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Removal of Fluoride from Groundwater by Carbonised *Punica granatum* Carbon ("CPGC") Bio-Adsorbent

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Abstract

This study applies the development and application of low cost, *Punica granatum* bio-adsorbent for the removal of fluoride in groundwater. The batch adsorption study was carried out to analyze the defluoridation by contact time variation, adsorbent dose, adsorbate concentration, adsorbent particle size and presence of co-anions at neutral pH. The analysis of the isotherm equilibrium data using the Langmuir and Freundlich equations by linear methods showed that the data fitted better with Freundlich model ($R^2 > 0.980$). Prepared adsorbent showed enhanced removal of fluoride concentration by 78.1% at equilibrium contact time of 75 minutes. Carbonised *Punica granatum* Carbon (*CPGC*) seeds showed a high affinity for fluoride ions compared with other conventional adsorbents. Therefore, it can be considered as a potentially "good", low-cost bio-adsorbent for de-fluoridation of water compared to other bio-adsorbent.

Keywords

Fluoride Removal, Adsorption, *Punica granatum*, Bio-Adsorbent

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1. Introduction

Water is one of the most important elements for all forms of life and is indispensable to the maintenance of life on earth. Safe drinking water is the primary need of every human being. Pure water is scarce and is not easily available at all. Water may be contaminated by natural sources or industrial effluents. One such contaminant is fluoride.

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Fluoride (F^-) is a fluorine anion characterized by small radius, great tendency to behave as ligand and easiness to form a great number of different organic and inorganic compounds in soil, rocks, air, plants and animals. Some of those compounds are quite soluble in water and notable in both surface and groundwater as an almost completely dissociated fluoride ion [1] [2]. The presence of naturally occurring fluorides or added fluoridated salts in drinking water allows its easy entrance in the body via the gastrointestinal tract [3] [4]. The epidemiological studies reveal that drinking water is the major source of fluoride daily intake and continuous consumption of drinking water with elevated fluoride concentrations (>1.5 mg/l) can induce birth, reproduction and immunological defects [5] [6], dental and skeletal fluorosis [7] [8]. Besides drinking water, fluoride can also enter into the human body through food, industrial exposure, drugs, cosmetics, etc. [1]. Fluoride occurrence and concentrations in water resources (surface water and groundwater), depend on several contributing factors, such as pH, total dissolved solids, alkalinity, hardness and weathering of fluoride bearing rock forming minerals like muscovite, biotite, fluorite, fluoro-apatite etc [1] [9]-[11].

However, in many countries worldwide, elevated fluoride concentrations are a result of fluorine polluted waste water discharges [12]-[15]. According to World Health Organization norms, the upper limit of fluoride concentration in drinking water is 1.5 mg/l [16]. Excessive presence of fluoride in water for drinking continues to be a serious public health concern in many parts of the world, including India. In India, this problem is common in states such as Andhra Pradesh, Tamilnadu, Karnataka, Kerala, Rajasthan, Gujarat, Uttar Pradesh, Punjab, Orissa as well as Jammu and Kashmir [17]. Many districts of Uttar Pradesh including Unnao, Agra, Mathura, Ghaziabad, Pratapgarh and Aligarh are facing the problem of inland salinity and excessive fluoride in groundwater [18] [19].

Raebareli is one of the districts of Uttar Pradesh state in India. The land area of this district is traversed by two rivers namely Ganga and Sai. The groundwater of this district is not safe because it is contaminated with fluoride, arsenic and coliform bacteria. The fluoride level in some of the villages within the of Raebareli District, the fluoride concentration is as high as 4.2 mg/l, which implies that the villagers are at high risk of developing severe, adverse health effects [20]. Arsenic (As) is also found in different parts of "Rae Bareli" district with concentration more than WHO permissible limit (0.01 ppb). Even few blocks have arsenic contamination more than 0.05 ppb. Homemade bio-sand filter is one of the options for the removal of fluoride, arsenic and coliform bacteria and this is also economical to construct, operate and maintain [21].

