



Hibiscus Rosa-Sinensis Leaf powder as an Adsorbent for Defluoridation from Aqueous Solution

J. Sharpudin¹, Barisha Kharjana², V. Christina Rajam³, A. Kousiya Jabin⁴

¹ Assistant Prof., Department of Civil Engineering, C. Abdul Hakeem College of Engineering and Technology, Vellore, Tamil Nadu, India

^{2,3,4} P.G. Student, M.E. Environmental Engineering, C. Abdul Hakeem College of Engineering and Technology, Vellore, Tamil Nadu, India

Abstract: Concentration of fluoride in drinking water is influenced by a number of factors, such as temperature, pH, solubility of fluorine bearing minerals, anion exchange capacity of aquifer materials (i.e., OH⁻ for F⁻). About 80% of diseases in the world are due to poor quality of drinking water and 65% is for fluoride contamination cause endemic fluorosis across the globe. There is no medicine for fluorosis, but treatment systems can regulate the amount of fluoride in water. This paper investigates the use of *Hibiscus rosa-sinensis* (Hibiscus leaf) as natural adsorbent for fluoride removal carried out at varied pH, contact time and adsorbent dosage. Based on batch experiments, maximum efficiency was observed 95% at pH-6.

Keywords: Fluorosis, World Health Organization (WHO), Adsorption, Hibiscus leaf.

I. INTRODUCTION

The inorganic contaminants commonly found in groundwater namely fluoride and other major ions like calcium, magnesium, chloride, carbonate, bicarbonate, sodium, potassium, sulphate and nitrate in addition to pH and electrical conductivity. Fluorine occurs as a negatively charged ion in water. It is one of the lightest halogen and the most reactive of all chemical elements (Kaminsky *et al.* 1990)^[34]. Concentration of fluoride in drinking water is influenced by a number of factors, such as temperature, pH, the absence or presence of complexing or precipitating ions and colloids, solubility of fluorine bearing minerals, anion exchange capacity of aquifer materials (i.e., OH⁻ for F⁻). The size and type of geological formations and the time period during which water remains in contact with a particular formation (Apambire *et al.* 1997)^[17]. Fluorosis affects millions of people around the world as it is dangerous and very deadly disease. In places where fluoride level are between 1.5 – 2.00 mg/l, it has some nutritional properties.

As prescribed by World Health Organisation and Indian Council of Medical Research (WHO 2011; ICMR 1975) fluoride maximum permissible limit is 1.5 mg/l. Due to excess of fluoride in drinking water more than 200 million

people suffer from endemic fluorosis (Mesdaghinia *et al.* 2010)^[13].

Likewise, fluoride in India contaminates 50% in groundwater sources and more than 90% of villages depend on groundwater for drinking purposes (Subarayan *et al.* 2012)^[16]. In 1937, state of Andhra Pradesh top for excess of fluoride in India. The highest concentration of fluoride which affects the people have found in Nalgonda District of Andhra Pradesh. The major health problems are dental and skeletal fluorosis and deformation of bones in children and adults if consume excessive of fluoride in drinking water (Susheela *et al.* 1993)^[21]. Children less than 7 years old are vulnerable to fluorosis as well as impact on teeth growth (Murray 1996). Lack of Ca, vitamins and proteins in diet is also an adverse health effects on human (Jacks *et al.* 1993)^[19].

Hibiscus rosa-sinensis is an evergreen flowering shrub belongs to Malvaceae family. Hibiscus leaf contained appreciable quantities of carotene, fair levels of iron, ascorbic acid phosphorus and calcium with traces of tannins, phytates and cyanide.



II. MATERIALS AND METHODS

The glassware were cleaned with acid and rinsed with water before use. Fluoride stock solution was prepared by taking 221.01 mg of sodium fluoride (NaF) dissolved in 1L distilled water. 100 ppm of fluoride contain in the stock solution.

A. Natural adsorbent preparation

The natural leaf was collected in an around Melvisharam (Vellore District) area. The leaf was washed with distilled water, dried in an oven for 2-3 hrs at 105°C. The dried leaf was finely powdered and use for acid treatment.

B. Acid treatment

22 gm of powdered adsorbent was added to 220 ml containing 1M HCl in a conical flask and was gently heat for 15 mins. Then, its was filtered and washed with distilled water using whatmann filtered paper and the solid sample was dried in a hot air oven for a period of 6-7 hrs at 105°C.

C. Characterization of adsorbent

Adsorbents were characterised according to European Council of Chemical Manufacturer's Federation (CEFIC).

