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Manjeet and Singh B. P

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RESEARCH ARTICLE

COMPARATIVE STUDIES OF PROSOPIS SPICIGERA, AZADIRACHTA INDICA AND ACACIA NILOTICA ON FLUORIDE REMOVAL

Manjeet[1]* and Singh B. P[2]

Chemistry Department, Deenbandhu Chhotu Ram University of Science and Technology, Murthal, Haryana, India

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ABSTRACT

A batch adsorption process was chosen on charcoal made from locally available trees viz- Prosopis Spicigera, Azadirachta Indica and Acacia Nilotica to assess their suitability for removal of fluoride from fluoride contaminated water. The efficiency of adsorption of fluoride was found to depend on physical parameters like dose of the adsorbent, pH, contact time, stirring rate, particle size of the adsorbent and initial adsorbate concentration in water. These parameters were studied to find out their optimum values for the uptake of fluoride. The fluoride removal efficiency in all the three adsorbents studied was found to be 96% (Prosopis Spicigera), 93.3% (Azadirachta Indica) and 72% (Acacia Nilotica). The optimum adsorbent doses were found to be 12 g/L for Prosopis Soicigera, 16 g/L for Azadirachta Indica and 18 g/L for Acacia Nilotica. Equilibrium was achieved in 2 hr for Prosopis Spicigera as well as for Azadirachta Indica and 90 min for Acacia Nilotica, an enhanced adsorption was obtained at pH 2. It was found that the adsorption of fluoride on these charcoals follows the Langmuir isotherm.

INTRODUCTION

Water plays an important role in man's daily activities. There are many applications of water in human life which includes drinking, bathing, livestock production, industrial, transportation, cooking, washing, irrigation, recreation and sports, hydroelectric power generation, construction, fishery and agriculture [1]. Population growth, industries and agricultural development are major reasons for constantly increasing demands for water. Hence, countries worldwide are endeavouring (striving) to improve the evaluation of their water resources. The quality, quantity and availability of drinking water are some of the most important environmental, social and political issues at international level [2]. In addition to other impurities, fluoride is found in natural water in small concentration. India is facing major problem in the form of high fluoride concentration in ground water sources [3].

High levels of fluoride present in concentrations up to 10 mg/L were associated with dental fluorosis (yellowish or brownish striations or mottling of the enamel) whereas low levels of fluoride, less than 0.1 mg/L, were associated with high levels of dental decay [4]. Excess fluoride causes pathological changes in teeth such as mottling of teeth or dental fluorosis [5]. Crippling skeletal fluorosis, which is associated with the higher levels of exposure, can cause calcification of ligaments, tendons and extreme bone deformity [6]. To mitigate high concentration of fluoride, de-fluoridation is the process of removal of fluoride ion from the drinking water. Some de-fluoridation techniques have been developed to control fluoride in water are such as reverse osmosis, precipitation, adsorption, electro- chemical, ion exchange and use of membrane. However, due to their high cost or lower efficiency or non applicability on mass scale, these techniques are not much in use [7] and [8].

This paper presents the investigation on the use of low cost and locally available materials viz, charcoal of Prosopis Spicigera, Azadirachta Indica and Acacia Nilotica. A comparative analysis of adsorption capacity of these three wood charcoals was done to find the more fluoride adsorptive behaviour of the charcoal so that these could be used as potential materials for de-fluoridation of water in future.

MATERIAL AND METHODS

Material Development

Charcoal based adsorbents were prepared from anaerobic combustion of wood of Prosopis Spicigera, Azadirachta Indica

Corresponding author:* **Manjeet
Chemistry Department, Deenbandhu Chhotu Ram University of Science and Technology, Murthal, Haryana, India

and Acacia Nilotica. Charcoal so formed from the trees was then crushed separately in to small pieces in *Hamam Dasta* and then grinded in a *Sujata* make mixer grinder. The grinded charcoal was then sieved through the sieves of various mesh sizes to obtain 212 µ, 300 µ and 425 µ sizes charcoal respectively. Charcoals of Azadirachta Indica and Acacia Nilotica were further activated by chemical treatments. Untreated charcoal of Prosopis Spicigera shows the same efficiency for removal of fluoride as chemical treated charcoal shows.

