# Fluoride Removal By Adsorption Using Activated Rice Husk

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II. FLUORIDE IN ENVIRONMENT

Abstract – Batch adsorption dynamics for the removal of Fluoride ions from aqueous solution using activated rice husk has been carried out under various experimental conditions. In the present study, activated rice husk is used as an adsorbent for the removal of fluoride from aqueous solution. The process parameters are adsorbent dosage, pH of the solution, contact time, initial fluoride ion concentration and temperature. The result showed that maximum fluoride ion removal is possible at a dosage of 5g/100ml of activated carbon for a contact time of 90 minutes and temperature of  $40^{\circ}$ C.

Keywords: IJSPR, Activated Rice Husk, Fluoride ion, Adsorption.

# I. INTRODUCTION

Water is one of the major elements essential for sustenance of all forms of life and is available in abundance in nature covering approximately three fourths of the surface of the earth. The chemical nature of water is one of the most important criteria that determines its usefulness for a specific need and as such not all the waters are fit for drinking; hence the problems of scarcity of drinking water. The presence of fluoride, in quantities in excess of limits is a serious matter of concern from a public health point of view. Like any other pollutant the fluoride pollution can also occur due to both natural and manmade reasons.[1]

Fluoride in drinking water is known for both beneficial and detrimental effects on health [2]. The fact that the problems associated with the excess fluoride in drinking water is highly endemic and widespread in countries like India prompted many researchers to explore quite a good number of both organic and inorganic materials adopting various processes from coagulation, precipitation through adsorption, Ion exchange etc [4]. Some are good under certain conditions while others are good in other conditions. Leaching of Fluoride from the earth crust is the chief source of fluoride content in ground water; however the other sources like food items also add to increase the overall ingestion of fluoride into the human body.

Defluoridation is the process of removal of fluoride ion in drinking water. Among these defluoridation process adsorption process is very effective, eco friendly, high removal efficiency and also low cost. Basically we used modified bio adsorbent for high removal of fluoride. The possible causes and sources through which fluoride exists in the environment are schematically shown in Figure 1.



Figure 1. Possible causes for fluoride in groundwater

Table1. Fluoride bearing minerals [5]

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Mineral	Chemical Formula	% Fluorine
Sellaite	MgF2	61
Villianmite	NaF	55
Fluorite (Fluorspar)	CaF2	49
Cryolite	Na3AlF6	45
Fluorapatite	Ca3(PO4)3F	3-4
Bastnaesite	(Ce,La) (CO3)F	9

Fluorine ( $F_2$ ) is a greenish diatomic gas. Fluorine is so highly reactive that it is never encountered in its elemental gaseous state except in some industrial processes [5]. The fluoride occurs notably as Sellaite, fluorspar, CaF<sub>2</sub>; Cryolite, Na<sub>3</sub>AlF<sub>6</sub>; Fluorapatite, 3Ca<sub>3</sub> (PO4)<sub>2</sub> Ca (F,C<sub>12</sub>). Other minerals containing fluoride are given in the table 1.[3]

As fluorspar it is found in sedimentary rocks and as Cryolite in igneous rocks. These fluoride minerals are nearly insoluble in water [6]. Hence fluorides will be present in ground water only when conditions favour their solution1. It is also present in sea water (0.8-1.4 ppm), in mica and in many drinking water supplies [8].

# III. GLOBAL AS WELL AS INDIAN SCENARIO OF FLUORIDE

Groundwater with fluoride concentration above the permissible limit set by WHO i.e. 1.5 mg/l have been recorded in several parts of the world. In 1984, WHO estimated that more than 260 million people living all over the world consume water with fluoride concentration above 1 mg/l (WHO, 1984)[7]. The problem of high fluoride in groundwater has been reported by several researchers in India, China, Japan, Sri Lanka, Iran, Pakistan, Turkey, Southern Algeria, Mexico, Korea, Italy, Brazil, Malawi, North Jordan, Ethiopia, Canada, Norway, Ghana, Kenya, South Carolina, Wisconsin and Ohio [9]. The other possible sources of intake of fluoride apart from drinking water are through food, beverages and dental products like tooth paste. A detailed description on the concentration of fluoride in groundwater and its sources in various regions of the world based on literature are given in Figure 2.



Figure 2. Occurrence of fluoride in groundwater in various parts of the world based on literature

# India

Out of the 85 million tons of fluoride deposits on the earth's crust, 12 million are found in India (Teotia and Teotia, 1994). Hence it is natural that fluoride contamination is widespread, intensive and alarming in India [10]. Endemic fluorosis is prevalent in India since 1937 (Shortt et al., 1937). It has been estimated that the total population consuming drinking water containing elevated levels of fluoride is over 66 million (FRRDF, 1999). Different parts of India where elevated

concentration fluorides in groundwater as reported in literature are shown in the figure 3.[11]



Figure 3. Range of fluoride in groundwater in India based on literature

#### IV. PROPOSED METHODOLOGY

#### Preparation of Adsorbent

There are a lot of bio-adsorbent present in the world that can be used to remove fluoride from water irrespectively their capacity. We used in this study Rice husk as a bio adsorbent. Rice husk is obtained from Paddy seeds and it was dried at  $100^{\circ}$ C for 6 hr to remove the adherent moisture. Then it grinded in mixer grinder and then sieved through standard sieve to obtain particle of mesh size 60. After that the sample was stored in an air tight container.

