Defluoridation of Ground Water Using Activated Carbon of Ber (Indian Jujube) Leaves

Ashvini Sharma, Hemant Purohit, Mohd Shahnawaz Hussain, Arun Kumar Suthar, Shobha Sharma

Abstract— The present study deals with defluoridation of ground water using activated carbon of ber (Indian jujube) leaves. In this study, Activated Ber leaves carbon (ABLC) prepared by heating the leaves in electric furnace was found to be useful for the removal of fluoride. Batch experiments were applied on water sample to study the influence of pH, adsorbent dose and contact time on adsorption efficiency. Fluoride removal reached a maximum of 72.5% by particle size 0.3mm of ABLC at pH 6.0. The adsorption of fluoride from aqueous solution with ABLC followed Freundlich equation. The values of adsorption capacity (K) and intensity of adsorption (1/n) indicate greater affinity for fluoride. Thermally activated Ber leaves carbon (ABLC) was good adsorbent.

Index Terms— Defluoridation, Ground water, Adsorption, Ber leaves, Activated carbon

I. INTRODUCTION

Fluoride is a health affecting substance. The physiological effects of fluoride ingestion on human health have been studied extensively. The acceptable fluoride concentration in drinking water is generally in the range of 0.5 - 1.5 mgl⁻¹. Concentration higher than that affects the metabolism of elements such as Ca, P in human body and lead to dental and skeletal fluorosis. The fluoride content of soils varies from under 20 to several thousand ppm, the higher records being mostly from areas with bedded phosphate on fluoride deposits. Fluoride is present in the soil and rock formation in the form of fluorapatite, fluorspar, amphiboles and micas weathering rockalkali contribute fluoride natural. The fluoride present in these minerals is substituted by (OH) ion under redox conditions resulting in the release of fluoride ions to the circulating waters.

India is among 23 nations in the world, where fluoride contaminated ground water is creating health problems. Many people including children in the country are affected with dental, skeletal or non skeletal fluorosis. In rural India, ground water (which contains high fluoride) remains the dominant source of drinking water. The contents of fluoride in ground water are increasing due to heavy withdrawal of water for agriculture purpose, poor recharging, low rainfalls and pollution from industrial effluents.

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Industries which release significant quantities of fluoride in process waste to streams include fluorosilicone acid and fluoro carbon polymer synthesis, gasoline production, manufactures of coke, ceramic, cement, enamel, fiberglass, electronics, pesticides, fertilizer's, surface heating operation, metal itching (with hydrofluoric acid) and wood preservatives.

Several methods for defluoridation of drinking water have been developed. Most of these methods are based on principles of precipitation, such as the use of lime softening, alum and lime addition, activated alumina, bone char, synthetic calcium hydroxyl apatite (HA) bauxite, ion exchange resin, electrodialysis and reverse osmosis etc.

A large number of plants and their wastes have been used to remove heavy metals and other contamination from water all over the world. Recently, considerable attention has been devoted to develop better and suitable adsorbents for defluoridation purpose but adsorption process is the cheapest, simplest, easily available and accessible process for Defluoridation in developing country like India. Materials like coconut shell carbon, activated carbon, activated alumina, bone char and ion exchange resins have been used as adsorbents. Activated carbon prepared from various raw materials exhibits good capacity for removal of fluoride from drinking water.

In the present study, removal of fluoride by adsorption on to low cost material like thermally activated Ber leaves carbon was investigated.

II. MATERIALS AND METHODS

1. PREPARATION OF ACTIVATED CARBON

The Ber leaves (*Indian jujube*) were collected from the local area. They were washed with water to remove dust and other impurities. They were dried in the sun and then burnt in the thermal furnace at 400°C for half an hour. The Ber leaves were ground to obtain small pieces. They were then washed with the 1% formaldehyde solution to remove the colour and then dried in oven maintained at temperature range of 120 - 140°C for a period of 12 hr. The dried material were ground and sieved through standard sieve to obtain particle of sizes up to 0.3 mm.

2. CHARACTERIZATION OF ADSORBENTS

Various characteristics of prepared adsorbents were studied by adopting the standard procedures.