D. Equipment used

Fluoride was estimated by using Fluoride Kit. Other equipment used are Erlenmeyer flask, beaker, standard measuring flask, measuring jar, pH meter, incubator shaker for agitating the samples at 120 rpm, whatmann filter paper and funnel respectively.

A. Effect of pH

By using *Hibiscus rosa-sinensis* leaf adsorbent, in each measuring flask 0.75 mg adsorbent dosage was added and agitate in orbital shaker for 150 min contact time and pH value were adjusted at 2.0, 4.0, 5.0, 6.0 and 7.0. The pH of the solution was maintained. Maximum removal efficiency of fluoride was obtained 95% at pH 6 adsorbent, above pH 6 it was reduced. Which is similar work with Sheo Prasad *et al.*, (2016)^[7] with pH above 6.5, decreased the percentage removal of fluoride. Thus, pH-6 was study for further experiment.

Table 2 Operating condition for effect of pH

Adsorbent dosage	0.95 mg (Hibiscus Leaf)
Time of contact	150 min
Volume of sample	50 ml

Table 3 Effect of pH

	pH	Initial Fluoride Conc. C ₀ (mg/l)	Final Conc. C _e (mg/l)	Removal % Of Fluoride
Hibiscus Leaf	2	9	1.82	79
	4	9	1.72	81
	5	9	0.90	90
	6	9	0.45	95
	7	9	0.99	89

III. RESULTS AND DISCUSSION

TABLE I
CHARACTERIZATION OF ADSORBENT

Parameters	Hibiscus leaf
Bulk density (g/cm ³)	0.26
Particle density (g/cm ³)	0.30
Water soluble (%)	1.14
Acid soluble (%)	2.46
Moisture content (%)	2.22

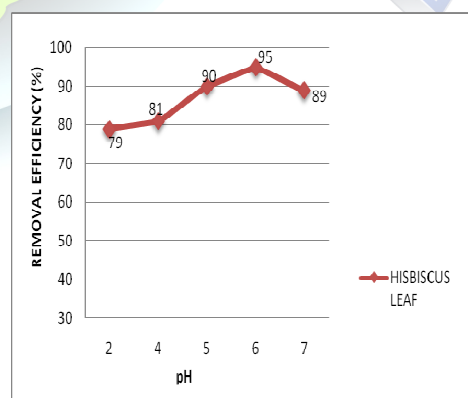


Fig. 1 Removal of Fluoride by variation in pH



B. Effect of Contact time

By using *Hibiscus rosa-sinensis* leaf adsorbent, in each measuring flask 0.75 mg adsorbent dosage was added at pH 6 and agitate in orbital shaker at varied contact time from 30-150 min. Maximum removal efficiency of fluoride was obtained 89% at 150 min. Similar work as A. Balouch *et al.*, (2013) ^[15] fluoride sorption using coal increased with increased in contact time. Thus, 150 min contact time was study for further experiment.

Table 4 Operating condition for effect of contact time

Adsorbent dosage	0.95 mg (Hibiscus leaf)
pH	6
Volume of sample	50 ml

Table 5 Effect of Contact Time (min)

	Contact Time (min)	Initial Fluoride Conc. C ₀ (mg/l)	Final Conc. C _e (mg/l)	Removal % Of Fluoride
Hibiscus Leaf	30	9	5.40	40
	60	9	3.60	60
	90	9	1.80	80
	120	9	1.10	87
	150	9	0.99	89

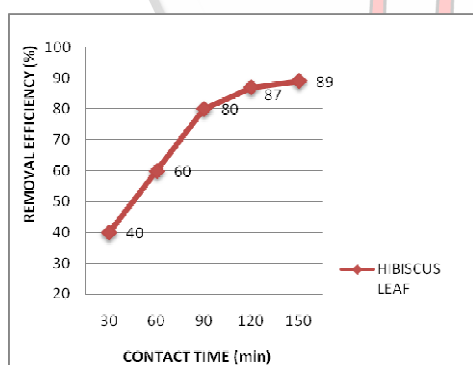


Fig. 2 Removal of Fluoride by variation in Contact Time

C. Effect of Adsorbent Dosage

By using *Hibiscus rosa-sinensis* leaf, with 150 min contact time, pH-6 and varied adsorbent dosage from 0.15, 0.35, 0.55, 0.75 to 0.95 mg. Maximum removal efficiency of fluoride was obtained 95% at 0.95 mg. Likewise the removal efficiency increases, while increasing and decreasing the adsorbent amount. Similar finding as Asha Gupta *et al.*, (2006) ^[18] the fluoride removal increased with an increased in adsorbent dosage with smaller particle size having active surface area. Whereas with large particle size the adsorption capacity is decreased. Thus, 0.95 mg adsorbent dosage was proceed for further experiment.

