



CHARACTERIZATION AND DEFLUORIDATION STUDIES OF ACTIVATED *DOLICHOS LAB LAB* CARBON

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ABSTRACT

The adsorption studies on the removal of fluoride from potable water using activated Dolichos Lablab Carbon (NDLC) was carried out under various experimental conditions. The maximum adsorption was observed to be a function of the solution contact time, initial concentration of fluoride and adsorbent dose. The effect of various process parameters have been investigated by following the batch adsorption technique at $30 \pm 1^\circ\text{C}$. The adsorbent capacity of adsorbent for fluoride was obtained by using linear Langmuir and Freundlich isotherms.

Keywords: dolichos lablab, Freundlich, Langmuir isotherms

INTRODUCTION

The development of indigenous low-cost materials for the water and waste water treatment is essentially needed, because the available commercial activated carbon (CAC) is costly and it is difficult to procure in developing countries like India. The ground water containing high levels of fluoride is used for drinking purpose, this usage of fluoride contaminated water over a period of time causes health problems, such as fluorosis i.e., deformations of skeletal, non skeletal, dental and also other hazardous effects. It also depends on the nutritional status and food habits of the people and geographical features of the region. The high fluoride content in water to be identified and removed to produce safe drinking water, in order to discourage the people from consuming water from this fluoride-contaminated areas. In recent years, various carbons from different species have been prepared and were used as adsorbents for defluoridation studies as well as for other purposes. The raw materials used for the preparation of carbon is available in plenty. Several attempts have been made to prepare carbons from coconut shell¹, sulphonated coconut shell², tea waste and ground nut shell, coffee husk³, avarambrak⁴, rice husk⁵, bone⁶ etc were used for defluoridation purposes. These studies have indicated that these adsorbents were less effective in removing fluoride content in water and showed poor additional characteristics. So there is a need to develop cost effective and efficient fluoride adsorbents for small and medium scale level operations. Even we have to keep an view of their maintenance expenditure. Adsorbent ⁷ *dolichos lablab* carbon (NDLC) was studied and this adsorbent was characterized after activation with HNO_3 . We are here with reporting the studies with improved adsorption capacity in removal of fluoride from water to be used for the drinking purposes.

Gampalagudum is one of the Mandals in Krishna District of Andhra Pradesh. High levels of fluoride in ground water was reported. It is proved that the consumption of drinking water containing high fluoride content causes fluorosis. High incidence of fluoride toxicity responsible for the various factors like skeletal, non-skeletal and dental deformations. The high fluoride content water samples were collected and checked the fluoride levels to discourage the people from consuming this contaminated water with out purification.

EXPERIMENTAL

Preparation of carbon from *dolichos lablab* (NDLC)

Activated carbon in powder form is prepared by the pyrolysis of arial parts of *dolichos lablab* (*Fabaceae*). They are collected, washed, dried, and crushed before carbonizing in uniform nitrogen flow in a horizontal tube furnace electrically heated at 600 °C for 4h, followed by activation. Then the activated carbon was cooled to room temperature and washed with deionized water until the effluent was clear in color. Finally, the activated carbon was dried in oven at 110 °C for 12 h. The activated carbon is chemically activated with 0.1M solution HNO₃AC under similar conditions to determine the effect of surface modification. The surface modification of carbons was also done by subjecting to liquid phase oxidation. The powdered activated carbon obtained after HNO₃ treatment has a particle size in the range of 40-50 mesh. The important characteristics of the above carbon, such as adsorption capacities, effect of contact time, effect of adsorbent dose, removal of fluoride in bulk water samples, recycling of fluoride loaded adsorbent, effect of pH, regeneration of the fluoride loaded adsorbent levels are discussed. The fluoride content of these samples were determined using SPADNS method⁸. A door-to-door survey was conducted in the study area to know the prevalence of skeletal, non-skeletal and dental fluorosis. The drinking water samples from all panchayat areas were collected in pre cleaned high-density polyethylene bottles and were tested in the laboratory. The fluoride content of these samples were determined using SPADNS method. The various water quality¹⁰, parameters¹¹ were selected and analyzed¹² by following standard methods¹⁴.

RESULTS AND DISCUSSIONS

Defluoridation of potable water

The fluoride content in the water samples were analyzed and given in Table 1. The natural fluoride content in drinking water depends on the source, climatic and geological factors. The concentration is usually below 1ppm in unpolluted surface water. The safe limit of fluoride for drinking water supply is 1.5 ppm¹⁵. The concentration of fluoride in all the samples in the study area has varied from 1.6 to 4.0 ppm except in the case of samples 18, 20.

Effect of contact time

In the adsorption system, contact time plays a vital role, irrespective of the other experimental parameters affecting the adsorption kinetics. In order to study the kinetics and dynamics of adsorption of fluoride by adsorbent sample HNO₃AC, the adsorption experiments have been conducted in the extent of removal of fluoride by varying the contact time from 30 to 75 minutes at fixed optimum initial concentration of fluoride 5ppm with a fixed dose of adsorbent samples 3g/L at room temperature. The effect of agitation time on the removal of fluoride by HNO₃AC is shown in Figure 1. The data clearly indicate that the removal of fluoride by these samples slightly increase with increase in the contact time. The rate of removal of fluoride is higher due to the availability of adequate surface area of the adsorbent with increase in contact time.

