Experimental Investigations on Removal of Fluoride in Groundwater using *Prosopis juliflora*

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***Abstract*— One of the methods is adsorption techniques, which is a low cost technique used for effective removal of fluoride from groundwater. This study started with preparation of activated carbon from *Prosopis juliflora*, characterized the activated carbon prepared from *Prosopis juliflora*, studied the effect of agitation speed and dosage, particle size on removal of fluoride in ground water using *Prosopis juliflora* activated carbon. From this study, it was observed that the maximum removal of fluoride found to be 73.21, 93.45, and 97.26 % respectively for the agitation speed of 500 rpm, dosage of 1.5 g and contact time of 45 min. against the particle size 3.2 mm.**

***Keywords*—groundwater, Sugarcane bagasse, *Prosopis juliflora*, Process parameter**

I. INTRODUCTION

Fluorine due to its high electronegative and reactivity cannot found in natural environment in elemental form. Fluoride (F^-) is a fluorine anion which has a great tendency to behave as ligand and easiness to form a great number of different organic and inorganic compounds in soil, rocks, air, plants and animals. These compounds are quite soluble in water, so fluoride is present in surface and groundwater as an almost completely dissociated fluoride ion and from the pollutants discharged through water and wastewater are contributing fluoride ions in the ground water [6]. The major source of fluoride to a living being is drinking water. The high level of fluoride (> 1.5 mg/l) in water is responsible for dental and skeletal fluorosis besides growth retardation. Many developing countries have being facing problems of fluorosis such as that of India. India and China have a few fluoride problems as per World Health Organization (WHO). Fluoride is a toxic chemical and it is a risk factor for thyroid hormone production in children when the exposure to fluoride occurs during intrauterine growth period.

There are several processes that can be adopted for the treatment of water and industry wastewater such as such as reduction followed by chemical precipitation, sedimentation, electrochemical processes [9,12], ion exchange, biological operations, bioremediation [7,8,10,17,18,22], biosorption [4,13,23], cementation, coagulation / flocculation [17], filtration and membrane processes, and solvent extraction.

Earlier works revealed the suitability of variety of agro-based materials like moriga oleifera seed, corncob, groundnut husk, rice husk, tea leaves carbon, saw dust to treat the industrial wastewater [2,3,5]. The adsorbent prepared from biomaterial has large surface area and micro porous character nature have made them potential adsorbents for the removal of heavy metals from industrial waste water [11,14-16,19-21].

The aim of this study is to conduct the experiment for removing fluoride ion present in ground water using *Prosopis juliflora* activated carbon under various conditions. The *Prosopis juliflora* has a high economic value which is used as fuel, anti-oxidant in human body but also it has many environmental impacts. The main objectives of this study are (i) to prepare activated carbon from *Prosopis juliflora*, (ii) to characterize the activated carbon prepared from *Prosopis juliflora*, and (iii) to study the effect of agitation speed, dosage, contact time and particles size on removal of fluoride in ground water using *Prosopis juliflora* activated carbon.

II. LITERATURE REVIEW

V. Jamode, V. S. Sapkal and V. S. Jamode (2004) assessed the suitability of inexpensive leaf adsorbents to effectively remediate fluoride-contaminated water. The leaves of neem (*Azadirachta indica*), pipal (*Ficus religiosa*) and khair (*Acacia catechu wild*) trees were used in this study. The study concluded that at the highest fluoride ion concentration (15 mg/l), the fluoride ion level in the effluent gradually decreased to 0 mg/l within 180 min at 29 ± 0.5°C when the dose of adsorbent is 10 g/l. The process adsorption by treated bio sorbents followed Langmuir isotherm, which comprises statistical and empirical data estimated from Isotherm equation.

S. Kagne, S. Jagtap, P. Dhawade, S.P. Kamble, S. Devotta, S.S. Rayalu (2007) investigated that the potential of cement hydrated at various time intervals for the removal of excess F^- from aqueous solution by using batch adsorption studies. It was found that the HC shows significant F^- removal over a wide range of pH (3–10). The maximum removal of fluoride was 92.37% with 10gm/l of hydrated cement, contact time 24 hours and initial fluoride concentration 5.9 mg/l. The experimental data generated from batch adsorption experiments fitted well into the linearly transformed Freundlich and Langmuir isotherms.