Table 6 Operating condition for effect of Adsorbent dosage

Time	150 min
pH	6 (Hibiscus leaf)
Volume of sample	50 ml

Table 7 Effect of adsorbent dosage

	Adsorbent Dosage (mg)	Initial Fluoride Conc. C ₀ (mg/l)	Final Conc. C _e (mg/l)	Removal % Of Fluoride
Hibiscus Leaf	0.15	9	5.40	40
	0.35	9	2.10	76
	0.55	9	0.80	80
	0.75	9	0.59	93
	0.95	9	0.45	95

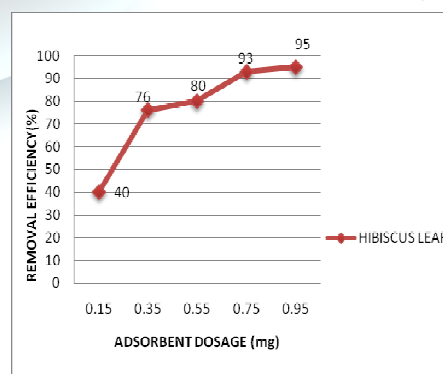


Fig. 3 Removal of Fluoride by variation in Adsorbent dose



IV. CONCLUSION

- For pH study, using Hibiscus leaf powder adsorbent the percentage of fluoride removal increase as pH increase from 2 to 6, the maximum was obtained **95% at pH 6**.
- For Contact time study, using Hibiscus leaf powder adsorbent the percentage of fluoride removal increase with increase in time from 330-150 min, the maximum was obtained **89% at pH 150 min**.
- For Adsorbent dosage study, using Hibiscus leaf powder adsorbent the percentage of fluoride removal was obtained **95% at 0.95 mg**.