Acid Treatment

Charcoal of Azadirachta Indica was digested chemically using acid treatment. The acid treatment incurred heating the charcoal powder (40 gm) with 1 N aqueous solution of HNO_3 (400 ml) for 20 minutes followed by washing with distilled water until the entire colour and acid had been removed.

Alkali Treatment

Charcoal of Acacia Nilotica was digested chemically using alkali treatment. The alkali treatment involved heating the charcoal powder (40 gm) with 1 N aqueous solution of NaOH (400 ml) for 20 minutes, again followed by washing with distilled water until the entire colour and alkali had been removed.

Dosing of the acid and alkali treated charcoal was done on dry basis [8] and [9].

Experimental Procedure

Adsorption studies were performed by batch method to obtain the optimum value of parameters. All the steps of the experiment were performed in 250 ml conical flasks at room temperature. Standard fluoride ion solution (mg/L) was prepared from sodium fluoride (NaF) and distilled water. Experiments were carried out by adding predetermined amount of wood charcoal in 30 ml standard fluoride solution to study the extent of adsorption of fluoride on wood charcoal. The contents of all the flasks were simultaneously agitated for a particular time on Remi shaking machine. After stirring, the contents of all the flasks were allowed to settle for two min and the supernatants were carefully decanted and filtered through Wattman filter paper No. 42. The filtered solution of the flasks were analysed for residual fluoride ion concentration by using Ion-selective meter (Hanna Instruments; Model = Hi 3222-02).

RESULTS AND DISCUSSIONS

Knowledge of the optimal conditions would give a better design and modelling process for the successful application of the adsorption process. The batch adsorption process was analysed to determine the optimal conditions of pH, dose of adsorbent, contact time, stirring rate and thus concentration of fluoride ions of the uptake on adsorbent was investigated.

Effect of Dose of Adsorbent

To study the effect of an increase in the dose of adsorbent on the fluoride removal efficiency, the experiment was conducted with increasing doses of wood charcoal ranging from 2 g/L to 18 g/L. pH of all the mixtures were adjusted to 2, by adding 0.5 N HNO_3 and the initial fluoride ion concentration was fixed 2 mg/L. The contents of all the flasks containing 30 ml (2 mg/L standard fluoride) solution were simultaneously mixed at 130 rpm for 120 min in a rotary shaker. The effect of adsorbent dose on fluoride removal is shown in Fig 1.

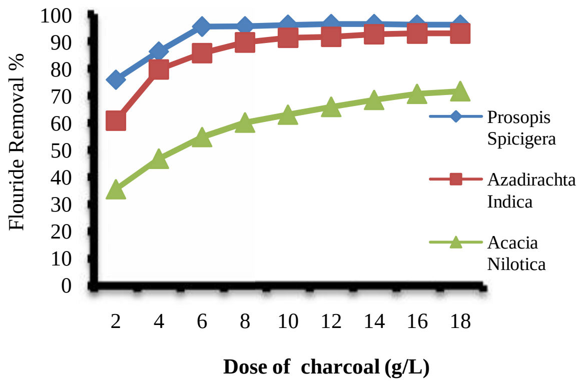

Fig 1 Effect of adsorbent dose on fluoride removal

(Initial F- concentration = 2 mg/L; contact time = 2 hr for Prosopis Spicigera and Azadirachta Indica, 90 min for Acacia Nilotica; pH = 2; stirring rate = 110 rpm for Prosopis Spicigera and Azadirachta Indica and 130 rpm for Acacia Nilotica)

It is observed that the adsorption of fluoride increases with the increase in the amount of adsorbent dose. This might be due to the fact that with higher doses of adsorbent more pore volume and larger surface area would be available to the adsorbate. Hence higher is the removal of fluoride ions. It is also observed that the removal of fluoride increases with the dose but after a certain dose range; there is no significant increase in removal [10]. The result shows that wood charcoal of Prosopis Spicigera removes 76.16% of fluoride with a dose of 2 g/L which increases to 96.8% with 12 g/L dose of adsorbent, Azadirachta Indica achieves 61% and 93.3% fluoride removal at doses of 2 g/L and 18 g/L and Acacia Nilotica achieves 35.71 % and 71.87 % fluoride removal at doses of 2 g/L and 18 g/L respectively, at room temperature of 25± 0.5^{0}C. There is less significant removal after a dose of 12 g/L for Prosopis Spicigera, 16 g/L for Azadirachta Indica and 18 g/L for Acacia Nilotica. Hence the respective doses were selected for the further studies.

