An Experimental Investigation on Removal of Fluoride Using Oryza Sativa and Azadirachta Indica Powders

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Abstract— An attempt has been made to work out the defluoridation of drinking water by using some natural adsorbents. Batch adsorption method was carried out at neutral pH by varying initial fluoride concentration, adsorbent dose and contact time. The adsorbents were prepared from Oryza Sativa (Rice husk) and leaves of Azadirachta Indica (Neem). Azadirachta Indica (Neem) and Oryza Sativa (Rice husk) exhibited highest fluorine removal efficiency about 92% and 90.8% at neutral pH, initial fluoride concentration of 5mg/l, contact time 60 min, adsorbent dosage of 25gm/250ml, stirring rate 150rpm at room temperature respectively. The obtained data were fitted to the Langmuir isotherm and Freundlich isotherm.

Key words: Defluoridation, Oryza Sativa, Azadirachta Indica, Langmuir isotherm, Freundlich isotherm, Polymath Regression

I. INTRODUCTION

Fluoride (F) is a saline component of fluorine. Fluorine is very volatile component of halogen family. 9 is its atomic number and 17 is periodic table in group. The appropriateness of seawater for inland, manufacturing and agronomic purpose depends upon the organic structure of external and subsurface of water and it is one of the prime factor. In ground water and in surface water the fresh water occurs. Ground water is the foremost and desirable basis of intake water in urban areas and particularly in rural areas, its contributes 0.6% of the overall aquatic resource on world, exceptionally in mounting father lands like India due to increase in economic growth, increase in urbanization and industrialization the ground water is getting polluted.

Small quantity of fluorine is existing in seawater, air, vegetation and wildlife. However a minor attention is helpful for the preservation for strong skeletons also false teeth, if the concentration exceeds more than 1.0mg/l causes harmful effects on human body. The fluoride range in the drinking water is 0.5 to 1.5 milligram/liter that suggested by World health organization (WHO). Fluoride enters into groundwater due to dissolution from minerals/ rocks available at aquifer bottom. Further, some quantity from industrialized methods as well as further developed practices etc.

The methods are ion-exchange method, precipitation, membrane filtration, nanofiltration, electrocoagulation, flotation and adsorption methods. Amongst them adsorption exists extensively established contamination elimination procedure since of its comfort of set-up and cost-effectiveness. In this present work, adsorbents are developed from Neem leaves, Rice husk and Activated Charcoal. In addition, they take great fiber presence creating them as appropriate for treat as bioadsorbents. Outcome of numerous constraints like adsorbent amount, primary absorption of fluoride ion along with contact time was considered.

II. MATERIALS AND METHODS

A. Adsorbent Preparation

Rice husk was partially carbonized at 250^{o}C to 300^{o}C in laboratory oven for 4 to 5 hours. The partly carbonized rise husk was completely carbonized at 500^{o}C to 600^{o}C in muffle furnace. After completely carbonization, the material from muffle furnace was cooled to room temperature. As to open the pores of the carbon the material was washed often with hot boiling water. In addition completely carbonized rice husk were treated with acid treatment. 1N HNO3 (nitric acid) in 1:1 ratio was mixed with rice husk biomass, was taken in 1000ml conical flask. Then the blend was heated at 600^{o}C to 700^{o}C in muffle furnace and treated biomass was washed until that most color was detached.

Neem leaves were washed with distilled water to get rid of wetness and soluble impurities and then it is set aside in dryer for 2 to 3 hours at 90^{o}C until the leaves color will change to pale yellow color. After that the leaves were crushed and sieved by 10-15 µm size. Then the neem leaves powder was washed with distilled water to eliminate moisture and free acid and set aside in dryer for 20-25 minutes. After drying, the powder was mixed with H_3PO_4 in silica crucible and placed in muffle furnace at 250^{o}C for 15-20 minutes. Then the solution is cooled to room temperature and washed with hot water approximately 7 times to remove free acid and moisture and kept it for dry for 20-25 minutes. After the activated black colored adsorbent is obtained it is used for additional purpose.

B. Preparation of Adsorbate solution

The fluoride stock solution of 100mg/liter was prepared by dissolving 221mg of anhydrous sodium fluoride in 1000ml distilled water.

C. Preparation of Total Ionic Strength Adjustment Buffer

This solution was prepared by dissolving 57ml of conc. Acetic acid, 58gm of sodium chloride and 12gm of sodium citrate dehydrate in 500ml distilled water. Stir well to dissolve the contents and cool it to room temperature. To adjust the pH of solution in between 5 and 5.5, 6N sodium hydroxide solutions were used and cool it room temperature. Transfer the solution to a 1lit conical flask and make up to the mark with distilled water.