G. Alagumuthu, V. Veeraputhiran and R. Venkataraman (2010) investigated the removal of fluoride from the water using cynodon dactylon as adsorbent. By conducting batch adsorption studies at constant temperature (25-32°C), the maximum removal of fluoride was 83.77 % while keeping 3.0 mg/L fluoride concentration and 1.25 g dosage of adsorbent at neutral pH, contact time 105 minutes. Various adsorption isotherm models were applied to evaluate the adsorption data. The adsorption of fluoride ions followed Redlich-Peterson isotherm as well as Langmuir isotherms. The authors concluded that the adsorbent used in this work shows superior adsorptive efficiency than previously studied defluoridation works using natural adsorbents was most effective.

R. Bhaumik, N. K. Mondal, B. Das, P. Roy, K. C. Pal, C. Das, A. Banerjee, and J. K. Datta (2011) studied the role of eggshell powder as an adsorbent for removal of fluoride from aqueous solution using batch technique. The maximum adsorption was achieved at pH 2.0-6.0. The investigators achieved around 94% removal of fluoride at initial metal ions concentration of 5 mg/l at optimum dose of 2.4 g/100ml and optimum time of 120minutes. Experimental data provided best fit with the Langmuir isotherm model, indicating monolayer sorption on a homogenous sur. The adsorption kinetics followed pseudo-second-order kinetic model indicating towards chemisorption. Intra-particle diffusion was not the sole rate controlling factor.

S. A. Valencia-Leal, R. Cortés-Martinez, R. Alfaro-Cuevas-Villanueva (2012) Evaluated the Guava Seeds (Psidium Guajava) as Low-Cost Bio sorbent for the Removal of Fluoride from Aqueous Solutions. Maximum adsorption occurred between pH 5.0 to 8.0.The adsorption of fluoride was found endothermic in nature Langmuir and Freundlich adsorption isotherm models were applied to evaluate the adsorption data. The pseudo second order model describes the fluoride sorption kinetics using guava seed at different temperature. The Langmuir model best describes the isotherm's experimental data, which may indicate that the sorption mechanism of fluoride ions on guava seed is chemisorption on a homogeneous material.

III. Materials And Methods

A. Prosopis Juliflora General View

Prosopis juliflora is one of the most economically and ecologically important tree species in arid and semi-arid zones of the world. It is an important species because of its high nitrogen fixing potential in very dry areas and in drought seasons and also because of it provides shelter and food to many species of animals on its nectar, pollen, leaves and fruits. The shrubs of *Prosopis juliflora* are highly esteemed for windbreaks, soil binders, sand stabilizers, living fences, fuel wood, bee plants and animal feed. These uses, together with fast growth, drought resistance and salt tolerance have led to its introduction in many arid zones. In India, original introductions are thought to have been *Prosopis juliflora* from Mexico or Jamaica. Differences in plant morphology may be due to further introductions of seed material of various origins and possible hybridization between them.

To conquer the hazardous wellbeing impact of fluorosis, different approaches for defluoridations are exists like coagulation, precipitation, membrane separation processes, ion exchange, adsorption techniques, electro-dialysis and electrochemical etc. Each approach has their advantages and limitations and worked productively under ideal condition to remove fluoride to more noteworthy range.

This study uses adsorption techniques. A few adsorbent materials have been attempted in the past to check their possibilities and techno-economic feasibility as defluoridating specialists. Activated alumina, activated carbon, activated alumina coated silica gel, calcite, activated saw dust, activated coconut shell powder, activated fly ash, groundnut shell, coffee husk, rice husk, magnesia, serpentine, tri-calcium phosphate, bone charcoal, activated soil sorbent, defluoron-1, defluoron-2 and so on various adsorbent materials reported in literature.

B. Spectrophotometer

Spectrophotometer is the quantitative measurement of the reflection or transmission properties of a material as a function of wavelength. Spectrophotometer uses photometers that can measure a light beam's intensity as function of its color (wavelength) known as spectrophotometers. Important features of spectrophotometers are spectral bandwidth, (the range of colors it can transmit through the test sample), and the percentage of sample-transmission, and the logarithmic range of sample-absorption and sometimes a percentage of reflectance measurement [1].

