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Research Note

Investigations on the Batch Performance of Fluoride Adsorption by Coconut Shell Carbon

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Abstract: Removal of fluoride using coconut shell carbon (CSC) was investigated in batch system. The fluoride removal performance of the coconut shell carbon (CSC) was evaluated as a function of the feed fluoride concentration, adsorbent dose and pH. The adsorbent was characterized as having high selectivity for fluoride and larger adsorption capacity. Adsorption was faster at the beginning followed by slower removal. pH was found to be a decisive parameter for fluoride removal. The experimental data followed the Freundlich isotherm model. Application of CSC for the effective removal of fluoride from water has been demonstrated.

Key words: Fluoride, coconut shell carbon, underground water, activated carbon, adsorption.

Introduction

High concentrations of fluoride occuring naturally in ground water have caused widespread fluorosis (both dental and skeletal) throughout many parts of the world. We purposely fluoridate a range of everyday products, notably toothpastes and drinking water because for decades we have believed that fluoride in small doses has no adverse effects on health, but it prevents dental decay. But now the benefits of fluoride are being questioned even in small amounts. More than 20 nations of the world are found endemic for fluorosis (Rukah and Alsokhny, 2004). Excess fluoride in ground water in 17 states of India also has caused havoc on human health and physiological activities (Meenakshi et al., 2004). The problem of excessive fluoride in ground water of India was first reported from the state of Andhra Pradesh (Short et al., 1937). In India, about 62 million people, including six million children, suffer from fluorosis because of consumption of water with high fluoride concentrations (Susheela, 1999). Due to its stronger electronegativity, fluoride gets attracted by positively charged calcium of teeth and bones. The major health problems caused by fluoride are dental fluorosis, teeth mottling, skeletal fluorosis and deformation of bones in children as well in adults (Susheela et al., 1993). Dental fluorosis is characterized by discoloration, blackened, mottled or chalky white teeth. Skeletal fluorosis leads to bone and joint deformities and osteosclerosis in advanced stage. According to WHO (1971), permissible limit of fluoride in drinking water is 1.0 mg/l, whereas at certain places of India, values as high as 30 mg/l have been reported (WHO, 1971; Handa, 1975). The fluoride content in the underground water is a function of many factors such as availability and solubility of fluoride minerals, velocity of flowing waters, temperature, pH, concentration of calcium and bicarbonate ions in water etc. (Chandra et al., 1981; Largent, 1961). Most of the fluoride intake in human body occurs through drinking water (Sarala and Rao, 1993). So, as excess fluoride in drinking water is harmful for human health, it is necessary to be removed from water before use.

Several methods are employed for the treatment of fluoride such as coagulation/precipitation (Saha, 1993), ion-exchange (Singh et al., 1999), electrodialysis

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(Hichour et al., 1999; Hichour et al., 2000; Adhikari et al., 1989), adsorption (Nava et al., 2003; Padmavathy et al., 2003; Raichur and Basu, 2001) and membrane process (Meenakshi et al., 2004; Aamer et al., 2001; Dieye et al., 1998). However effective method has to be evolved for removal of fluoride from underground water to avoid acute and chronic poisoning. As most of the available treatment techniques for removal of fluoride involve high investments, they are not suitable at small scale. Activated carbon adsorption is one alternative which overcomes the limitations presented by other removal methods. However, the high costs of commercially available activated carbon prompts for use of cheaper and locally available adsorbents for removal of fluoride from underground water. Application of low cost adsorbents for treating fluoride-rich water has been recommended by many researchers (Mariappan et al., 2002; Kumar and Seth, 2002). In this paper, the potential of CSC has been investigated for fluoride reduction from ground water.

Materials and Methods

Adsorbent Preparation

Coconut Shell Carbon is a low cost material and is readily available. Coconut shell carbon has unique surface properties as sorbent for the removal of fluoride from water. CSC derived from coconut is very economical compared to commercially available activated carbon. Coconut shells were procured from Malai mandir, a temple situated in Munirka about six kilometres from Indian Institute of Technology, Delhi. Cocunut shells were dried, ground, sieved and then kept in oven at 100° C for 24 hrs. The activated carbon was prepared by treating one part of coconut shells with 1.5 parts by weight of concentrated sulfuric acid (H_2SO_4) and then it was kept in an air-oven maintained at the temperature range of 140-160° C for a period of 24 hrs. The carbonized material was then washed away with distilled water to remove free acid and dried at 105-110° C. The characteristics of carbon produced are presented in Table 1.