Several defluoridation technologies of drinking water, such as ion exchange [22], precipitation-coagulation [23], reverse osmosis [24], electro dialysis [25] and nano filtration, have been developed for fluoride removal from water [26]. Ion exchange is a reliable and simple way to remove fluoride from water while its selective up-taking for fluoride is usually not so satisfactory for water because of the coexisting anions such as sulphate, carbonate, phosphate and hydroxide in the drinking water. These anions may impede the effective stabilization of fluoride onto the resins [27]. Precipitation-coagulation process is a good way to remove fluoride from the water [28], for its easy operation, low cost and extensive application, while one of its dominant disadvantages is its difficulty to decrease the equilibrium concentration of fluoride in water below the allowable limit level regulated by WHO, hence it can only be employed as one of the primary unit operations in the practical water treatment. Moreover, the produced mud still has the potential to cause secondary pollution and hazard. As for other methods, such as reverse osmosis [29], electrodialysis [30], and nanofiltration [31], they exhibit excellent purification capacity for fluoride removal, but expensive operation cost also limits their extensive application, especially for the wide countryside areas.

In comparison, adsorption is one of the most popular purification methods due to its high selectivity, low cost, high efficiency and easy post-treatment after adsorption. Recently, a series of low-cost adsorbents derived from agricultural waste or natural materials [32] [33] have been investigated for toxic substances like fluoride removal from aqueous solutions, such as apple and orange juicing residue, waste tea, peanut hull, rice straw, and coconut shell. Several adsorbents especially for the removal of fluoride anion from the aqueous solution have been developed, namely activated alumina, bone char, calcite, clay, zeolite, rare metal-loaded resins, etc. [34] [35] Activated alumina is quite effective for the removal of fluoride, while the leaked Al^{3+} in the drinking water may bring about senile dementia for people. Bone char is usually applied to the purification of fluoride and arsenic, but the source of this material is not so abundant. As for calcite, clay and zeolite, their cost-effectiveness still needs to be improved. In contrast to these above-mentioned adsorbents, the rare metal loaded adsorbents exhibit fairly good adsorption properties for their selective affinity to fluoride anion, high adsorption capacity, easy operation and harmless characteristic, and draw the interests of more and more researchers. For this kind of adsorbent, it is significant to choose the suitable matrix materials used for loading of rare metals and consequently affect the adsorption behaviour for fluoride anion directly.

The biomass produced in the agricultural industry is one of the most promising candidates for this application for its effectiveness, availability and abundance. In this aspect, orange juice residue has been used as the matrix material to load rare metal ions, showing quite excellent adsorption properties for anions including fluoride, arsenic and phosphate [36] [37]. While for the wide application of this technique, more biomass should be sieved to develop highly cost-effective adsorbents in fluoride purification.

In this study, a bio-adsorbent CPGC was developed for fluoride removal with a kind of easily available bio-waste, Pomegranate seeds, which can be an alternative to the costly de-fluoridation products such as ion exchange resins and electro dialysis. In India, 810 × 10^3 metric ton Pomegranate fruit is consumed every year, and lots of seed waste is disposed, causing a severe problem in the community. So, for the environmental interest, this bio-waste could be a low-cost adsorbent and remove fluoride from groundwater. The aim of the present study is to evaluate the feasibility of using pomegranate seed waste for fluoride removal from groundwater and the effect of common experimental parameters, such as the initial pH value of the solution, equilibrium concentration of fluoride, contact time, adsorbent dose and coexisting anions.

2. Experimental Methods

2.1. Adsorbent Preparation

The *Punica granatum* seed (powdered sample), common name, Pomegranate, was purchased from market. Then the material was dried at 378 - 383 K for 24 hours. It was washed with doubly distilled water to remove the free acid and dried at the same temperature for 3 hours. Later, the dried adsorbent was carbonised thermally in Muffle furnace between 1073 K to 1084 K temperature ranges. The resulting product was cooled to room temperature and sieved to the desired particle sizes, namely, <55, 55 - 106, 106 - 150, 150 - 225 and 225 - 305 micron. Finally, the product was stored in vacuum desiccators until required for treatment. The physiochemical properties of the adsorbent are listed in **Table 1**.