Effect of pH

To study the influence of pH on removal of fluoride test mixture containing 2.0 mg/L of F^- and optimum doses were studied on various pH values i.e. 2, 4, 6 and 8. The pH of the solution containing fluoride ion was adjusted by adding 0.5 N HNO_3 and 0.1 N NaOH. As the pH of the test fluoride solution increases from 2.0 to 8.0, the fluoride removal reduces from 96% to 58% for Prosopis Spicigera, 93.1% to 42% for

Azadirachta Indica and from 71.94% to 20% for Acacia Nilotica. Graphically the results of the experiments are shown in Fig 2.

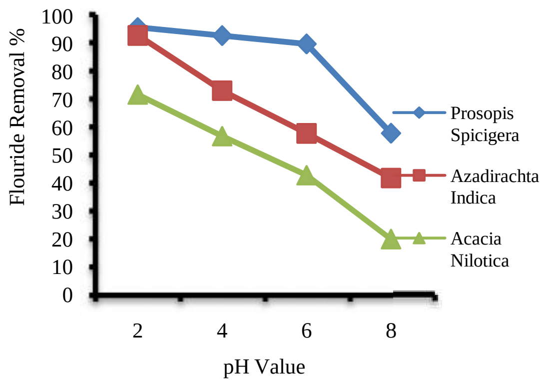

Fig 2 Effect of pH on fluoride removal

(Initial F- concentration = 2 mg/L; contact time = 2 hr for Prosopis Spicigera and Azadirachta Indica, 90 min for Acacia Nilotica; adsorbent dose = 12 g/L for Prosopis spicigera, 16 g/L for Azadirachta Indica and 18 g/L for Acacia Nilotica; stirring rate = 110 rpm for Prosopis Spicigera and Azadirachta Indica and 130 rpm for Acacia Nilotica)

Thus it concluded that removal of fluoride ions decreases with the increase in pH of the solution. The reason for the better adsorption at low pH might be due to large number of H^+ ions present at these pH values which in turn neutralizes the negatively charged OH^- ions present on the adsorbent surface thereby reducing hindrance to the diffusion of fluoride ions [8], [10], [11] and [12]. Thus pH 2 was taken for further study.

Effect of Contact Time

The study of effect of contact time was done with the standard fluoride solution of 2 mg/L and an optimum dose of adsorbent was added at optimum pH value. The contact time of these mixtures were varied from 30 min to 150 min. The result of the study is shown in Fig 3.

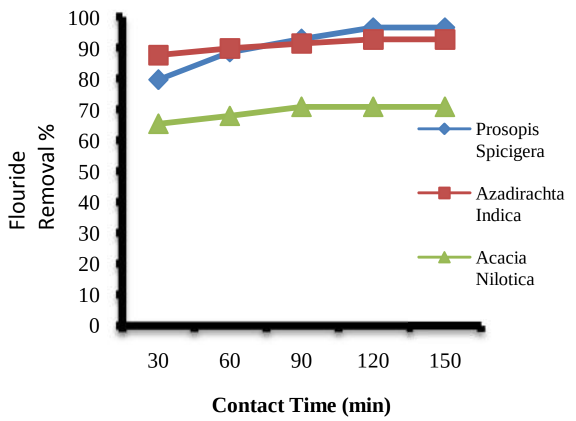

Fig 3 Effect of contact time on fluoride removal

(Initial F- concentration = 2 mg/L; pH = 2; adsorbent dose = 12 g/L for Prosopis spicigera, 16 g/L for Azadirachta Indica and 18 g/L for Acacia Nilotica; stirring rate = 110 rpm for Prosopis Spicigera and Azadirachta Indica and 130 rpm for Acacia Nilotica)

It is evident that adsorption of fluoride increases with increase in contact time and after some time it can also be seen that equilibrium was established at about 2 hr. But further increase of contact time did not increase the uptake of fluoride by the adsorbent. This might be due to non availability of the adsorption sites on the adsorbent surface [8]. The fluoride uptake increases from 80% to 97% for Prosopis Spicigera, 88% to 93.1% for Azadirachta Indica and 65.62% to 71.12% for Acacia Nilotica at 30 min to 150 min of contact time respectively. Equilibrium time was obtained at 2 hr for Prosopis Spicigera and Azadirachta Indica. Equilibrium time at 90 min was obtained for Acacia Nilotica.

