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FLUORIDE REMOVAL FROM WATER BY ADSORPTION ON NATURAL ADSORBENT PREPARED FROM BEET ROOT LEAVES

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ABSTRACT

Among all the quality parameters, Fluoride is one of the water quality parameters of concern. Due to its dental and bone deficiency to living entities the WHO limits the fluoride content that should not be greater than 1.5 mg l⁻¹. In the present study investigation is performed the removal ability of beet root leaves as adsorbent for removal of fluoride ion from aqueous solution. The optimum conditions were studied to get maximum adsorption. It was observed that maximum removal of fluoride ion was at pH 7.5, maximum removal was achieved upto 62%. Removal of fluoride was confirmed by EDAX. Kinetics data was best fitted for First order kinetics.

KEY WORDS: Adsorption, Fluoride, Beet root leaves, Natural adsorbent, Water treatment

INTRODUCTION

Adsorption technique is arguably one of the most versatile of all the defluoridation techniques due to a number of reasons such as cost, diverse end-uses, socio-cultural acceptance, regulatory compliance, environmental benignity and simplicity. There are a lot of materials which occurs naturally and biologically, may have unique properties to act as an adsorbent. These types of materials are available in large quantities in nature and due to these, they are of low cost with several properties such as, exchange ability, molecular sieve etc. and therefore, they can be used as adsorbent on large scale¹⁻⁶.

1. Fluoride is an element which is present in almost every water at different concentrations levels and also present in our day to day needs, viz. toothpastes, mouthwashes, cosmetics, drugs, chewing gums, etc. Although a small amount of it is useful for human health for preventing dental problems, but it is harmful when it is present in excess. According to World Health Organization (WHO) it is recommend that in drinking water the fluoride content should present in the range of 1-1.5 mg/l. Intake of excess of fluoride more than recommended quantity results in several health issues, such as dental and skeletal fluorosis. There are about 23 countries including India are in the world which are facing this chronic disease (Hong et al., 2001; Yazdi, et al., 2011) According to survey, it is estimated that nearly 62 million people of India are affected with various disease due to fluoride (Ali, et al., 2019; Adimalla, et al., 2019) Several studies are continuously going on in the field of removal of fluoride from the drinking water to attain the limit standardised by WHO. Several attempts are made using several techniques such as, adsorption, electrodialysis, reverse osmosis, coagulation/ precipitation, ion exchange, dialysis, nano-filtration, ultra-filtration, etc. are used to treat drinking water containing fluoride (Kabay et al., 2008; Sismanoglu, et al., 2004).

Tan *et al.* (2020) prepared a zirconium-based metal organic framework adsorbent via solvothermal method and studied its adsorption efficiency for removal of fluoride ions from water. They observed that at optimum conditions, the maximum removal efficiency for fluoride at equilibrium was 92.3% and maximum adsorption capacity of 19.42 mg g⁻¹. They found that the adsorption kinetic for the adsorption of fluoride is

well described by pseudo-second-order kinetics and well-fitted with the Langmuir isotherm model. Borgohain *et al.* (2020) synthesised porous MgO nanostructures with high surface areas by wet chemical method via green pathway. They observed that as-synthesised nanostructures exhibit exceptionally high adsorption capacity (nearly 90%) for the fluoride removal from water. They revealed that adsorption of fluoride is thermodynamically favourable and is exothermic in nature.

 ZrO_2 mesoporous fibers were successfully synthesized by Yu *et al.* (2018) using the electrospinning device combining with the softtemplate method. They investigated the fluoride adsorption activity of as-synthesised fibers and found that the maximum adsorption capacity was 297.70/mg/g⁻¹ with a removal efficiency of 95%. They revealed that the adsorption data were well fitted with the Freundlich isotherm model and pseudo-second-order kinetic model.

Fito *et al.* (2019) designed activated carbon from the *Catha edulis* stem to investigate the removal of fluoride from aqueous solution. They attain maximum adsorption of 18 mgg⁻¹ with a removal efficiency of 73% at the optimum condition using synthesised activated carbon. They revealed that the data founds best fit with Freundlich isotherm model (R² 0.98), which suggest that the adsorption process was multilayer. They conclude that the removal of fluoride was positivelysupported by the adsorbent dose, and pH made a negative impact on removal. In the present study investigation is performed the removal ability of beat root leaves as adsorbent for removal of fluoride ion from aqueous solution.

EXPERIMENTAL

Preparation of standard fluoride solution: Anhydrous sodium fluoride (NaF) is used to prepare stock solution for the whole experimental procedure. Stock solution with a concentration of 100 mgl⁻¹ (100 ppm) of fluoride was prepared by dissolving 221 mg of in 1000 ml distilled water. Further dilutions and aliquots were prepared using this stock solution.

Preparation of adsorbent from the Beet root leaves: In present experiment beet roots leaves collected and washed with distilled water several times then dried under sunlight for 4-5 days. These dry leaves crushed using mortal pestle, and fine powder was prepared using mixer grinder. To obtain fine powder, appropriate sieve of 60 BSS size was used. **Measurement of Fluoride:** Progress of the adsorption was measured using Fluoride Ion Meter Panomex Model PX/IMC/321. This instrument is calibrated in the range of blank or 0.0 ppm to 100 PPM. Control experiment were perform for 240 min. to optimize the rate effecting parameters, such as pH, initial concentration, amount of adsorbent, contact time, temperature, etc.