D. Batch Adsorption Experiments

Batch adsorption experiments were conducted by taking the known concentration of fluoride stock solution of 250 ml and known quantity of adsorbent which is treated with acid was added to it. All the experiments were conducted at a room temperature and at neutral pH 7. Then the mixture was

agitated by using a mechanical agitator at a speed of 150 rpm for 60 min. The substances of the beakers were then allowed to settle down for 2 min and then supernatants were carefully conveyed and filtered through filter paper. The filtrate was collected and assessed for residual fluoride ion concentration by using fluoride ion-meter.

$$\%\text{Sorption} = \frac{(Ci - Ce)}{Ce} * 100$$

Where, C_i and C_e is the initial and final concentration of fluoride (mg/l) respectively.

1) Freundlich Isotherm

The linear Freundlich adsorption isotherm is given by,

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

where,

q_e is the amount of adsorbate adsorbed per unit weight of adsorbents, mg/gm

C_e is the equilibrium adsorbate concentration in solution, mg/l

K_f and 1/n are the Freundlich constants.

2) Langmuir Isotherm

The Langmuir isotherm is expressed as,

$$q_e = q_{max} b C_e / (1 + bC_e)$$

The linear form of Langmuir isotherm is:

$$C_e / q_e = C_e / q_{max} + 1 / q_{max} b$$

Where,

q_e– the amount of fluoride removed at equilibrium (mg/gm)

q_{max}– Langmuir constant related to the adsorption capacity

b – Langmuir constant related to the energy adsorption

C_e– the equilibrium concentration of fluoride (mg/ltr)

III. Results and Discussion

A. Effect of Contact time

Contact time varied from 30 min to 150 min and kept other parameters as constant i.e. adsorbent dose of 5gm/250ml, initial fluoride concentration was 5mg/l and stirring rate was150 rpm at neutral pH were adjusted for both Oryza Sativa and Azadirachta Indica. The samples were taken at regular intervals, filtered and analyzed at room temperature. Observations were represented in Fig.1. As the fig represents that the fluoride removal efficiency increases with increase in contact time. About 88% removal of fluoride was reported for Azadirachta Indica (Neem) and 83% removal for Oryza Sativa (Rice husk).

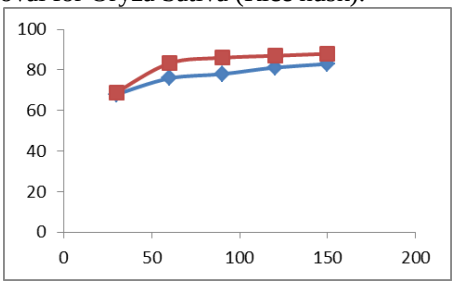

Fig. 1: Effect of contact time on % of removal

B. Effect of Adsorbent Dose

The effect of adsorbent dose varied from 5gm/250ml to 25gm/250ml and other parameters kept constants i.e. initial fluoride concentration was 5mg/l, contact time 60 min and stirring rate was150 rpm at neutral pH were adjusted for both Oryza Sativa and Azadirachta Indica. Observations were made at adsorbent dose of 5, 10, 15, 20 and 25gm/250ml and these experiments were conducted separately for both adsorbents.

The removal percentage of fluoride increases with increase in adsorbent dosage as shown in Fig.2. respectively. About 92% removal of fluoride was reported for Azadirachta Indica (Neem) and 90.8% removal for Oryza Sativa (Rice husk).

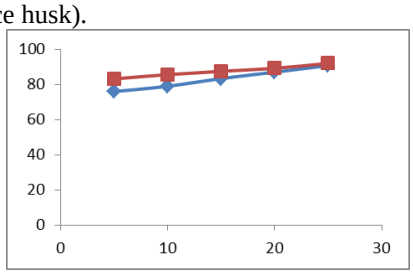

Fig. 2: Effect of Adsorbent Dose on % of removal

C. Effect of Initial Fluoride Concentration

The effect of initial fluoride concentration studies were conducted by varying it from 5mg/l to 25mg/l keeping other parameters as constants i.e. adsorbent dose was 5gm/250ml, contact time of 60 min and stirring rate of 150 rpm at neutral pH.

Fig.3. represents that the percentage removal of fluoride decreases with increase in fluoride concentration. The percentage removal of fluoride found to be 83% for Azadirachta Indica (Neem) and 76% removal for Oryza Sativa (Rice husk).

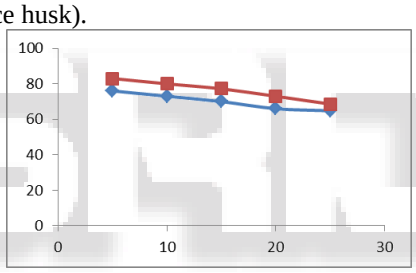

Fig. 3: Effect of initial fluoride concentration on % of removal

D. Adsorption Models

The experimental data were fitted to Langmuir and Freundlich isotherms and estimated best suitable model for this study.