Effect of dose of adsorbent

The percentage of removal of fluoride as a function of carbon dose is useful to find out the optimum amount of carbon to be used in the removal of fluoride. The percent removal of fluoride by adsorption on the selected adsorbent sample has been obtained with various doses of adsorbent sample HNO₃AC range 1.0-5.0 g/L with optimum initial concentration of fluoride 5ppm and contact time 30 minutes at room temperature. The relative percentage removal of fluoride is found to be insignificant after the dose of 3g/L for the various samples, which is fixed as optimum dose of adsorbent sample. The percentage removal of fluoride increases exponentially with increasing dose of adsorbent sample. This may be due to the increase in availability of active sites caused by the increase of effective surface area, especially at higher adsorbent doses⁹. Figure 2 shows the effect of carbon dosage on fluoride adsorption onto HNO₃AC.

Removal of fluoride in bulk water samples

The adsorption capacity of adsorbent in bulk water samples have been studied by varying the dose 10-25gm of the adsorbent HNO₃AC by using standard 5 ppm fluoride solution at optimum conditions. The

results reveal that when dose of the adsorbent increases, the percent removal of fluoride increases and the amount adsorbed exponentially decreases. It may be due to the availability of active sites on the adsorbent surface.

Recycling of fluoride loaded adsorbent

The adsorption capacity of the adsorbent, for continuous use in further defluoridation experiments is important. When the discharge of effluents is of large quantity, continuous cyclic adsorption process is always effective. If cost is affordable it plays great role in economical defluoridation. In the present water management study, the fluoride-loaded adsorbent was studied five times. When the 5 ppm standard fluoride solution was passed on the adsorbent, it can remove the 83.6 percent fluoride. The same carbon was used for the next defluoridation experiment. Another 5 ppm standard fluoride solution was passed through fluoride loaded carbon. The fluoride-loaded adsorbent removes the fluoride by 80.8 percent. In this way, the adsorbent was tested for five times Figure 3. In all experiments, we were observing the continuous removal of fluoride up to maximum permissible limit.

Determination of pH before and after passing through the adsorbent

In water management pH of water shows great effect on the quality of water. In the present study fluoride solution having initial pH 6.17. It is observed that the fluoride solution after passing on the different activated samples of NDLC having the pH values in between pH 7.40 – 8.24. The slight increase in the pH value may be due to the presence of alkaline matter.

Regeneration of the fluoride loaded adsorbent

In majority of applications for the removal of fluoride reported earlier, the disposal of adsorbent as waste is not an economic option and therefore regeneration is carried out to an extent that the adsorbents can be reused. It was also very important in adsorption technique. In the present water management study, fluoride loaded carbon was regenerated by passing the adsorbent through 0.1N NaOH solution. From the results, it can be concluded that when the standard 5 ppm fluoride solution is passed through the regenerated carbon, the fluoride was removed from 77.60 to 58.0 percent. It clearly indicates that the fluoride-loaded carbon was regenerated.

Adsorption isotherms

The adsorption isotherm is highly significant in the defluoradation by the adsorption technique, as it provides an approximate estimation of the adsorption capacity of the adsorbents. The equilibrium data for the removal of fluoride by adsorption on various adsorbent samples at $30 \pm 1^{\circ}\text{C}$ have been used in Freundlich and Langmuir isotherms¹¹.

$$\text{Langmuir Isotherm: } (C_e/Q_e) = (1/ab) + (C_e/a)$$

Where Q_e is the amount of fluoride adsorbed per unit weight of the adsorbent (in mg.g^{-1}), C_e is the equilibrium concentration of fluoride (in ppm) and 'a' and 'b' are Langmuir constants. The Langmuir constants have been also determined from the linear correlations of C_e/Q_e against C_e . The adsorption isotherm parameters along with the adsorbent samples are shown in Figure 4. The applicability of Langmuir isotherm model indicates the formation of monolayer coverage of adsorbate on outer surface of the adsorbent. Further, the essential characteristics of the Langmuir isotherm can be described by a separation factor R_L , which is defined by. $R_L = 1 / (1 + b C_i)$ Where C_i is the initial concentration of fluoride (ppm) and 'b' is the Langmuir constant (g. L^{-1}). In the present, study the values of R_L indicating that the sorption process is favorable for all these adsorbent samples.

CONCLUSIONS

The results of experiments have shown that the percentage of fluoride removal has increased with the increase of contact time and dose of adsorbent. Among the four activated adsorbent samples prepared from NDLC under consideration, HNO_3AC possesses the highest or maximum adsorption capacity. Hence it is the best and the most effective adsorbent sample in the removal of the decreased fluoride

content in water. The next one in order based on its efficacy in removing the fatal content is HCl AC, H₂SO₄ AC, NAC, Na₂CO₃ AC and NaOH AC is not far behind the first two. The results show that the ground water of study area is not suitable for drinking purpose.