2.2. Batch Adsorption Study

The batch adsorption de-fluoridation study was conducted for the optimization of various experimental conditions like contact time, initial fluoride concentration, adsorbent dose, particle size and influence of co-ions with fixed dosage. The mixture was agitated in a thermostatic shaker at a speed of 250 rpm at room temperature. The reagents used in this present study are of analytical grade. A fluoride ion stock solution (100 mg/l) was prepared and other fluoride test solutions were prepared by subsequent dilution of the stock solution. Analytical grade sodium chloride, sodium nitrate, sodium sulphate and sodium phosphate were used to prepare 1.0 mol/L stock solutions, and mixed at an arbitrary volume ratio to adjust the concentration of the coexisting anions in the solution. Analytical grade sodium citrate and sodium nitrate were employed to prepare the total ion strength adjusting buffer (TISAB) solution for the pre-treatment of all sampling solutions before testing by using ion selective electrode of fluoride.

All the experiments were carried out at room temperature. Fluoride ion concentration was measured with a specific ion selective electrode by use of total ionic strength adjustment buffer II (TISAB II) solution to maintain

Table 1. The physicochemical properties of CPGC adsorbent.

Sr. No.	Parameter	Value
1.	Surface area	6.42 $m^2 \cdot g^{-1}$
2.	Density	1.18 $g \cdot cm^{-3}$
3.	Particle size	>55 µm
4.	Specific volume	0.92 $dm^3 \cdot kg^{-1}$
5.	Moisture	25.76 percent mass
6.	Ash content	28.6 percent mass

pH 5 - 5.5 and to eliminate the interference effect of complexing ions [38]. The pH of samples was measured by Orion ion selective equipment. All other water quality parameters were analyzed by using standard methods [39]. Effect of different initial fluoride concentrations viz., 2, 4, 6, 8 and 10 mg/l were studied by keeping the mass of sorbent at 0.75 g and volume of solution at 100 ml in neutral pH.

The fluoride concentration retained in the adsorbent phase, q_e (mg/g), is calculated by Equation (1).

$$q_e = \frac{(c_0 - c_e)V}{w} \tag{1}$$

where q_e is the amount of fluoride adsorbed (mg/g); V is the volume of solution; c_o and c_e are the initial and residual concentration at equilibrium (mg/l), respectively, of fluoride and W is the weight (g) of the adsorbent.

3. Results and Discussion

3.1. Effect of Contact Time and Initial Fluoride Concentration

Contact time plays a very important role in adsorption dynamics. The effect of contact time on adsorption of fluoride onto CPGC is shown in **Figure 1**. Batch adsorption studies using the concentrations 2, 3, 4, 6, 8 and 10 mg/l of fluoride solution with 0.75 g of the adsorbent were carried out at 298 K as a function of time to evaluate the de-fluoridation and adsorption rate constants. The adsorption of fluoride increases with time and gradually attains equilibrium after 75 minutes. From **Figure 1**, the time to reach equilibrium conditions appears to be independent of initial fluoride concentrations. Therefore, 75 minutes was fixed as minimum contact time for the maximum de-fluoridation of the sorbent. The adsorption of fluoride decreased from 88% to 47% by increasing fluoride concentration from 2.0 to 10.0 mg/l. Further, it was observed that the removal curves are smooth and continuous indicating the possibility of the formation of monolayer coverage of fluoride ion at the interface of adsorbent.

3.2. Effect of Particle Size

The de-fluoridation experiments were conducted using CPGC with five different particle sizes viz. 55, 55 - 106, 106 - 150, 150 - 225 and 225 - 303 μm. As the adsorption process is a surface phenomenon, the de-fluoridation efficiency of the sample with 55 μm registered high de-fluoridation efficiency due to larger surface area (**Figure 2**). The variation in the percentages of fluoride removal by the sample with different particle sizes was studied. Hence, the material with particle size of 55 μm has been chosen for further experiments. Higher percentage of adsorption by CPGC with smaller particle size is due to the availability of more specific surface area on the adsorbent surface.