C. Magnetic Stirrer

A magnetic stirrer or magnetic mixer is a laboratory device that employs a rotating magnetic field to cause a stir bar immersed into a liquid to spin very quickly, thus stirring it. The rotating field may be created either by a rotating magnet or a set of stationary electromagnets, placed beneath the vessel with the liquid. It should be used with the closed vessel such as standard flask or conical flask with closets. With the help of magnetic stirrer we can change the parameter such as temperature, agitation speed, contact time and dosage of the material used.

D. Collection of Stem and Chopping

The stem of *Prosopis juliflora* of 3years plant is collected and it is chopped in the center of timber sawing. Then the chopped material is sun dried for 3 days then it is collected and stored in non-contaminated place. It should be used for converting the activate carbon.

Stem

Chopped

E. Sieving

The ready product of chopped one is to be sieved. The general size of the sieving is 4.75mm to 75 micron. But in this project is used the sizes of 3.2mm of retained in the corresponding sizes.

F. Activated Carbon

The size of 3.2 mm is used for activated carbon preparation. Totally nine sets of activated carbon is prepared with the use of muffle furnace. By changing the parameter of contact time and temperature, nine sets of activated carbon is prepared.

The various sets were prepared from the saw powder of the *propis juliflora*, obtained from wastelands and these were kept in the muffle furnace with various contact time with various temperature for the particle size of 3.2mm. The temperatures such as 150 °C, 200 °C and 250 °C with various contact time of 15 minutes, 30 minutes, 45 minutes are followed in this study.

IV. Results And Discussion

A. Characterization of Activated Carbon

To determine the characterization of activated carbon this study uses methylene blue test (according to IS 875). In this method, 0.5gm of methylene blue is dissolved in one liter of distilled water and need to know the wavelength of the methylene blue.

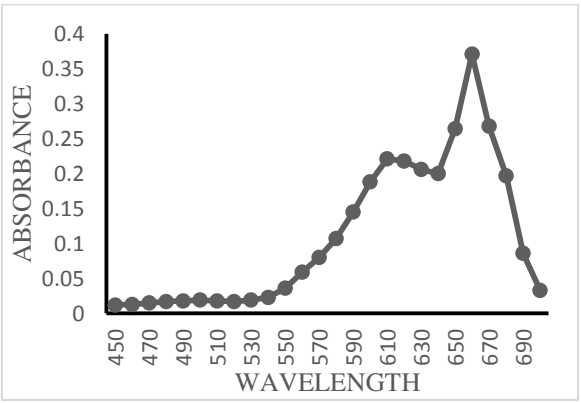

Fig. 1 Absorbance characteristics of *propis juliflora in Methylene Blue*

Fig. 1 shows that methylene blue has highest absorbance value at the wavelength of 660 nm. Thus, further study was completed with this wave length 660 nm.

B. Initial Absorbance Value

The various concentration in the form of mg/l is made from the stock solution of methylene blue. The concentration of 1 mg/l, 2 mg/l, 3 mg/l, 4 mg/l, and 5 mg/l are made from standard solution. Then the absorbance value is taken from the spectrophotometer at the 660 nm. The carbon produced at 200 °C and 30 min. was used for the removal of methylene blue and the same optimum conditions are followed for fluoride removal from ground water.

C. Sodium Fluoride Calibration

To find the unknown concentration of fluoride in the groundwater, the calibration curve should be plotted by known concentration of the fluoride. The available sample for fluoride is sodium fluoride. It is diluted to 1 mg/l, 2 mg/l, 3 mg/l, 4 mg/l, 5 mg/l, and 6 mg/l. To find the highest wavelength, the concentration of least value is selected for calibration, which is 1 mg/l.

To find the absorbance, reagent should be added which is methylene orange. It is prepared by adding 0.327 gm of methylene orange and 0.0665 gm of aluminium chloride and 25 ml of ethanol. One drop of reagent is added to the sample which is having the fluoride content. The below value gives the calibration of 1 mg/l of sodium fluoride.