Effect of Stirring Rate

The effect of stirring rate was studied by varying the agitation speed from 30 rpm to 130 rpm at optimum pH with optimum doses of all the three adsorbents with optimum contact time. The effect of stirring rate on fluoride removal is shown in Fig 4.

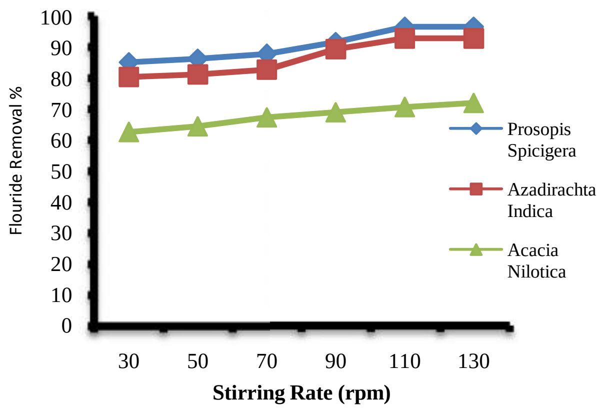

Fig 4 Effect of stirring rate on fluoride removal

(Initial F- concentration = 2 mg/L; pH = 2; adsorbent dose = 12 g/L for Prosopis spicigera, 16 g/L for Azadirachta Indica and 18 g/L for Acacia Nilotica; contact time = 2 hr for Prosopis Spicigera and Azadirachta Indica and 90 min for Acacia Nilotica)

Fluoride removal depends upon the stirring rate. An increase in adsorption was observed from 85.5% to 97% for Prosopis Spicigera, 80.7% to 93.19% for Azadirachta Indica and 62.8% to 72.3% for Acacia Nilotica when the stirring rate was increased from 30 rpm to 130 rpm. The percentage adsorption is less at lower stirring rate and increases with the stirring rate upto 110 rpm for Prosopis Spicigera and Azadirachta Indica. The adsorption increases upto 130 rpm in case of Acacia Nilotica and thereafter remains almost constant. Due to stirring, proper contact was developed between F^- ion solution and the binding sites, which promoted effective transfer of adsorbate ions on to the adsorbent sites [13].

Effect of Particle size of Adsorbent

The experiment was conducted to evaluate the influence of adsorbent particle size on the fluoride removal. The experiment was done with fluoride ion solution of 2 mg/L, pH 2, and optimum time of contact, optimum dose of adsorbents and with stirring rate. The results are shown in Fig 5.

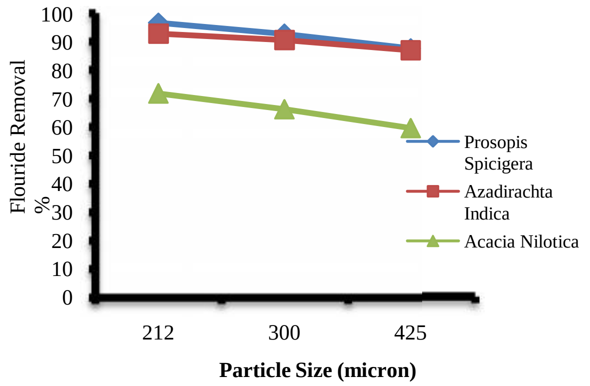

Fig 5 Effect of particle size on fluoride removal

(Initial F^- concentration = 2 mg/L; pH = 2; adsorbent dose = 12 g/L for Prosopis spicigera, 16 g/L for Azadirachta Indica and 18 g/L for Acacia Nilotica; contact time = 2 hr for Prosopis Spicigera and Azadirachta Indica and 90 min for Acacia Nilotica; stirring rate = 110 rpm for Prosopis Spicigera and Azadirachta Indica and 130 rpm for Acacia Nilotica)

The removal of fluoride increases as the particle size decreased from 425 µ to 212 µ. The fluoride removal efficiency decreases from 97% to 88% for Prosopis Spicigera, 93.18% to 87.27 % for Azadirachta Indica and 72% to 59.8% for Acacia Nilotica. The relatively higher adsorption rate as the particle size decreases due to increase in more active surface area [14].