3.3. Effect of Adsorbent Dose

One of the parameters that strongly affect the adsorption capacity is the concentration of the adsorbents. The effect

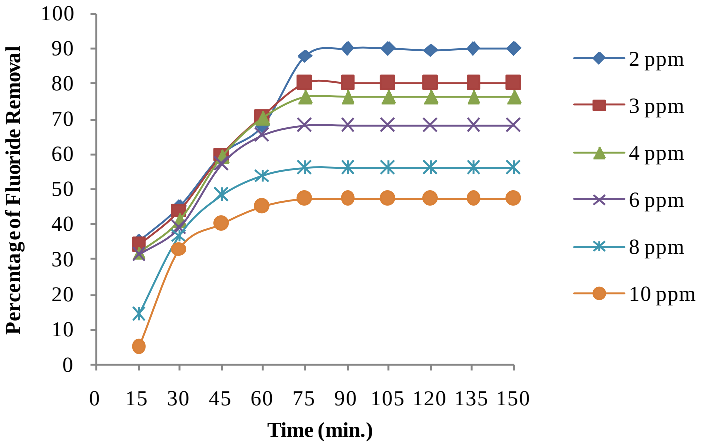

Figure 1. Effect of contact time and fluoride concentration on fluoride removal.

of adsorbent dosage on the removal of fluoride from ground water was studied at neutral pH and fluoride concentration of 3 mg/l for 150 min. **Figure 3** indicates that, increasing adsorbent dosage increases fluoride uptake, indicating that for a fluoride removal an optimum CPGC dosage of 0.75 g/L was required. The results further illustrates that the fluoride removal efficiency increases up to this optimum dosage beyond which fluoride uptake has no significant change with CPGC dosage. The increase in the adsorbent dosage provides greater surface area and hence increases the adsorption. However, by increasing the adsorbent dosage beyond the optimum dosage, there may be a slight improvement in fluoride removal efficiency but the adsorption capacity decreases.

3.4. Effect of Interfering Co-Ions

The effects of coexisting anions such as sulphate, nitrate, chloride and bicarbonate on fluoride adsorption by the CPGC adsorbent were examined and the results are given in **Figure 4**.

Chloride and nitrate did not perceptibly interfere with fluoride removal even at a concentration of 500 mg/l, while sulphate began to show some adverse effects when the SO_4^{2-} concentration increases. However, bicarbonate showed great competitive adsorption with fluoride. The fluoride adsorption amount decreased quickly from 78.1 to 52% with the increase of bicarbonate concentration 0 - 500 mg/l. This may be attributed to the competition of bicarbonate ions with the fluoride ions at the active site, on the surface of the sorbents. The selective nature of the fluoride by the sorbent depends on size, charge, polarizability, electro negativity difference, etc. The order of interference for fluoride removal was in the following order, $HCO_3^- > SO_4^{2-} > Cl^- > NO_3^-$ for the adsorbent CPGC.

3.5. Adsorption Isotherms

The sorption isotherms express the specific relation between the concentration of sorbate and its degree to

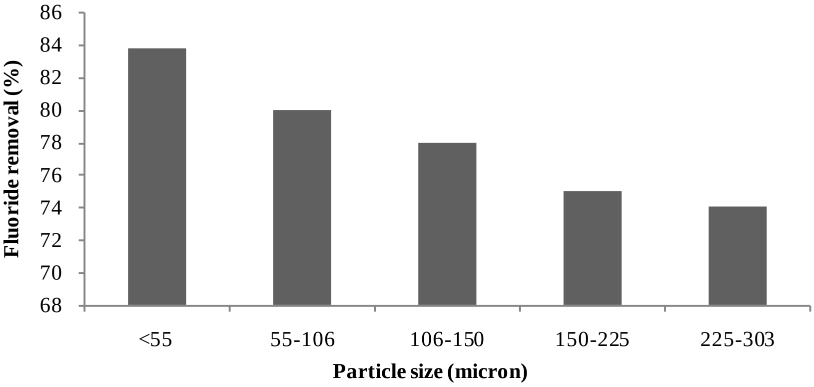

Figure 2. Effect of particle size on fluoride removal.