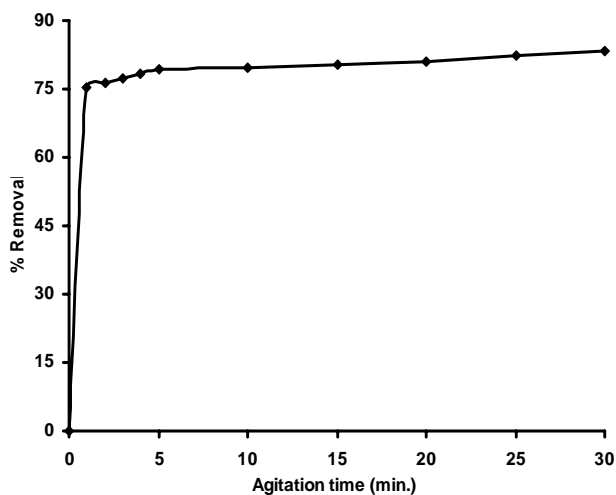


Fig.-1: The effect of agitation time on the removal of fluoride

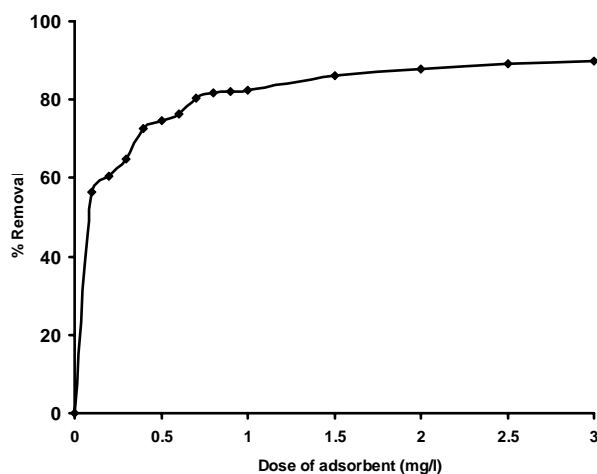


Fig.-2: The effect of carbon dosage on fluoride adsorption

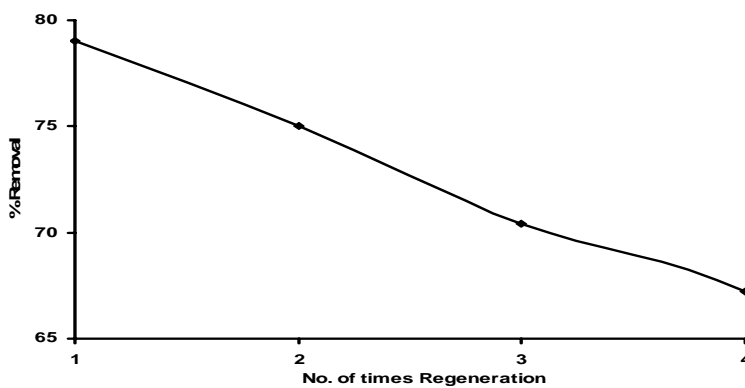


Fig.- 3: Recycling of fluoride loaded adsorbent

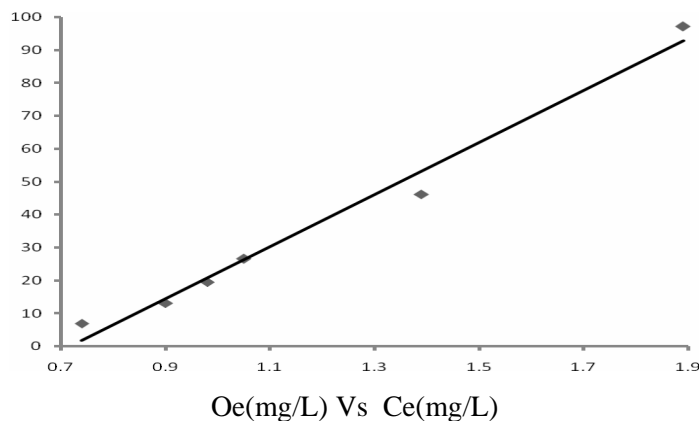


Fig.- 4: Longmuir isotherms for the removal fluoride by adsorption on NDLC

Table-1: Water fluoride ranges in before after defluoridation in 15 panchatyats

S.No	Name of the Panchayat	Fluoride(ppm)	After Defluoridation(ppm)
1	Ummadadevara Palli	1.4	0.7
2	Nemali	3.2	1.7
3	Konojerla	2.5	1.1
4	Vutukuru	1.2	0.8
5	Penugolna	3.2	1.6
6	Rajavaran	2.6	1.3
7	Mettagaudam	3.4	1.1
8	Tunikipadu	3.3	1.3
9	Gunwale Paleam	3.1	1.6
10	Satyla Padu	3.2	1.7
11	Maeduru	1.9	1.4
12	Gampalagudam	2.4	1.2
13	Ammireddygaduum	3.2	1.4
14	Chinna Komera	2.1	1.2
15	Gosavedu	1.6	0.7

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