Batch Studies

Sodium fluoride was used as a source of fluoride. Stock fluoride solution (1000 mg/L) was prepared by adding known amount of NaF in distilled water and this solution was used to prepare fluoride solutions for further experimental investigations. Batch studies were conducted to investigate the fluoride sorption rates and sorptive capacities by CSC. The reaction mixture consists of various CSC doses (2.0 to 16.0 g/L) and 100 ml of desired concentration of fluoride in the range of 2.5 mg/L to 10.0 mg/L kept in flask of 250 ml capacity. The flasks were shaken in a rotary shaker till equilibrium. At the end of desired time interval, flasks were removed from the shaker and filtered through Whatman filter paper to remove sorbent fines. The amount of fluoride removed from the solution was determined from the differences of initial and final solution concentrations. The fluoride was analysed by Ion Selective Electrode method.

Table 1: Characteristics of carbon

Bulk density	:	0.67g/cc
Moisture	:	16.26%
Ash content	:	0.33%
pH	:	6.7
Water solubility	:	1.32%
Acid solubility	:	2.25%
Decolourizing power	:	39 mg/g
Ion exchange capacity	:	1.20 meq/g
Surface area	:	13.0 m^2/g (N_2-BET method)
Iron	:	0.03%

Adsorption experiments were conducted in different batches where the adsorbent dose, treatment time, pH of the solution and initial fluoride concentration were changed. In these experiments, parametric ranges were treatment time: 15 to 300 min, adsorbent dose: 2 to 16 g/L, pH: 1-12 and initial fluoride concentration: 2.5 to 10.0 mg/L. Influence of various operating parameters was studied by varying one parameter at a time and keeping the others constant.

Results and Discussion

(a) **Effect of adsorbent dose and initial fluoride concentration on removal:** The effect of adsorbent dose at various initial fluoride concentrations on fluoride removal efficiency of CSC is shown in Figure 1. An increase in fluoride reduction has been observed with increasing adsorbent dose, because at higher adsorbent dose, large surface area and greater number of adsorption sites become available. It has also been observed that rejection of fluoride decreases at higher feed concentrations, thus the adsorbent capacity gets reduced at high fluoride concentrations.

(b) **Effect of pH:** The effect of pH on fluoride removal by CSC is shown in Figure 2. At an adsorbent dose of 10.0 g/L and ambient temperature of 30° C, the maximum fluoride removal was obtained at pH 7.0, which decreases sharply with pH change in either direction.

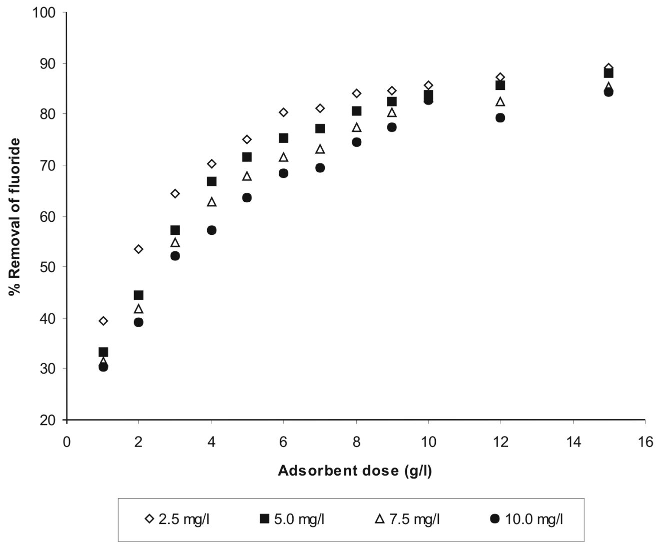

Figure 1: Effect of adsorbent dose on the adsorption of fluoride at pH 7.0.

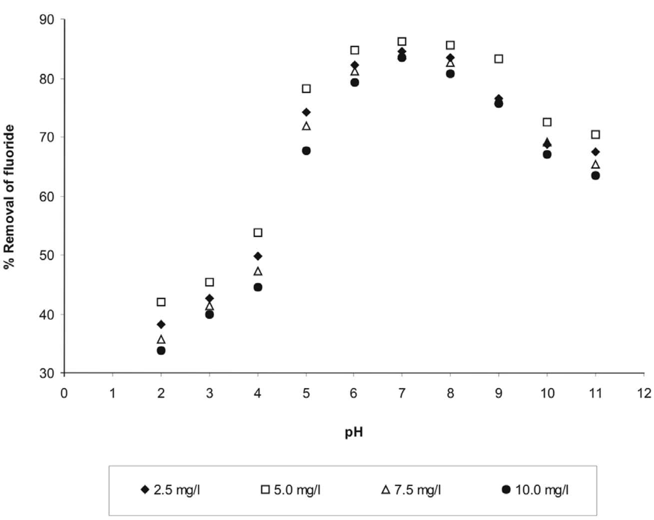

Figure 2: Effect of pH on the removal of fluoride at an adsorbent dose of 10 g/L.

