Research Article

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Use of aralu (*Ailanthus exelsa*) stem charcoal for defluoridation of drinking water

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Abstract: Excessive intake of fluoride (F), mainly through drinking water, is a serious health hazard. Endemic fluorosis is found in at least 20 states of India, affecting more than 65 million of mainly under privileged people including 6 million children. Fluoride ion concentrations in India's groundwater vary widely, ranging from 0.01 mg/L to 48 mg/L. Defluoridation of ground water and supply of safe drinking water is the only immediate solution to this problem. There are several methods used for the defluoridation of drinking water, of which adsorption processes are generally found attractive because of their effectiveness, ease of operation, simplicity of design, and for economic and environmental reasons. The present investigation is an attempt towards a cost effective feasible solution for defluoridation of drinking water by using Ailanthus exelsa as absorbent. The adsorption capacity of Ailanthus exelsa stem charcoal has been evaluated. Adsorption studies for defluoridation on Ailanthus exelsa charcoal powder showed that the adsorbent were highly influenced by temperature, pH of water and initial fluoride concentration.

Keywords: Biosorption, Ailanthus exelsa, pH, Adsorption.

INTRODUCTION

Management of contaminants such as fluoride is a major public issue. Fluoride enrichment in natural water can occur either by geological processes or from industries[1]. Fluoride of geogenic origin in groundwater used as a source of drinking water is a major concern because fluoride content above permissible levels is responsible for human dental and skeletal fluorosis. Consequently, water sources containing elevated levels of fluoride needs to be treated. The fluoride load in the aquatic environment due to industrial discharge is at least 100 fold higher compared to that which arises due to leaching of fluoride bearing minerals [2]. Fluoride, as a dissolved constituent of drinking water, is perhaps the only substance producing divergent health problems. Fluoride concentration in the range of 0.8 to 1.20 mg/L is considered to be beneficial, concentration higher than 1.5 mg/L are reported to be harmful to the teeth and bone of men and animals. The toxicity of fluoride is influenced by high ambient temperature, alkalinity, calcium and magnesium contents in the drinking water. The drinking water standard for fluoride set by WHO is 1 mg/L [3]. In India, endemic fluorosis affects more than 65 million populations and is a major problem in 20 states of India. The most affected states in India are Rajasthan, Andhra Pradesh, Orissa, Gujarat, Madhya Pradesh and Chhattisgarh states [4]. Environmental factors include annual mean temperature, humidity, rainfall, tropical climate, duration of exposure etc. Besides, other factors such as pH in terms of alkalinity, age, calcium in diet, fresh fruits and vitamin-C reduces fluoride toxicity. Whereas, trace elements like molybdenum enhances the fluoride toxicity. Defluoridation is normally accomplished by precipitation (coagulation) and adsorptive processes. Fluoride removal by chemical precipitation using alum, iron, lime and magnesium compounds, and calcium phosphate was investigated by several researchers [5, 6, 7, 8, 9, and 10]. Adsorption process has been reported to be effective, environmental friendly and economic [11]. Use of biosorbents/biomass from various microbial sources, leaf based sorbents, and water hyacinth, for fluoride removal was reported by various investigators [12,13,14]. Used fungal biosorbent for removing of fluoride from water has also been studied [15]. Aspergillus Niger [16], A. fumigates [17], *A. flavus* [18] and Fusarium moniliforme [19] have also been investigated for defluoridation of water. Defluoridation of drinking water is the only pragmatic approach to solve the fluoride pollution problem as the use of alternate water sources and improvement of nutritional status of population at risk have their own limitations and are expensive affairs[20]. Generally, methods reported in literature are based on adsorption, ion exchange, precipitation. All these methods, their principle of operation, advantages, disadvantages, limitations and applications have been critically reviewed [21,22]. Adsorption techniques are advantageous for defluoridation as the processes are capable of removing fluoride up to 90% and are cost effective [23,24]. Therefore, it is aimed to find a suitable highly cost effective and environmental friendly

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method for removal of fluoride in the drinking water that could be used by under privileged community. Fluoride removals by using synthetic adsorbents are common. Reports on the fluoride removal using algal biomass and tree leaves [25] are available in recent literature.

In the present investigation a cost effective feasible solution for defluoridation of drinking water by using *Ailanthus exelsa* (Aralu) as absorbent has been attempted.