Effect of Initial fluoride ion concentration at optimized conditions

Studies on the effect of initial fluoride ion concentration was conducted by varying the initial fluoride ion concentration of the standard fluoride solution from 2 mg/L to 12 mg/L at optimized set of conditions. The result of analysis is shown in Fig 6.

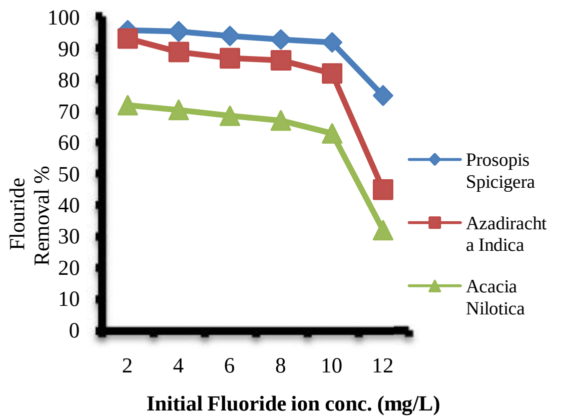

Fig 6 Effect of initial fluoride ion concentration on fluoride removal

(Initial F^- concentration = 2 mg/L; pH = 2; adsorbent dose = 12 g/L for Prosopis spicigera, 16 g/L for Azadirachta Indica and 18 g/L for Acacia Nilotica; contact time = 2 hr for Prosopis Spicigera and Azadirachta Indica and 90 min for Acacia Nilotica; stirring rate = 110 rpm for Prosopis Spicigera and Azadirachta Indica and 130 rpm for Acacia Nilotica)

The removal of fluoride ion decreases with increase in initial fluoride concentration. The adsorption capacity of charcoal was found to increase with decrease in initial fluoride ion concentration. This was probably due to the fact that for a fixed adsorbent dose, the total available adsorption sites were limited, thereby adsorbing almost the same amount of fluoride and thus, manifesting a decrease in percentage removal of fluoride corresponding to an increased initial fluoride ion concentration [13] and [14].

Adsorption Isotherms

The adsorption data of the adsorbents for the removal of fluoride ions have been correlated with Freundlich and Langmuir isotherm.

Freundlich Isotherm

Freundlich isotherm deals with physico-chemical adsorption on heterogeneous surfaces. The general form of Freundlich isotherm is given as.

$$q_e = K_f\, C_e^{1/n} \qquad (1)$$

The linearised Freundlich adsorption isotherm is given by equation:

$$\log(q_e) = \log(K_f) + 1/n \, \log C_e \qquad (2)$$

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

C_e is the equilibrium adsorbate concentration in solution, mg/L. K_f and 1/n are the Freundlich constants.

Freundlich isotherm was obtained by plotting with $\log(q_e)$ against $\log C_e$ as shown in Fig 7, 8 and 9.