The important characteristics of activated carbon are presented in Table 1. Characteristic of carbon reveals that activated Ber leaves carbon (ABLC) was found basic. The adsorbent solubility in acid is more than water. The characteristics of carbon reveal that the ABLC possessed less

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bulk density and particle density. Fluoride removal by ABLC was carried out to assess the suitability of ABLC from aqueous solution. The effect of different parameters, viz. pH, adsorbent dose, and contact time were studied in batch experiments.

S. No.	Parameters	Activated leaves (ABLC)	Ber carbon
1.	pH	9.1	
2.	EC mmhocm ⁻¹	0.51	
3.	Bulk density gcm ⁻³	0.28	
4.	Particle density gcm ⁻³	0.3	
5.	Solubility in water (%)	6.3	
6.	Solubility in acid (%)	9.1	
7.	Moisture content (%)	8.8	
8.	Porosity (%)	13	

Table 1: Important characteristics of activated Ber leave carbon (ABLC)

3. BATCH STUDIES

Batch adsorption tests were conducted to investigate the effect of controlling parameters like pH, adsorbent dosage, size of adsorbent and contact time. All the experiments were conducted at room temperature of $27 \pm 1^{\circ}$ C. All optimization experiments were performed with 100 ml synthetic sample of standard fluoride. To study the effect of pH, adsorbent dose was 0.5 g/100 ml while time duration was kept 1 hr and pH was varied from 2 to 10. The experiment to study the effect of adsorbent dose was carried out at 1 hr contact time, pH 6.0 and ambient temperature. The effect of contact time studied at dose of adsorbent was taken as 0.5 g/100 ml, pH 6.0 and contact time was varied from 10 min to 100 min at ambient temperature.

After desired contact period, conical flasks were removed and allowed for two minutes to settle down the adsorbents. The solutions were filtered through sonar filter paper no. 1. The filtrate was then analyzed according to the standard method.

In batch process studies pH, adsorbent dose, contact time was optimized on synthetic sample of 5 ppm of fluoride. The effect of the initial concentration of fluoride was also studied. After optimization of these parameters the possible optimized conditions were applied on the ground water samples collected from Barmer to check the suitability of the adsorbent for field condition.

The percentage removal of the fluoride and the amount of fluoride adsorbed were calculated by the following equations.

% removal =
$$\frac{(C_i - C_e)}{C_i} * 100$$

Amount adsorbed (qe) =
$$\frac{(C_i - C_e)}{M} * V$$

Where Ci = initial concentration of fluoride solution in mg/L Ce = equilibrium concentration of fluoride solution in mg/L M = mass of the adsorbent in grams (gm) V = Volume of test solution in liters (L)

III. RSULTS AND DISCUSSION

1. EFFECT OF pH

Fig. 1 shows the effect of pH on fluoride removal by ABLC. It is evident that fluoride removal reached a maximum of 72.5% by particle size 0.3mm of ABLC at pH 6.0. Initially it was observed that the percent fluoride removal increase as the pH increases from 2.0 to 6.0 then decreases above 6.0. Therefore, solution pH was maintained at 6.0 for further studies.

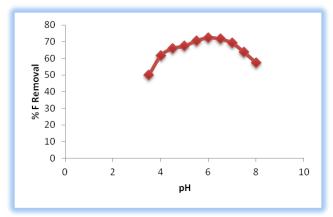


Figure 1: Effect of pH on fluoride removal by ABLC

In the acidic pH range, the amount of fluoride adsorbed slightly decreased and this can be attributed to the formation of weak hydrofluoric acid. In the alkaline pH range there was sharp drop in adsorption which may be due to the competition of the hydroxyl ions with the fluoride for adsorption.

2. EFFECT OF ADSORBENT DOSE

Fluoride uptake by carbon at different dose of 0.1~g to 1.0~g particle sizes of 0.3~mm has been given in Fig. 2. The results showed that increase in adsorbent dosage increased the amount of fluoride removal. It is observed from the results that fluoride removal increases from 50.0~% to 72.5~% at 0.1~g to 1.0~g of ABLC/ 100~ml. The significant removal efficiency was reported at 0.8~g dose of ABLC beyond which further addition of carbon revealed fluoride removal at slower rate. So for further study 0.8~g of ABLC/100~ml dose were taken.