EXPERIMENTAL

Preparation of Aralu Stem charcoal Adsorbent

In the present study, Aralu (*Ailanthus exelsa)* have been used as adsorbing media. Aralu plants (Fig.1) are easily available in this region. Activated Charcoal (Fig.2) was prepared from dry stems or timber of Aralu tree. Charcoal was prepared by charring the dried plant materials at about 100-200°C for about 3-4h in a muffle furnace. By this time all of the material was completely carbonized. The carbonized material was cooled and then grounded in a laboratory blender and made into fine powder of mesh size (230-400). The fine powder of *Ailanthus exelsa* charcoal is then sorted in desiccators and is used for biosorption studies. A fluoride stock solution of 210 mg/L was prepared by dissolving 0.21 g of AR Grade Sodium Flouride (Merck) in 1L of double distilled water at room temperature. The solution was diluted as required to obtain working solution. The initial pH of the working solution was adjusted to 5.0 by addition of 0.1N HNO_3 or 0.1N NaOH solution except experiment examining the effect of pH. Fresh dilutions were used for each study [26,27].

Fig.1: Aralu Stem and Leaves

Fig.2: Aralu (*Ailanthus exelsa) Charcoal*

Adsorption Experiments

Batch adsorption experiments were conducted to examine time-dependent absorption behavior and the effects of temperature and solution pH on adsorption performance. Following a systematic process, the adsorption uptake capacity of fluoride in batch system was studied in the present study. The data obtained in batch mode studies was used to calculate the equilibrium fluoride adsorptive quantity by using the following expression:

$$qe = \frac{(Co - Ce)\,V}{m}$$

where qe is the amount of adsorbed fluoride onto per unit weight of the biomass in mg/g, V is the volume of solution treated in litre, Co is the initial concentration of fluoride ion in mg/L, Ce is the residual fluoride ion concentration in mg/L and m is the mass of adsorbent in g/L. Ten Adsorption studies were carried out at different conditions viz. contact time (30-180 min), pH (2-10), initial fluoride concentration (2-21 mg/L) and temperature (25, 30, 40 and 45°C). About 0.5 g of the adsorbent was mixed with 100 mL of a desired fluoride solution with known pH and agitated in a mechanical shaker at 100 rpm for certain time. The solutions were filtered and collected for the determination of fluoride ion concentration Each determination is repeated thrice and the results given are the average values.

***Effect of initial ion concentration** (for isotherm study):* Equilibrium experiments were carried out by stirring 0.5 g of *Ailanthus exelsa* Charcoal with 100 ml of

F ion solution of different initial concentrations (20-210mg/L) at pH 5.0 and at room temperature.

Effect of biosorbent concentration

Batch adsorption tests were done at different concentration of adsorbent from 0.1g to 0.6g at pH 5.0, for a contact time of 20 minutes at room temperature (25°C ± 2°C). The samples were then agitated and filtered and the filtrates were analyzed as mentioned before.

Effect of solution pH on biosorption

The effect of pH on the adsorption capacity of *Ailanthus exelsa* was investigated using 100 ml solution of 210mg/L of fluoride ion for a pH range of 2.0 to 10.0 at 30°C. Flasks were agitated on a shaker for 10 minutes to ensure that the equilibrium was reached. Then the mixture was filtered and the concentration of fluoride in the filtrates was measured.

RESULT AND DISCUSSION

Effect of contact time: The effect of contact time on Fluoride biosorption on Aralu (*Ailanthus exelsa*) was studied and the results were shown in Figure.3.

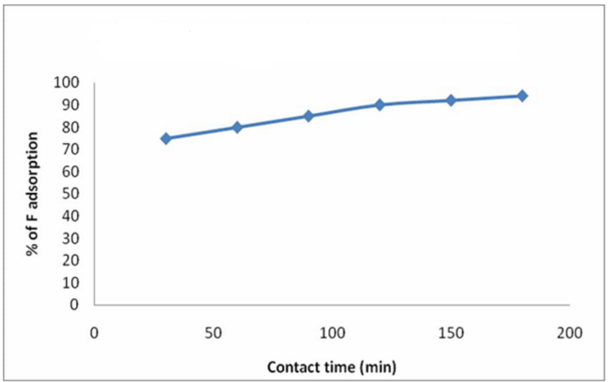

Fig.3. Effect of contact time on the percentage removal of fluoride

From Fig. 3, it was found that the adsorption quantity of Fluoride ion on *Ailanthus exelsa* increases as the contact time increased. The biosorption of fluoride onto *Ailanthus exelsa* was rapid for first 30 minutes (75%) and equilibrium was nearly reached after 180 minutes (94%). Hence, in the present study, 180 minutes was chosen as the equilibrium time. Basically the removal rate of sorbate is rapid, but it gradually decreases with time until it reaches equilibrium. The rate of percent fluoride removal is higher in the beginning due to the larger surface area of the adsorbent being available for the adsorption of the fluoride ion. It is also relevant that, since active sorption sites in a system have a fixed number and each active sites can absorb only one ion in a monolayer, the fluoride uptake by the sorbent surface will be rapid initially, slowing down as the competition for decreasing availability of active sites intensifies by the fluoride ions remaining in the solution.