(c) **Adsorption isotherms:** Both Langmuir and Freundlich equations were applied for the adsorption equilibrium, but the experimental data was found in agreement with Freundlich isotherm equation.

Freundlich equation

It can be represented as follows:

$$q_e = x/m = K.C_e^{1/n} \quad (1)$$

Equation (1) was linearized to determine the process parameters as given below:

$$\log (q_e) = \log (K) + 1/n \log (C_e) \quad (2)$$

where x is the amount of solute adsorbed (mg/L), m is the adsorbate dose (g/L), C_e is the concentration of solute in the solution at equilibrium, mg/L and K and n are Freundlich constants characteristic of the system which indicate the adsorption capacity and adsorption intensity respectively. The K and $1/n$ can be determined from the linear plot of log (q_e) vs. log (C_e). As the Freundlich equation indicates the adsorptive capacity (or loading factor on the adsorbent), $q_e(x/m)$ is a function of the equilibrium concentration of the solute. Therefore, higher capacities are obtained at higher equilibrium concentrations. The experimental and simulated results have been compared in Figures 3 and 4.

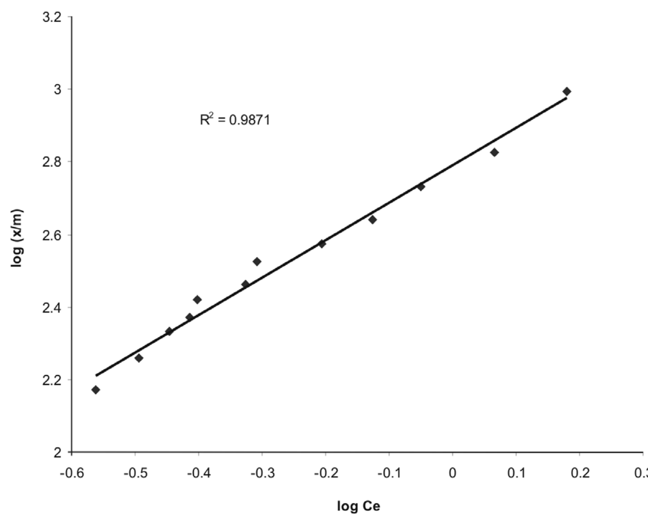

Figure 3: Freundlich plot for the adsorption of fluoride at an adsorbent dose: 10g/L, initial fluoride concentration: 2.5 mg/L and pH 7.0.

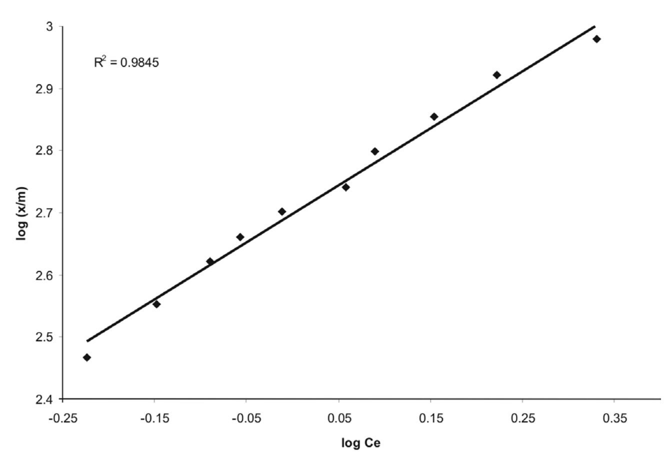

Figure 4: Freundlich plot for the adsorption of fluoride at an adsorbent dose: 10g/L, initial fluoride concentration: 5.0 mg/L and pH 7.0.

Conclusions

- CSC has high potential for fluoride removal from water and it is cost effective due to abundant availability.
- Rejection increases with time until an arbitrary equilibrium point is reached, at which maximum rejection was observed.
- pH has significant effect on rejection rate of fluoride. This behaviour is due to the strong hydrogen bondings formed by fluoride ion in acidic solution.
- Initial feed concentration and adsorbent dose are also the determining factors for adsorbents fluoride removal capacity.
- The experiment is in good agreement with Freundlich isotherm equations.

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