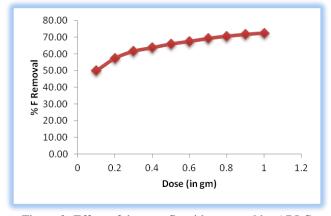


Figure 2: Effect of dose on fluoride removal by ABLC

As the particle size increases, surface area available per unit weight of the sorbent decreases. Moreover, as the particle size increases the time required for migration of the sorbate into inner pores also increases and this may result in longer time for equilibrium and a lower sorption capacity. A higher fluoride removal was observed by availability of more surface area.

It is also evident from the result that as adsorbent dose was increased, there was less commensurate increase in adsorption which may be due to lower adsorptive capacity utilization of sorbent and unit adsorption decreases significantly with increasing mass of sorbent per unit volume. This effect had been termed as "solid concentration effect" i.e. overcrowding of particles.

3. EFFECT OF CONTACT TIME

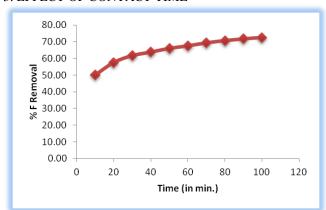


Figure 3: Effect of time on fluoride removal by ABLC

Fig. 3 exhibits the variation of fluoride removal from aqueous solution by ABLC with respect to contact time. It has been revealed from the results that fluoride removal by ABLC increases from 50.0 % to 72.5% at 10 min to 100 min was observed. There was slow removal (%) of fluoride after 60 min so this time period was taken as optimum time.

The perusal of results shows higher removal rate initially and slower rate after lapse of time. Vacant adsorption sites, high solute concentration gradient, electrostatic affinity and ion – exchange may attribute to the higher removal in the initial stages.

4. ADSORPTION ISOTHERM

Experimental data on the adsorption of fluoride on activated carbon at room temperature under optimum conditions of contact time and dosage of adsorbent were found to obey Freundlich adsorption isotherm and the model was applied to the adsorption equilibria for ABLC.

$$Log qe = log K+1/n log Ce$$

The results revealed that the adsorption of fluoride on ABLC under optimum conditions at room temperature ($27 \pm 1^{\circ}$ C) also obeyed the Freundlich adsorption isotherm. Freundlich adsorption isotherm relates fluoride adsorbed by per unit mass of the adsorbent qe with fluoride at equilibrium (Ce). The constants K and n represent the adsorption capacity and intensity of the adsorption, respectively. The data obtained in this study gave good fits to the Freundlich adsorption isotherm. The plot of Log qe against Log Ce was linear at various initial concentrations justifying the application of Freundlich adsorption isotherm (Fig. 5.4). The value of K 0.1 for ABLC indicated greater affinity for fluoride and the 1/n

values, 0.21 for ABLC showed the good adsorption character of the carbons (Table 3).

S. No.	Amount of Dose (in gm)	Ce (in mg/L)	qe (in mg/gm)	Log Ce	Log qe
1	0.1	1	1.00	0.00	0.00
2	0.2	1.7	1.15	0.23	0.06
3	0.3	2.3	1.23	0.36	0.09
4	0.4	2.9	1.28	0.46	0.11
5	0.5	3.4	1.32	0.53	0.12
6	0.6	3.9	1.35	0.59	0.13
7	0.7	4.3	1.39	0.63	0.14
8	0.8	4.7	1.41	0.67	0.15
9	0.9	5.1	1.43	0.71	0.16
10	1	5.5	1.45	0.74	0.16

Table 2: Calculation of Freundlich isotherm

Adsorbent	Value of K (intercept)	Value of 1/n (slope)
Activated Ber leaves carbon (ABLC)	0.01	0.21

Table 3: Value of K and 1/n for ABLC

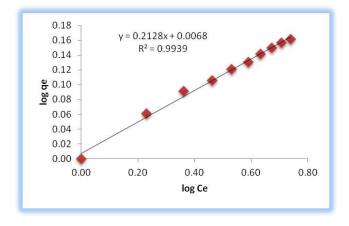


Figure 4: Plot for freundlich isotherm for the adsorption of fluoride by ABLC

IV. CONCLUSION

The present study indicates that removal of fluoride from aqueous solution depends on pH, contact time, particle size and dose of the adsorbent. The removal of fluoride is more for the finer-variety of carbon. The adsorption of fluoride from

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aqueous solution with ABLC followed Freundlich equation. The values of adsorption capacity (K) and intensity of adsorption (1/n) indicate greater affinity for fluoride. Thermally activated Ber leaves carbon (ABLC) was good adsorbent.

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