Effect of initial concentration of ions

Biosorption experiments with Aralu (*Ailanthus exelsa*) were conducted for solutions containing 2 mg/L to 10mg/L fluoride ion. As seen in the Figure 4, at lower concentrations of fluoride ion (2mg/L-6mg/L), biosorption was completed rapidly to about 94-80% success rate, but at higher concentrations biosorption was achieved slowly with a success rate of 60-75%.

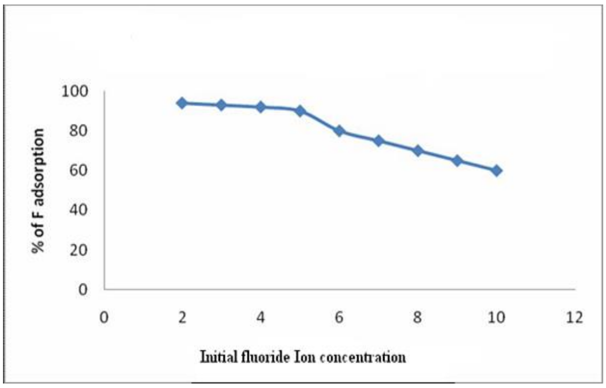

Fig. 4: Effect of initial concentration of fluoride ion on its removal of from solution.

At lower concentrations, all fluoride ions present in the solution would interact with binding sites and then facilitated more than 80% adsorption. At higher concentration, more fluoride ions are left unabsorbed in the solution due to the saturation of binding sites. This appears due to the increase in the number of ions competing for available binding sites in the biomass.

Effect of biosorbent concentration

The percentage removal of fluoride ion increases with increase in *Ailanthus exelsa* doses from 0.1g to 0.5g. There was a non-significant increase in removal of percentages of fluoride when adsorbent dose increases beyond 0.5g. This suggests that after a certain dose of bio adsorbent, the maximum adsorption is attained and hence the amount of ions remain constant even with further addition of dose of adsorbent. The increase in fluoride removal percentage with increase in adsorbent dose is due to the greater availability of the exchangeable sites or surface area at higher concentration of the adsorbent.

Effect of solution pH

The pH of the solution is perhaps the most important parameter for adsorption. To understand the adsorption mechanism, the adsorption of fluoride as a function of pH was measured and the result are shown in Figure 5.

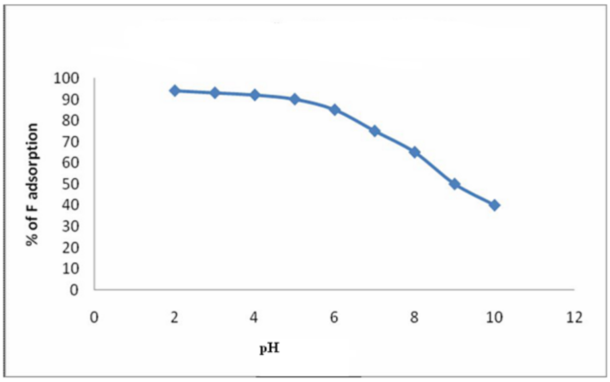

Fig. 5 Effect of pH on the removal of fluoride from solution.

Fluoride removal recorded its minimum values at pH 10.0. There was an increase in biosorption capacity of the biomass with decrease in pH from 10.0 to 5.0 and showed marginal increasing trend from pH 4.0 lower pH. In lower pH values, occupation of the negative sites of the adsorbent by H^+ and H_3O^+ ion leads to reduction of vacancies for fluoride ion and consequently causes decrease in fluoride ion biosorption. As the pH was raised, the ability of the ions for competition with H^+ and H_3O^+ ions was also increased. Although the sorption of fluoride ions raised by growing pH, further increment of pH caused declining in adsorption due to the competition of the hydroxyl ions with the fluoride for adsorption.

CONCLUSION

The Aralu (*Ailanthus exelsa*) stem charcoal is found to be an efficient adsorbent for the defluoridation of drinking water. The biosorbent was successful in removal of fluoride ions from aqueous solution of 10mg/l fluoride concentration with about 94% efficiency. Biosorption equilibrium was achieved within 180 minutes. It was observed that the adsorption was pH dependent with maximum adsorption achieved at pH 5.0.

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