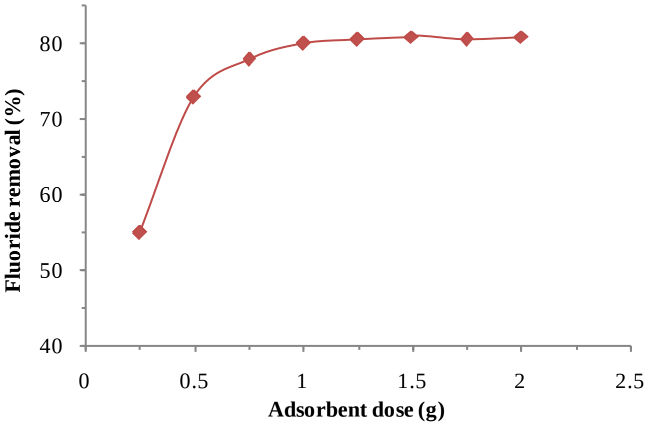

Figure 3. Influence of adsorbent dose on de-fluoridation.

accumulation onto sorbent surface. The fluoride sorption capacity of CPGC at 298 K was evaluated using Langmuir and Freundlich isotherm models.

The experimental data was applied in accordance with the linearized form of the Langmuir isotherm model. It can be represented as Equation (2):

$$q_e = \frac{q_m K_a c_e}{1 + K_a c_e} \tag{2}$$

where q_m is q_e for a complete monolayer (mg/g); K_a is the adsorption equilibrium constant (cm^3/mg). To evaluate the adsorption capacity for a particular range of adsorbate concentration, the aforementioned equation can be used in the following linear Equation (3):

$$\frac{c_e}{q_e} = \frac{1}{q_m} c_e + \frac{1}{K_a q_m} \tag{3}$$

The constants q_m and K_a in Equation (2) can be determined from the slope and intercept of the linear plot/graph of c_e/q_e versus c_e in **Figure 5**. Results indicate that the adsorbent CPGC has a high affinity for fluoride adsorption under these conditions. The present data fit well in Langmuir isotherm model with $R^2 > 0.970$. The average monolayer adsorption capacity (q_m) obtained for CPGC is 1.68 mg/g.

Freundlich adsorption isotherm is based on adsorption on heterogeneous surface, and is the earliest known relationship describing the adsorption equilibrium and is given as Equation (4):

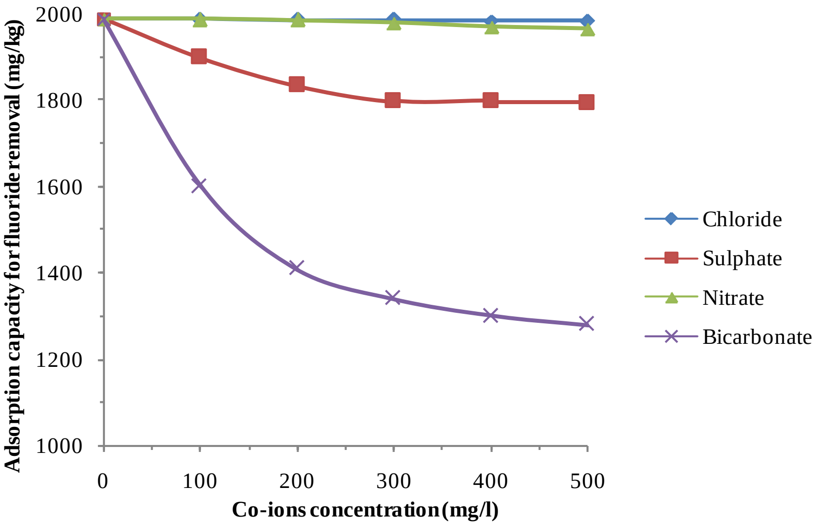

Figure 4. Variation in adsorption capacity for different concentrations of co-ions.