#### Preparation of Activated Carbon

5gm of sample was taken in a silica crucible and it was mixed with 5ml Phosphoric acid. The resulting mixture was then put in Muffle furnace at  $400^{\circ}$ C for 10 minutes and then upto  $750^{\circ}$ C for 35 minutes. After that the activated mixture was transferred and the excess acid was removed by filtering through Whatmann 41 filter paper by hot water and cold water. The filtrate pH was checked and it was maintained at pH 6.5.The wetted mass was thoroughly dried at  $100^{\circ}$ C.

#### Preparation of stock solution

Here the sample is NaF (Merck) which is used for the preparation of stock solution. The percentage of fluoride in NaF is 45.247. So 100 mg/l stock solution is prepared by dissolving 221mg (We used sodium fluoride having

purity of 97%.So we take 227.84gm of NaF) of sodium fluoride in 1000ml distilled water.

## Preparation of Zr- SPANDS solution

To prepare  $3.72 \times 10^{(-3)}$  M SPANDS (Merck) solution, 0.4750 gm of SPANDS reagent is dissolved in 250 ml of double distilled water. The zirconyl acid solution,  $3.72 \times 10^{(-2)}$ M is prepared by dissolving 0.0665 gm of ZrOCl<sub>2</sub>.8H<sub>2</sub>O in 25 ml of double distilled water. Then 25 ml of HCl acid is added and then distilled water is added until the total volume of solution is 250ml. The conc. Of Zr-SPANDS solution is prepared by mixing the two solutions at 1:1 volume ratio.

# V. EXPERIMENTAL RESULTS&DISCUSSION

Adsorption experiment and analysis-

We studied batch adsorption for analysis. Adsorption experiments were carried out for the determination of particle size, concentration, optimum pH value, contact time, weight of the adsorbent &temperature. The influence of pH (3.0-10.0), contact time (30,60,90,120,150and 180 min), initial fluoride concentration (3,3.5,4,4.5,5,and 5.5g/L), temperature(20 to  $70^{\circ}$ C), weight of the adsorbent dose (2,3,4,5,6,7g/l) were evaluated during the study in a 50 ml flasks with fluoride solution of known concentration. The Powdered neem leaves as an adsorbent were added in each flask and the flasks were kept for constant shaking for 1hr and then the solids were separated through filtration. The solutions were collected and developed the colour and fluoride concentration in the solution was determined by using spectrophotometer. Each experiment was conducted three times and average values are reported. The amount of fluoride adsorbed per unit adsorbent (mg fluoride per g adsorbent) was calculated according to a mass balance on the fluoride concentration using Eq:

$$q_{e} = \frac{(c_{i} - c_{f})x}{w}....(1)$$

Where, X = volume of the solution

$$W = weight of adsorbent (g)$$

The percent removal (%) of Fluoride was calculated using the following equation:

Removal (%) = 
$$\frac{(c_i - c_f) \times 100}{c_i}$$
 .....(2)

## Effect of pH

The effect of pH on the adsorption of fluoride by using activated carbon is shown .The adsorption pattern of fluoride by using activated carbon has been studied varying the pH from 3 to 10. Initially at low pH the percentage of removal is low.When pH increasing percentage removal also increases; it reaches maximum at pH 5 for activated carbon then again decreases.



Figure 4: Effect of pH on removal of fluoride

## Effect of contact time

The effect of contact time on the adsorption of fluoride by using activated carbon shown the adsorption pattern of fluoride by using activated carbon has been studied varying the contact time from 30 minutes to 180 minutes. It was observed that with fixed amount of adsorbent the removal of fluoride increased with time and attained equilibrium after 90 minutes. After reaching maximum point of removal the adsorbent reached in saturated condition.



Figure 5: Effect of contact time on removal of fluoride

## Effect of Adsorbent dose

The effect of adsorbent dose on the adsorption of fluoride by using activated carbon bagasse is shown. The adsorption pattern of fluoride by using activated carbon has been studied varying the adsorbent dose from 2 g/100ml to 7 g/100ml.It can be seen the percentage of removal increased with increasing dosage of adsorbent. The largest percentage removal was exhibited at 5 g for activated carbon.



Figure 6: Effect of adsorbent dose on removal of fluoride

## **Effect of Initial concentration**

The effect of initial concentration on the adsorption of fluoride by using activated carbon is shown. The adsorption pattern of fluoride by using activated carbon has been studied varying the initial concentration from 3 mg/L to 5.5 mg/L.It can be seen that the percentage removal of fluoride decreased with increasing initial concentration. At initial concentration 3 mg/L the fluoride removal percentage is maximum.



Figure 7: Effect of concentration on the adsorption of fluoride

# **Effect of Temperature**

The effect of temperature on the adsorption of fluoride by using activated carbon bagasse is shown. The adsorption pattern of fluoride by using activated carbon has been studied varying the temperature 20 ° C to 70 ° C. Initially Fluoride removal increased with increasing temperature & then decreased for activated carbon. The maximum percentage of removal of at 40°C for activated carbon.



Figure 8: Effect of temperature on fluoride removal

## VI. CONCLUSION

The experiment was aimed to remove the fluoride ion concentration from aqueous solution by using activated rice husk adsorbents using adsorption technique. The adsorption of fluoride was depends on pH, initial concentration, contact time, temperature as well as adsorbent dose which were optimized. The equilibrium agitation time for the adsorption of fluoride is 90 min. Percentage removal is increased up to pH of 5.The removal rate is increased with increasing the adsorbent dose and at 5gm/100ml adsorbent dose the removal was found to be maximum. In the following conditions maximum removal was achieved 92%.The present study concluded that the activated rice husk material as an adsorbent can be utilized effectively for fluoride removal.

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