REFERENCES

- [1] A.S. Parlikar *et al.*, "Defluoridation Of Water by Moringa Oleifera-A Natural Adsorbent", International Journal of Engineering Science and Innovative Technology (IJESIT) Volume 2, Issue 5, September 2013.
- [2] P. K. Shrivastava and A. Deshmukh, "Defluoridation of Water with Natural Zeolite," Journal of the Institution of Public Health Engineers (India), Vol. 14, No. 2, 1994, pp. 11-14.
- [3] Deshmukh Waheed, S. Attar, S. J. and Waghmare, M. D., Investigation on Sorption of Fluoride in Water Using Rice Husk as an Adsorbent, Nature Environment and Pollution Technology, An International Quarterly Scientific Journal, (2009) Vol.8, No.2, pp. 217-223.
- [4] A. Narsimha *et al.*, "Contamination of fluoride in groundwater and its effects on human health: a case study in hard rock aquifers of Siddipet, Telangana state, India", September 2017 Issue 5, pp 2501-2512.
- [5] Rajan *et al.*, "The fluoride removal capacity of zirconium impregnated walnut shell carbon (ZIWSC)", ISSN: 2277-9655.
- [6] Sutapa Chakrabarty and H.P.Sarma, "Defluoridation of contaminated drinking water using neem charcoal adsorbent: Kinetics and equilibrium studies", International Journals of Chem Tech Research. Vol.4, No.2, April-June 2012, 511-516.
- [7] Sheo Prasad Shukla *et al.*, "Removal of fluoride from aqueous solution using *Psidium guajava* leaves", Desalination and Water Treatment (2016) 1-8.
- [8] A. Tor, N. Danaoglu, G. Arslan and Y. Cengelloglu, "Re- moval of Fluoride from Water by Using Granular Red Mud: Batch and Column Studies," Journal of Hazardous Material, Vol. 164, No.1, 2009, pp. 271-278.
- [9] Shubha Dwivedi *et al.*, " Bioadsorption of Fluoride by *Ficus religiosa* (Peepal Leaf Powder): Optimization of process Parameters and Equilibrium study", Research Journal of Chemical Sciences Vol. 4(7), 52-60, July (2014)
- [10] Gandhi, N., Sirisha, D., Chandra Shekar, K.B. and Asthana, S., Removal Of Fluoride From Water And Waste Water By Using Low Cost Adsorbents, International Journal of ChemTech Research, (Oct-Dec 2012), Vol.4, No.4, pp1646-1653.
- [11] Meenakshi, Maheshwari R.C., Fluoride in drinking water and its removal, Elsevier B.V, (16 February 2006).
- [12] Kaminsky LS, Mahoney MC, leach J, Melius J, Miller JM (1990) Fluoride: benefits and risks of exposure. Crit Rev Oral Biol Med 1:261-281.
- [13] Mesdghinia Alireza, Vaghefi Kooshir Azam, Montazeri Ahmad, Mohebbi Mohammad Reza, Saeedi Reza (2010), "Monitoring of fluoride in groundwater resources of Iran. Bull Environ Contam Toxicol 84:432-437.
- [14] Veeraputhiran V. and Alagumuthu G., Treatment of High Fluoride Drinking Water Using Bioadsorbent, Research Journal of Chemical Sciences (July 2011), Vol. 1(4), 49-54.
- [15] A.Balouch *et al.*, "Sorption Kinetics, Isotherm and Thermodynamic Modeling of Defluoridation of Ground Water Using Natural Adsorbents", American Journal of Analytical Chemistry 2013, 4, 221-228.
- [16] Subarayan BG, Viswanathan Gopalan, Siva IS (2012), "Prevalence of fluorosis and identification of fluoride endemic areas in Manur block of Tirunelveli district, tamil Nadu, South India. Appl water Sci 2:235-243.
- [17] Apambire WB, Boyle Dr, Michel FA (1997), "Geochemistry, genesis and health implications of fluoriferous groundwaters in the upper regions of Ghana", Environ Geol 33:13-24.
- [18] Asha Gupta *et al.*, "Removal of fluoride by thermally activated carbon prepared from neem (*Azadirachta indica*) and kikar (*Acacia arabica*) leaves", Journal of Environmental Biology March 2008, 29(2) 227-232.
- [19] Jacks G, Rajagopalan K, Alveteg T, Jonsson M (1993), "Genesis of high-F groundwaters, Southern India", Appl Geochem 2:241-244.
- [20] Murray JJ (1996), "appropriate use of fluorides for human health", WHO, Geneva.
- [21] Susheela AK, Kumar A, Bhatnagar M, Bahadur R (1993), "Prevalence of endemic fluorosis with gastrointestinal manifestations in people living in North-Indian Villages", Fluoride 26:97-104.
- [22] Patil Satish, Renukdas *et al.*, "Defluoridation of water using bioadsorbents: Kinetic and Thermodynamic Study", International Journal of Research in Chemistry and Environment, (January 2013) Vol.3, Issue 1, pp.125-135.
- [23] V.Tomar, S. Prasad, D.Kumar, "Adsorptive removal of fluoride from water samples using Zr-Mn composite materials", Microchem J., 111 (2013) 116-124.
- [24] Shashikant R.Mise *et al.*, "Fluoride Removal from Water Using Activated Carbon Derived From Phoenix Dactylifera (Date Plum) Seeds", ISSN: 2348-4748, Volume 1, Issue 4, April 2014.
- [25] Test Methods for Activated Carbon according to European Council of Chemical Manufacturer's Federations (CEFIC).
- [26] Sutapa Chakrabarty and H.P.Sarma, "Defluoridation of contaminated drinking water using neem charcoal adsorbent: Kinetics and equilibrium studies", International Journals of Chem Tech Research. Vol.4, No.2, April-June 2012, 511-516.
- [27] WHO, "Fluorides and Oral Health," World Health Organisation Technical Report series 846, Geneva, 1994.
- [28] Sarfraz Ahmad Khan *et al.*, "Comparative Study of Defluoridation from Water using Waste Materials as Adsorbents", International Journal of Innovations in Engineering and Technology (IJJET) Volume 6 Issue 1-October-2015.
- [29] Meenakshi, Maheshwari R.C., "Fluoride in drinking water and its removal", Elsevier B.V, (16 February 2006).
- [30] R. X. Liu, J. L. Guo and H. X. Tang, "Adsorption of Fluoride, Phosphate, and Arsenate Ions on a New Type of Ion Exchange Fiber," Journal of Colloid and Interface Science, Vol. 248, No. 2, 2002, pp.268-274.
- [31] S.S. Tripathy *et al.*, "Removal of fluoride from drinking water by adsorption onto alum-impregnated activated alumina, Sep. Purif.



- Technol., 50 (2006) 310-317.
- [32] Abas Siti Nur Aeisyah *et al.*, "Adsorption Process of Heavy Metals by Low-Cost Adsorbent: A Review", World Applied Sciences Journal. 2013; 28(11):1518-1530.
- [33] Hugo Alberto Sánchez-Sánchez *et al.*, "Fluoride Removal from Aqueous Solutions by Mechanically Modified Guava Seeds", International Journal of Sciences: Basic and Applied Research (IJSBAR) (2013) Volume 11, No 1, pp 159-172.
- [34] Kaminsky *et al.*, (1990), "Fluoride: Benefits and Risk of Exposure", Oral Biology and Medicine.