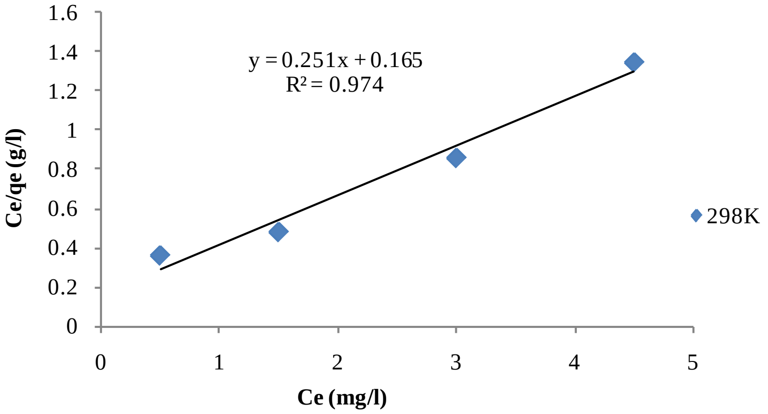

Figure 5. Langmuir isotherm for fluoride adsorption at various temperatures.

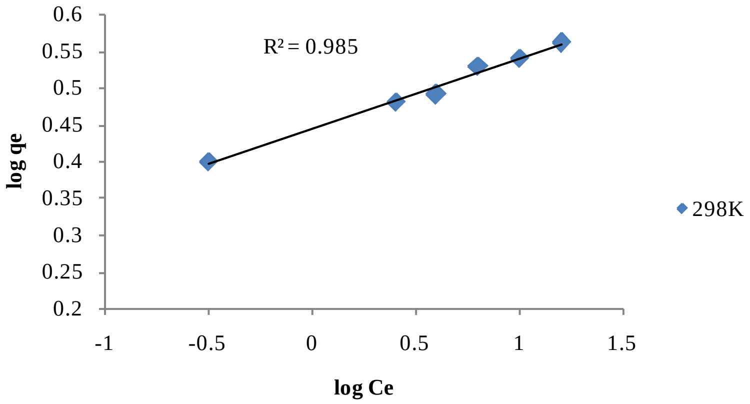

Figure 6. Plot of Freundlich isotherm for fluoride adsorption on CPGC.

$$q_e = \frac{x}{m} = K_f c_e^{\frac{1}{n}} \quad (4)$$

where x is the amount of solute adsorbed (mg), m is the mass of adsorbent used (g), K_f and $1/n$ are empirical constants, indicating the adsorption capacity and adsorption intensity, respectively. Above Equation (4) may be converted to a linear form (5) by taking logarithms:

$$\log\frac{x}{m} = \log K_f + \frac{1}{n}\log c_e \quad (5)$$

The values of K_f and $1/n$ were obtained from the slope and intercept of the plot between $\log(q_e)$ and $\log(c_e)$. The Freundlich equation deals with physicochemical adsorption on heterogeneous surfaces. Linearized form of the Freundlich equation is demonstrated in **Figure 6**. Adsorption data fitted well in linearized form of model equation ($R^2 > 0.980$) which indicated the acceptability of the model.

4. Conclusion

Among various bio-waste materials, *Punica granatum* seeds were first time utilised in de-fluoridation of groundwater. In the present study, the fluoride is removed from aqueous solutions using Carbonised *Punica granatum* Carbon (*CPGC*) seeds as bio-adsorbent. It can be concluded that CPGC has good properties for the sorption of fluoride ions from aqueous solutions. The equilibrium time for removal of fluoride concentration is determined to be 75 min. The fluoride saturation capacity of CPGC is 1.68 mg F^-/g at room temperature. The best fitting adsorption isotherm is Freundlich model ($R^2 > 0.980$), which indicates that fluoride bio-adsorption onto CPGC is characterized by chemisorptions on heterogeneous surfaces. The particle size is an important parameter that affects the sorption for fluoride on CPGC since the sorption of these ions increases as the particle size decreases. Finally, this low-cost material can be employed as an adsorbent for fluoride removal from groundwater, in particular in domestic systems where fluoride related problems exist.

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