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# Green Synthesis of Iron Nano-Impregnated Adsorbent for Fast Removal of Fluoride from Water



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#### ABSTRACT

Iron nano-impregnated adsorbent was synthesized, characterized and applied for fluoride subtraction from the water. Maximum fluoride removal (90%) was at 4.0 mg/L concentration, 25.0 min. contact time, 7.0 pH, 2.5 g/L dose and 293 K temperature. Iron nanocomposite adsorbent was selective for fluoride removal. The experimental data obeyed Langmuir, Freundlich and Temkin models. The values  $\Delta G^{\circ}$  were -1.89, -0.86 and -0.74 kJ/mol at 293, 298 and 303 K temperatures.  $\Delta H^{\circ}$  value was -7.61 kJ/mol; indicating exothermic adsorption.  $\Delta S^{\circ}$  value was  $-2.30 \times 10^{-2}$  kJ/mol K; a signal of entropy decrease during adsorption. The adsorption process was in the order of 293 > 298 > 303 K. Kinetic modeling confirmed pseudo-first-order and liquid film diffusion mechanisms. The mechanism of adsorption is also determined.

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#### 1. Introduction

Fluoride is one of the most harmful pollutants in the ground water. Water at some places in the world is not fit for drinking purpose due to fluoride contaminant. Geological and various anthropogenic activities are responsible for the ground water contamination by fluoride. Industries discharging fluoride are super phosphate, zinc smelters, ceramic works, aluminum smelters, brickworks, coal-fired power plants, steel mills, uranium enrichment facilities and oil refineries [1]. As per WHO, the permissible limit of fluoride is 1.0 mg/L [2]. High concentration of fluoride causes poor development of infant's brain, osteosclerosis, dental fluorosis, cancer and impairment in human beings [3]. Fluoride pollution is a global problem in various countries such as USA, Canada, Brazil, Pakistan, India, Sri Lanka, China, Thailand, Japan, New Zealand, and some countries of Africa and Europe continents. The fluoride problem is recognized globally, and Fig. 1 shows dental and skeletal fluorosis world wide.

In view of these facts, it is essential to develop, fast, selective, economic and eco-friendly method for the removal of fluoride from water. There are some techniques employed for fluoride removal, but adsorption is considered as the best one due to its unique features [4–18]. Literature survey indicates few papers on fluoride removal by adsorption [19–25]. These methods have certain drawbacks such as poor adsorption capacities, long contact time, high dose and extremely

\* Corresponding author. *E-mail addresses:* drimran\_ali@yahoo.com, drimran.chiral@gmail.com (I. Ali). low or high pHs. These limitations could not make these methods feasible to remove fluoride in real life problems. Therefore, the attempts were made to develop iron nano-impregnated adsorbent by green technology. The developed adsorbent was used for the removal of fluoride from water. The results of this study are discussed herein.

#### 2. Experimental

#### 2.1. Chemicals, Reagents, and Instruments

Sodium fluoride was obtained from Merck, Darmstadt, Germany. 1-Butyl-3-methylimidazolidium bromide was purchased from Sigma-Aldrich Co., USA. Deionized water was prepared using Millipore-Q, Bedford, MA, USA system. pH meter of Control Dynamics (Model APX175 E/C) was used to measure pH of the solutions. The centrifuge of Remi (model C-30BL) was used to separate the adsorbent. Powdered X-ray diffraction was carried out on Philips PX-1830 diffractometer using Cu K $\alpha$  radiation ( $\lambda = 1.54$  Å), a Cu filter on secondary optics, 25 kV voltage, 30 mA current and a proportional counter detector. The residual concentration of fluoride was determined by UV-vis. spectrophotometer (T80, P.G. Instrument Ltd., U.K.) at 570 nm wavelength as per standard procedure [26].

#### 2.2. Preparation of Iron Nano-impregnated Adsorbent

Green technology was exploited to prepare iron nanoparticles (NPs) as per the standard procedure [27,28]. The black tea (100.0 g/L) was



Fig. 1. The dental and skeletal fluorosis globally.

heated at 80 °C for 1 h. The extract was filtered followed by addition of 0.20 M ferrous sulphate solution in 1:2 ratio. The solution was kept for 24 h. The NPs formed were separated, washed with deionized water three times and drying in an oven at 250 °C for 24 h. 500 mg of 1-Butyl-3-methylimidazolidium bromide was dissolved in 100 mL acetate buffer (0.05 M, pH 4.5). 5.0 g NPs were transferred to 100 mL solutions of 1-butyl-3-methylimidazolidium bromide. It was sonicated for 24 hrs. The treated iron NPs were separated and washed with deionized water three times and dried in an oven at about 100 °C for 24 h. The prepared iron nano-impregnated particles were ready for use in adsorption experiments.

#### 2.3. Characterization of Iron nano-impregnated Adsorbent

The synthesized iron nano-impregnated particles were characterized by UV–vis. spectrometry, field emission scanning electron microscope (FESEM) and XRD techniques. The morphology of the material was ascertained by scanning electron microscope (FESEM). Images of samples were recorded at different magnifications at 10 kV operation. X-Ray diffraction (XRD) patterns of native and iron nano-impregnated particles were obtained using Philips PX-1830 diffractometer using Cu K $\alpha$  radiation ( $\lambda = 1.54$  Å), a Cu filter on secondary optics, 25 kV voltage and 30 mA current and a proportional counter detector. Iron nanoimpregnated particles were scanned from 10° to 80° 2 $\theta$  at a scanning rate of 3° 2 $\theta$  per minute.

#### 2.4. Preparation of Fluoride Solutions

The standard solution of fluoride (10.0 mg/L) was prepared in deionised water. Further dilution for UV–vis. spectrometry (0.50–5.0 mg/L) and adsorption studies (0.5-7.0 mg/L) were carried out in deionised water.

#### 2.5. Adsorption Studies

All the adsorption experiments were carried out on thermostatic water bath shaker at a fixed temperature for a given period. After adsorption, the solid and liquid parts were separated by centrifugation. Fluoride concentrations in the solution samples were determined by UV Visb. spectroscopy. Adsorption isotherms were studied in the range of 0.5-7.0 mg/L as a concentration with 1.0-10.0 pH range, 2.5-50 min. contact time, 0.5-5.0 g/L dose and 293–303 K temperatures. The different mathematical models were used to ascertain isothermal and kinetic parameters. The data obtained in batch studies was used to calculate the equilibrium fluoride uptake capacity. It was calculated using the following equation.

$$Q_e = (C_0 - C_e)/m \tag{1}$$

where,  $Q_e$  is the amount (mg/g) of fluoride adsorbed at equilibrium.  $C_o$  is initial concentration (mg/L).  $C_t$  is the equilibrium concentration (mg/L) at time 't'. *m* is the weight of adsorbent in g/L. The percentage removal of fluoride was calculated using the following equation.

$$\% \text{ Removal} = [(C_0 - C_e) / C_0] 100$$
(2)

where, C<sub>0</sub> and C<sub>t</sub> have the usual meanings.

#### 2.6. Kinetics Studies

Kinetics for fluoride adsorption was analyzed by uptake of fluoride from aqueous solutions at different times. For studying adsorption isotherms, the various solutions of fluoride were stirred with a known amount of adsorbent till equilibrium. The residual fluoride concentration was determined by UV–vis. spectrometry. The batch tests were Download English Version:

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