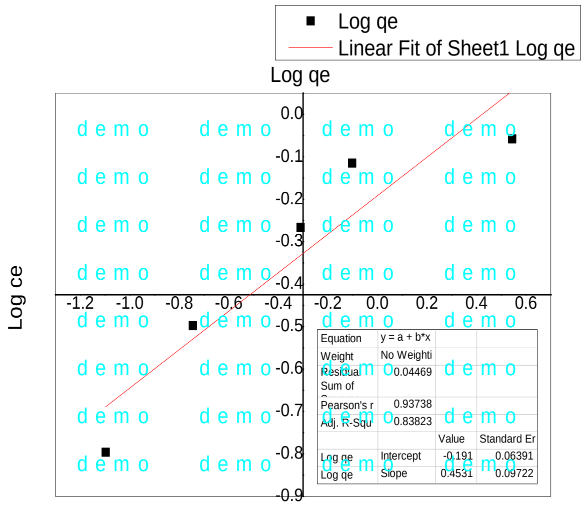

Fig 7 Freundlich isotherm for Prosopis Spicigera

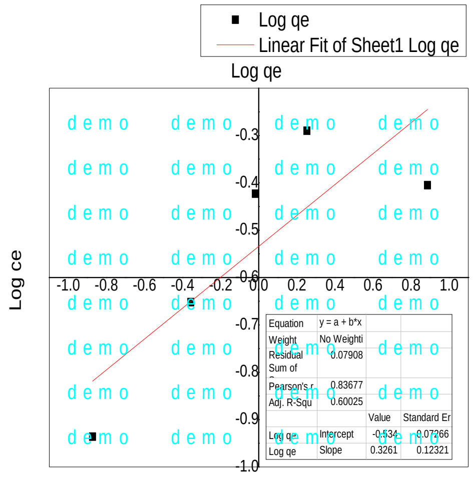

Fig 8 Freundlich isotherm for Azadirachta Indica

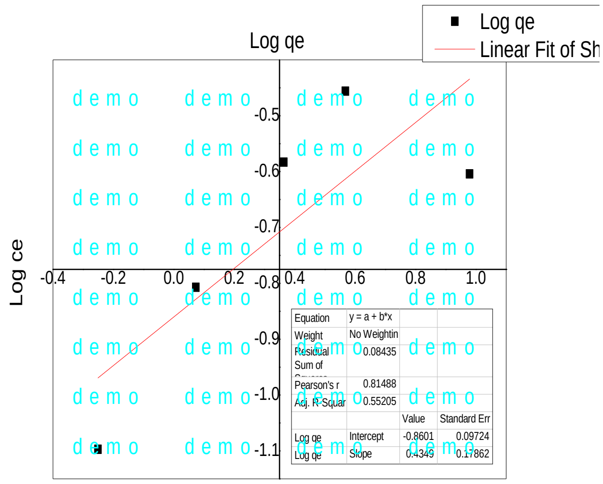

Fig 9 Freundlich isotherm for Acacia Nilotica

Langmuir Isotherm

Langmuir isotherm is valid for single layer adsorption. It is based on the assumption that all the adsorption sites have equal affinity for molecules of the adsorbate and there is no transmigration of adsorbate in the plane of the surface [9]. The Langmuir equation is commonly written as,

$q_e = abC_e/(1+bC_e)$ (3)

The linear form of Langmuir isotherm can be expressed as

$1/q_e = (1/a) + (1/abC_e)$ (4)

Where, q_e is the amount of solute adsorbent per gram of adsorbent (mg/g).

C_e is the equilibrium adsorbate concentration in solution, mg/L. a is the number of moles of solute adsorbed per unit weight of adsorbent in forming a monolayer on the surface.

b is a constant related to energy. Langmuir isotherm was plotted with $1/q_e$ against $1/C_e$ as shown in Fig 10, 11 and 12.