Methylene Orange Sodium Fluoride

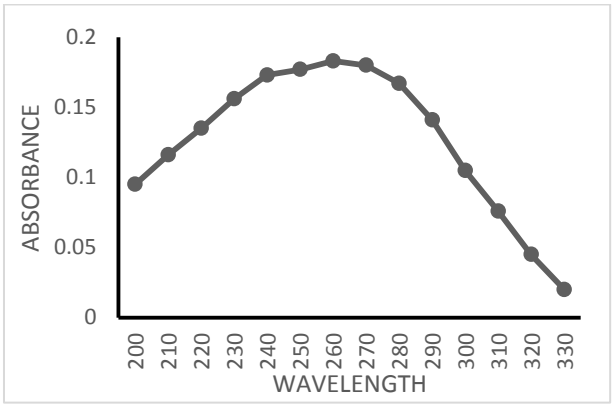

Fig. 2 Absorbance characteristics of *propis juliflora in Sodium Fluoride*

Fig. 2 shows that sodium fluoride has highest absorbance value at the wavelength of 260 nm. Thus, further study was completed with this wave length 260 nm.

D. Effect of Agitation Speed

The fluoride solution is collected in the magnetic stirrer and removal efficiency is studied by changing the agitation speed of 125 rpm, 250 rpm, 375 rpm and 500 rpm with the contact time of 15 minutes. After 15 minutes the solution was kept undisturbed for 24 hours. The 24 hours is due to the settlement of the activated carbon. The amount of activated carbon used is 500 mg and optimum agitation speed, for which the maximum removal was obtained as 500 rpm. The maximum removal due to agitation speed is presented in Table 2. From Table 2, it may be observed that the maximum removal was 73.21 %.

TABLE 2
MAXIMUM REMOVAL OF FLUORIDE DUE TO AGITATION SPEED

Agitation Speed	Percentage Removal (%)
125	4.46
250	12.79
375	65.17
500	73.21

E. Effect of Activated Carbon Dosage

The maximum removal efficiency is obtained by changing the activated carbon dosage of 0.5 g, 1 g, 1.5 g and 2 g with the contact time of 15 minutes and an optimum agitation speed of 500 rpm. After 15 minutes the solution is kept for 24 hours without any changes. The 24 hours is due to the settlement of the activated carbon. The optimum dosage for which the maximum removal was obtained as 1.5 g. The maximum removal due to adsorbent dosage is presented in Table 3. From Table 3, it may be observed that the maximum removal was 93.45 %.

TABLE 3
MAXIMUM REMOVAL OF FLUORIDE DUE TO ADSORBENT DOSAGE

Dosage (g)	Percentage Removal (%)
0.5 gm	73.21
1.0 gm	81.25
1.5 gm	93.45
2.0 gm	85.41

C. Effect of Contact Time

The maximum removal efficiency is obtained by changing the contact time of 15, 30, 45 and 60 min with an optimum adsorbent dosage of 1.5 g and an optimum agitation speed of 500 rpm. After each contact time, the solution is kept for 24 hours without any changes. The 24 hours is due to the settlement of the activated carbon. The optimum contact time for which the maximum removal was obtained as 45 min. The maximum removal due to contact time is presented in Table 4. From Table 4, it may be observed that the maximum removal was 97.26 %.

TABLE 4
MAXIMUM REMOVAL OF FLUORIDE DUE TO ADSORBENT DOSAGE

Contact Time (min)	Percentage Removal (%)
15	93.45
30	97.16
45	97.26
60	97.20

V. CONCLUSION

The *Prosopis juliflora* is taken from the wasteland which was converted into activated carbon. The characteristics of the activated carbon is determined from the methylene blue solution with different concentrations the optimum wavelength is acquired from spectrophotometer. The experiments are conducted to optimize fluoride removal by different parameters of agitation speed, dosage, and contact time with help of magnetic stirrer apparatus. The maximum removal percentage of fluoride is 97.26 % was obtained for the *Prosopis juliflora* activated carbon particle size of 3.2 mm with an optimum agitation speed of 500 rpm, dosage of 1.5 g and contact time of 45 min. From the experimental investigation, it is concluded that *Prosopis juliflora* activated carbon is effectively used as adsorbent for removing fluoride in groundwater.

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