RESULTS AND DISCUSSION

(a) Effect of pH: pH is a critical parameter in case of adsorption, due to the pH value of the solution the nature of the upper surface of the adsorbent may differ or change. pH was studied in the range of 4.0 to 10.0 and result was represented graphically in Fig. 1. It is observed that higher removal of fluoride of approx 59% is at approx neutral pH value of 7.5. Adsorption increase with increase in pH upto 7.5, and after that the removal % gradually decreases.It may be due to the interference of ionic strength of the solution.



Fig. 1. Effect of pH

(b) Effect of amount of adsorbent: To study the impact of adsorbent dose, 80 ml of fluoride solution (10 ppm) was taken, and the doses of adsorbent varied in the range of 0.25 g. to 2.0 g. The obtained



Fig. 2. Effect of amount of adsorbent

results were graphically represented in the Fig. 2. It was observed that as we increase the doses of adsorbent, % of removal of fluoride increases. This happen due to the fact that, as the quantity of adsorbent increases, the surface area or the adsorption site increases, and it gives more opportunity to attach fluoride ion on the surface of the adsorbent.

(c) Effect of contact time: Adsorption is a phenomenon which depends upon time. A plot of % of removal of fluoride onto the adsorbent (Pista Shell) with respect to contact time of 0-240 min. was studied with a initial concentration on 10 ppm and different adsorbent dose of adsorbent (0.5, 1.0, 1.5 and 2.0 g.) and represented in Fig. 3. It is observed that, at all adsorbent dose similar pattern of adsorption was observed. As we increase the adsorbent dose, the % of removal increases with respect to time. After attaining a peak at 180 min., the platue like graph is observed, which indicates that the equilibrium phase is obtained. It is observed that at initial the adsorption was fast, it is explained by the fact that the at initial more adsorption site was available for the adsorbate ion.



Fig. 3. Effect of Contact time

(d) Effect of temperature: The adsorption of fluoride was also recorded at optimum conditions at pH- 7.5, concentration of 10 ppm, amount of adsorbent 0.10 g and at temperatures RT, 35, 45 and 55 °C (Fig. 4). Fig. 4 shows the removal of fluoride via adsorption on beet root leaves increases with the increase in temperature. Removal % slightly increasewith increase in temperature. This continues increase suggest that the adsorption equilibrium is endothermic in nature.

(e) Adsorption Kinetics studies: Kinetics is the study of rates of chemical processes and factors that affects the equilibrium. It also suggest about mechanism of the adsorption process, in addition to



Fig. 4. Effect of temperature

the adsorption capacity.

(i) **Pseudo-first order**: A linear form of Lagergren pseudo-first order kinetic model can be described as -

 $\log (q_e - q_t) = \log q_e - k_1 x t$

where, q_e and q_t are the amounts (mg g⁻¹), of fluoride adsorbed at equilibrium and time t, respectively; k_1 is the rate constant of pseudo-first order kinetic model (sec⁻¹).

(ii) Pseudo-second order: The linear form of pseudo-second order is expressed as-

$$t/q_t = 1/k_2 / q_e^2 + 1/q_e^2$$

where, k_2 is pseudo-second order constant. A plot of t/q_t against t gives a linear relationship. q_e and k_2 can be determined from the slope and intercept of this plot.



Fig. 5. First order kinetic plot

The best-fit model was determined on the linear regression correlation coefficient values (R^2). It is observed that the adsorption of fluoride follows First order of kinetics (R^2 =0.913).

(f) Adsorption Isotherm: Adsorption is a mass transfer process that can generally be defined as the interaction between solid and liquid phases. Both; Langmuir and Freundlich equations were studied to describe the relationship between the amount of fluoride adsorbed and its equilibrium concentration in solutions.

The equation for Langmuir isotherm is-

$$1/q_{e} = 1/q_{m} \times K_{I} \times C_{e} + 1/q_{m}$$

The Freundlich equation is based on adsorption on a heterogeneous surface and its linear form is given as:

$$\log q_e = K_f + 1/n \log C_e$$

R² values of the both isotherm suggest that, both models describe well the adsorption of fluoride molecule on Beet root leaves.

(g) Characterisation: The characterization of adsorbent prepared by beetroot leaves (untreated) and after adsorption (treated) was Characterized

using SEM (Scanning electron microscopy) and EDAX. The obtained image of SEM shows porous surface, which will be suitable for the adsorption of fluoride (Fig. 7). EDAX image of adsorbent shows the presence of fluoride in a small concentration, which conclude that fluoride is successfully adsorbed on the surface of the adsorbent.

CONCLUSION

The adsorption and removal of fluoride ion was examined on an adsorbent prepared from Beetroot leaves. It was found that maximum removal was attained at pH 7.5 with a removal of approx 62%. Equilibrium was achieved after 180 min. of contact



Fig. 6 a) Langmuir isotherm plot b) Freundlich isotherm plot



Fig. 7. SEM images of adsorbent a) untreated and b) Treated with fluoride water.



Fig. 8. EDAX images of adsorbent (treated)

time. It was also observed that temperature is also an important factor for the removal of fluoride from water as the temperature increases the adsorption slightly increases. The kinetic data best fitted with First order kinetics. This study concludes that Beetroot leaves will be a good candidate for the treatment of water containing higher fluoride content.

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Conflict of Interest

The authors declare that there is no conflict of interests regarding the publication of this article.

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