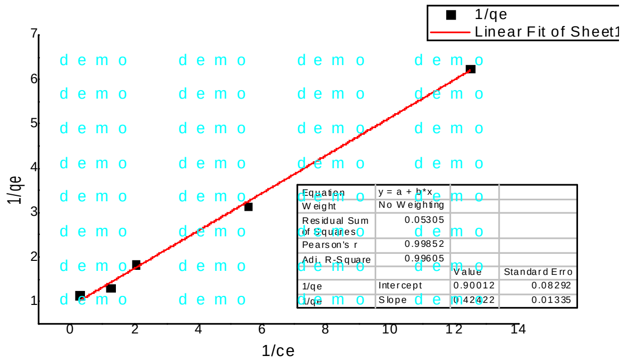

Fig 10 Langmuir isotherm for Prosopis Spicigera

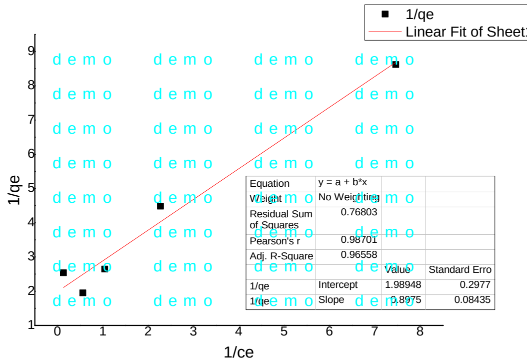

Fig 11 Langmuir isotherm for Azadirachta Indica

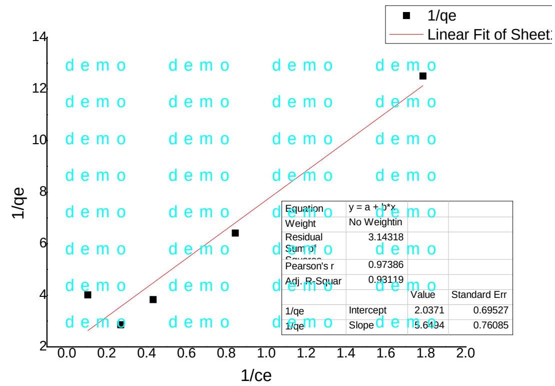

Fig 12 Langmuir isotherm for Acacia Nilotica

It could be seen that the value of R^2 for Prosopis Spicigera, Azadirachta Indica and Acacia Nilotica is 0.996, 0.965 and 0.931 i.e value for Langmuir isotherm is near to unity and hence the process of defluoridation using wood charcoal of all these three trees follows the Langmuir isotherm well. Thus it shows that the fluoride efficiency by wood charcoal of Prosopis Spicigera is more than Azadirachta Indica; which in turn more than Acacia Nilotica.

CONCLUSION

The results of the experiments show that these low cost and easily available adsorbents could be successfully used for the removal of fluoride from the water over a wide range of concentrations. The percentage of fluoride removal is found to be a function of adsorbent dose, pH, contact time, stirring rate, particle size of adsorbent and the initial adsorbate concentration in water. The process of adsorption follows Langmuir isotherm.

The exhausted charcoal can be disposed off safely after use by burning. Thus these bioadsorbents could be suitably utilized for the treatment of fluoride bearing water on domestic or community levels especially in rural areas.

References

1. Simmons I.G. (1999). Earth, Air and Water Resources and Management in the late 20th century. *Edward Arnold, London,*8 : 254.
2. Dahiya S., Singh B., Gaur S., Garg V.K. and Kushwaha H.S. (2007). Analysis of Ground water Quality using Fuzzy Synthetic Evaluation. *Journal of Hazadous meterials* 147: 938-946.
3. Agarwal V., Vaish A.K. and Vaish P. (1997). Ground water quality: Focus on Fluoride and Flourosis in Rajasthan. *Current Sci.* 73: 743-746.
4. Edmunds W.M. and Smedley P.L. (1996). Groundwater Geochemistry and Health: an Overview. *Appleton, Fuge and McCall [Eds] Environmental Geochemistry and Health, Geological Society Special Publication*, 113: 91-105.
5. Mahavi A.H. *et al*. (2006). Survey of Fluoride Concentration in Drinking Water Sources and Prevalence of DMFT in the 12 years old students in Behshar city. *J. Med. Sci.* 6: 658-661.
6. IPCS (2002). Fluorides. *Environmental Health Criteria 227, International Programme on Chemical Safety, World Health Organization, Geneva.*
7. Tembhurkar A.R., Dongre S.R. (2009). Comparative Studies on Fluoride Removal using Natural Adsorbent viz Azadirachta Indica (Neem) and Ficus Religiosa (Pipal), *IE Jornal- EN*, (1): 18-23.
8. Jamode A.V., Sapkal V.S. and Jamode V.S. (2004). Defluoridation of water using Inexpensive Adsorbents. *J. of Indian Inst. Sci.,* 84: 163-171.
9. Tembhurkar A.R. and Dongre S.R. (2009). Comparative Studies on Fluoride Removal using Natural Adsorbents viz Azadirachta Indica (Neem) and Ficus Religiosa (Pipal), *IE(1) Journal-EN.*
10. Srimurli M. (1998). AStudy on Removal of Fluorides from Drinking water by Adsorption onto Low Cost Materials. *Journal of Environmental Pollution, (99):* 285.
11. Bhargava D. S. and Killedar D. J. (1993). Effect of Stirring rate and Temperature on Fluoride Removal by Fishbone Charcoal. *Indian Journal of Environmental Health, (35)*: 81.
12. Bulusu K. R. And Nawlakhe W. G. (1998). Defluoridation of water with Activated Alumina; Batch Operations. *Indian Journal of Environmental Health, (30)*: 262.
13. *Hem J.D.* (*1989).* Study and Interpretation of the Chemical Characteristics of Natural water. *US Geological survey, Washington, D.C., Water Supply Paper* 3: 263.
14. USEPA (1996). R.E.D. Facts. *Cryolite, EPA-378-F-96-016, United States Environmental Protection Agency*.

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