1) Freundlich Isotherm

Freundlich isotherm was plotted with Log Qe vs. Log Ce as shown in above Fig.4. and Fig.5.

From the (Fig.4.) graph, the Freundlich isotherm constants of K_f and 1/n are found to be 0.688 and 0.718 respectively. Hence, the Freundlich isotherm is,

$$q_e = 0.344\ Ce^{1.393}$$

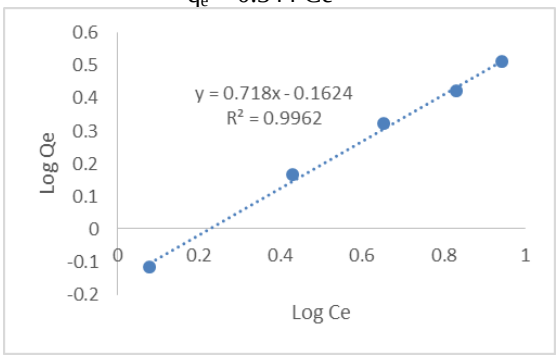

Fig. 4: Freundlich isotherm for Oryza Sativa

From the (fig.5.) graph, the Freundlich isotherm constants of K_f and 1/n are found to be 00.9761 and 0.6442 respectively. Hence, the Freundlich isotherm is,

$$q_e = 0.9761\ Ce^{0.6442}$$

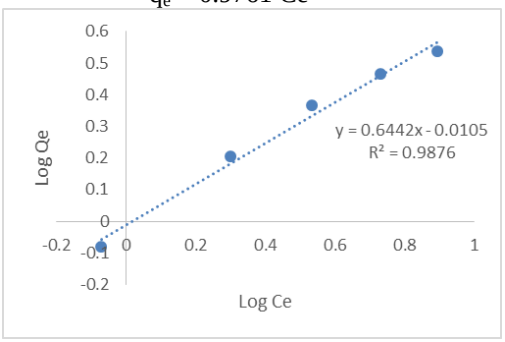

Fig. 5: Freundlich isotherm for Azadirachta Indica

2) Langmuir Isotherm

Langmuir isotherm was plotted with Ce/Qe vs. Ce as shown in above Fig.6. and Fig.7.

From the (Fig.6.) graph, the Langmuir isotherm constants of Qo and b are found to be 6.45 and 0.108 respectively. Hence, the Langmuir isotherm is,

$$q_e = 9.21\ Ce / (1 + 0.108Ce)$$

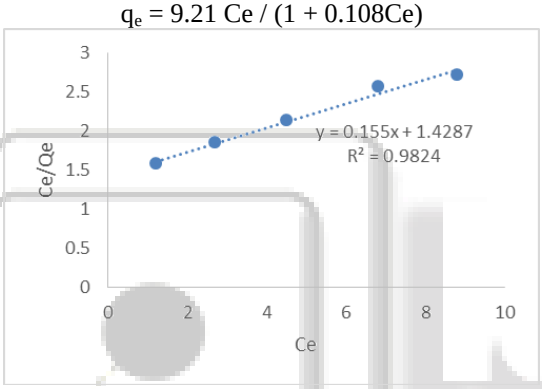

Fig. 6: Langmuir isotherm for Oryza Sativa

From the (Fig.7.) graph, the Langmuir isotherm constants of Qo and b are found to be 5.55 and 0.2058 respectively. Hence, the Langmuir isotherm is,

$$q_e = 1.142Ce / (1 + 0.2058Ce)$$

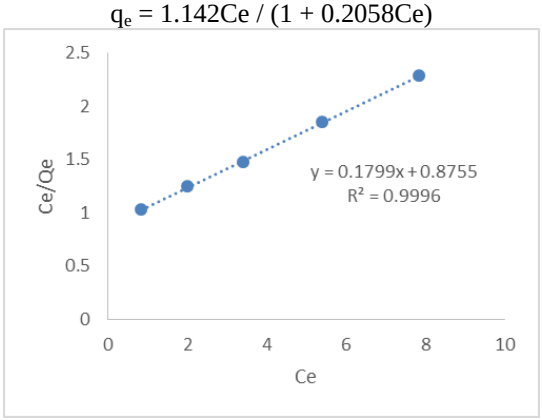

Fig. 7: Langmuir isotherm for Azadirachta Indica

IV. Conclusions

This study focused on the locally available low cost adsorbents and highlighted the significance of adsorption process with its benefits. This study concludes that the locally available adsorbents such as Azadirachta Indica (Neem) leaf powder and Oryza Sativa (Rice husk) are potential adsorbents to remove the fluoride by using batch adsorption method. The capacity of different adsorbents for reduction of fluoride depends upon the parameters like adsorbent dose, initial fluoride concentration, agitation time, pH, temperature and stirring rate. Because of neutral pH, the high fluoride adsorption was obtained in all the adsorbents. This was very favorable and useful for the drinking water treatment, especially in rural areas.

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