentration, it can bind to the functional amino acid groups located around the active center of an enzyme to cause an inhibitory effect which leads to decrease enzyme activity [6,7]. According to World Health organization (WHO), the acceptable limit of fluoride in drinking water is 1.5 mg L^{-1} [8]. In some areas of India a typical fluoride concentration in underground water is found to be in the range of 1.5–6.5 mg L^{-1} [9].

A number of drinking water defluoridation techniques have been developed. Among them reverse osmosis (RO), is one of the well-known techniques applied for removal of fluoride [10]. However, RO has serious drawback for its high operational and maintenance costs. Application of solar distillation for

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defluoridation demonstrated approximately 97% fluoride removal [11]. But the method does not produce large volume of potable water, which is its serious drawback. On the other hand, nanofiltration shows remarkable suitability for defluoridation of water. However, the use of inorganic component such as $CaCO_3$ as filter and membrane produces a smell in the water. In addition, this method requires a huge maintenance cost [12]. Application of precipitation and coagulation method for defluoridation of water has also been investigated [13,14]. However, the shortcomings of most of these methods are high operational and maintenance costs, secondary pollution (generation of toxic sludge, etc.) and complicated procedure involved in the treatment [15].

In comparison with the abovementioned techniques for defluoridation of water, adsorption is a simple, low cost and easily applicable method [16,17]. Literature has reported various adsorbents for defluoridation such as egg cell dust, sugarcane rice husk ash, tea ash, leaf biomass, algal biomass, sugarcane baggies charcoal, bio-char, etc [18–20,16,21,22].

The interest in fluoride removal by biomass based adsorbents is growing due to their low cost and availability. It is well reported that functional groups such as hydroxyl, carbonyl, amine, amide, carboxyl, sulfhydryl, imidazole, phosphonate, and phosphodiester present on the biosorbent surface contributed to biosorption [23–25]. Nowadays, it is a great challenge in the area of public health to remove fluoride from drinking water by using low-cost adsorbents at local levels, especially in fluoride affected areas. Moreover, field sample was collected from fluoride affected area of Birbhum district, West Bengal, where plenty of coconut trees are available. With this in perspective, present work was undertaken to explore the potentiality of aluminum impregnated coconut fiber ash for removal of fluoride from synthetic fluoride solution.

2. Materials and methods

2.1. Collection of adsorbents

The coconut fiber was collected from fruit shops of local market (24°35′0″N latitude and 87°5′25″E longitude). After collection it was cut into small pieces and dried in sunlight. After drying it was burned in muffle furnace at 423 K for one and half hours. The ash was washed with distilled water, dried in sunlight and then dried in oven at 353 K for overnight. It was sieved through mesh size of 150 mm and kept in plastic air tied container for further use. This material considered as coconut fiber ash (CFA). All chemical reagents were of analytical grade. Deionized water was used throughout the experiments.

2.2. Preparation of aluminum coated CFA

2.2.1. Method of coating of aluminum hydroxide on CFA

According to Ganvir and Das [26] aluminum impregnation has been done by using stirred tank reactor with stirring, vacuum/pressure filter and oven/drier (Fig. 1). Initially, 100 g CFA was taken in stirred reactor tank and 500 mL of 0.6 M aluminum sulfate solution was added and stirred the mixture. The reactor was stirred at 180–200 rpm with 1.0 M sodium hydroxide solution. In this condition, sodium hydroxide reacts with aluminum sulfate to produce aluminum hydroxide which gets deposited on CFA. In this process, the addition of sodium hydroxide is very vital and it was controlled by proper checking of pH of the mixture. Once the pH of the reaction media reaches the desired value of 5–7, sodium hydroxide addition was stopped. Finally the resulting adsorbent contains the mixture of sodium sulfate and aluminum hydroxide coated CFA. Thereafter, the whole slurry was filtered and subsequently dried at 400 K to get the desired adsorbent. However, for further use the CFA was thoroughly washed with double distilled water in order to complete removal of sodium sulfate and dried in oven at 370 K.

2.2.2. Adsorbent characterization

The scanning electron microscopy (SEM) images of the adsorbent sample are helpful to understand its surface structure. In this study images were recorded by using SEM analyzer (HITACHI, S-530, Scanning Electron Microscope and ELKO Engineering) at an accelerating voltage of 20.0 kV. Automated Mercury Porosimeters (Quantachrome, model Pore Master 60 GT) were used to determine the pore size distribution since this method is suitable for determining larger pores such as mesopores or micropores. The FTIR study was done to record the potential functional groups involved in the adsorption process by using FTIR (BRUKER, Tensor 27). The crystalline structure of the adsorbent was characterized using Ni-filtered Cu Kα radiation from a highly stabilized and automated Philips X-ray generator (XRD, PW 1830) operated at 35 kV and 25 mA at a scanning speed of 2° min^{-1} from 10° to 70°. The XRD data were matched with standard JCPDS data.

2.3. Adsorption experiment

An artificial fluoride solution was prepared by dissolving 2.21 g sodium fluoride solid granules in 1 L of deionized water and subsequently diluted to the required concentrations for the adsorption experiments. The adjustment for pH was done using HCl or NaOH. All the experiments were carried out in 250 mL conical flask, with 100 mL F^- solutions at different experimental temperature. These flasks, along with test solution and adsorbent, were shaken by the magnetic starrier, to study various parameter. The percentage of fluoride removal (% F) and the amount of F adsorbed per unit weight of adsorbent at time t (q_t, mg g^{-1}) and at equilibrium (q_e, mg g^{-1}) were calculated using the following equation, respectively:

$$\%F = \frac{C_0 - C_e}{C_0} \times 100 \quad (1)$$

$$q_t = \frac{(C_0 - C_t)v}{m} \quad (2)$$

$$q_e = \frac{(C_0 - C_e)v}{m} \quad (3)$$

where v (L) is the volume of fluoride solution, and C_0 (mg L^{-1}) is the initial concentration of F. C_t (mg L^{-1}) is the concentration of F at a given time t, C_e (mg L^{-1}) is the concentration of F at equilibrium and m(g) is the dry weight of the adsorbents. The relative parameters for the isotherm and kinetic equations were obtained using χ^2 relationship between the calculated and