



## Removal of Fluoride in Polluted Water using Gular Plant (*Ficus recemosa*) Leaves as Bio-Adsorbent

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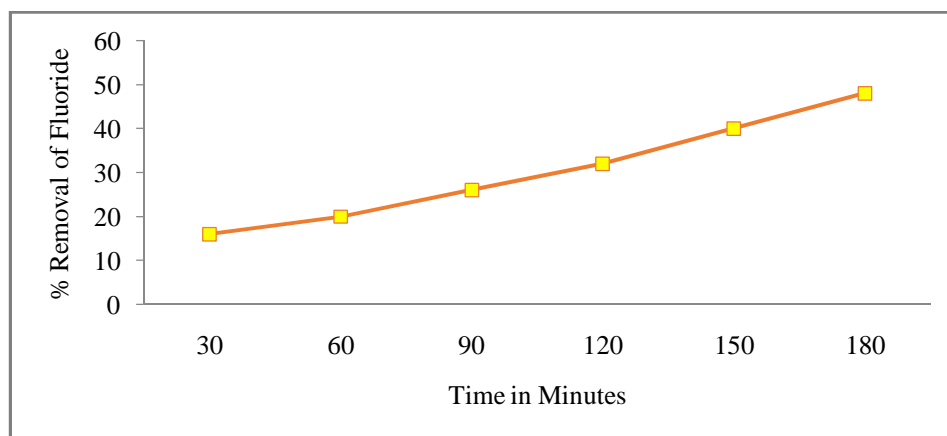
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### ABSTRACT

Fluoride is a poison associated with cancer in young men; Osteoporosis; Low I.Q; And hip fractures accumulate in our bones in the elderly, to name a few, the permissible limit of fluoride concentrations in drinking water is  $1.5 \text{ mg L}^{-1}$  according to WHO. Therefore, knowledge of using the best technology with maximum efficiency is required, removing its efficiency. Among various techniques, adsorption plays a major role in fluoride removal. As cost is an important consideration in most developing countries, efforts have been made to explore the possibility of using various low-cost advertisers that are abundant, readily available, and derived from waste materials. The "M. S. Swaminathan Research Foundation" (MSSRF) showed drumstick seeds to have significantly higher deflation capacity than active alumina. In the present work, the use of Gular Plant (*Ficus recemosa*) leaves as low cost adsorbents and their feasibility for the removal of fluoride from ground water is studied with various affecting parameters like different pH and contact time.

### Graphical Abstract



Defluoridation by Gular plant (*Ficus recemosa*)

**Keywords:** Adsorption, Bio-adsorbent, Efficiency, Defluoridation, Low Cost Adsorbents.

## INTRODUCTION

Water is contaminated when natural sources or industrial wastes are found in it. One such pollutant is fluoride. The natural presence of which usually occurs through the formation of fluorapatite, geochemical deposits, natural water systems, and soil in the form of earth's crust [1, 2]. In industrial processes Fluorine is used for the manufacturing or production of steel, aluminum, iron, glass, plastic and in refrigerants such as Freon [3–8]. Fluorine is also used for the production of phosphate fertilizers for agricultural use [3, 9–11]. In drinking water, fluoride must be within a range that varies slightly above and below  $1 \text{ mg L}^{-1}$  according to WHO standards [12]. In low water levels, fluoride levels are up to  $1.5 \text{ mg L}^{-1}$ . According to the Bureau of Indian Standards BIS (IS-10500) [13], the desirable limits and permissible limits of fluoride in drinking water are  $1.0$  and  $1.5 \text{ mg L}^{-1}$ , respectively. The entry of fluoride into the body is through water, food, industrial exposure, medicines, cosmetics etc. The major source of daily intake (75%) is drinking water. Fluoride is attracted to positively charged calcium in teeth and bones because it has strong electrical negativity. Dental fluorosis, tooth decay, skeletal fluorosis and bone deformity in children as well as adults are the major health problems caused by fluoride, and high concentration of fluoride also affects plants and animals. The effect on agriculture was also evident due to the inhibition of plant metabolism due to necrosis, needle scratching and tip-burning diseases. Also major signs of fluorosis were seen in animals. After studying in the areas around Zinc Smelter, Debari, Udaipur, Rajasthan, the concentration of fluoride in soil and vegetables was found [14]. When concentrations of fluoride are high, which increases the risk of miscarriage; [15] it has also been observed that high concentrations of fluoride have been studied in endemic areas of Iran, with increased blood pressure, body mass index, and waist circumference [16]. Physico-chemical study of some fluorosis-affected pediatric dental caries and their BMI value was done in Rajauli sub-division of Nawada, Bihar [17].

The adsorption process is considered to be one of the most effective methods compared to other techniques of removing fluoride from drinking water, its removal for fluoride based on initial cost, flexibility, and simplicity of design, ease of operation and maintenance. This technique depends mainly on the efficiency of the adsorbents [18]. It is reported that Detoxification (Removal of Trace Elements like Lead, Manganese) of Aqueous Waste Industrial Effluent Water by KAZA'S Carbons-Tools [19]. Adsorption-Remediation was observed of Fluoride by Municipal Solid Waste Ashes [20]. Activated KAZA's Carbons –was used for Defluoridation of Potable Water [21].

Adsorption relies on solid adsorption that is bounded by a solid surface with a weak intermolecular force.

The adsorption of fluoride on solid adsorbent usually occurs in three stages [18]

1. Diffusion or transport of fluoride ions from the bulk to the outer surface of the adsorbent  
Solution at the boundary layer around the adsorption particle called external mass transfer;
2. Adsorption of fluoride ions on particle surfaces.
3. Adsorbed fluoride ions possibly exchange with structural elements

Depending on the chemistry of the adsorbent solids, or the adsorbed fluoride ions are transferred to internal surfaces for porous material (intrauterine diffusion) [22]. It has been observed that Defluoridation of water can be worked with environmental wastes [23]. In the literature, defluoridation was studied using different biosorbents such as Cynodondactylon-based activated carbon [24], thermally activated carbon prepared from neem (*Azadirachta indica*) and kikar (*Acacia arabica*) leaves [25], carbonaceous adsorbents such as wood charcoal or bone charcoal, coconut coir or animal bones [26], metal ions impregnated activated charcoal [27], activated charcoal prepared from wheat husk- and alum-treated fly ash [28], bone charcoal, peels of *Citrus document*, *Citrus medica*, and *Citrus aurantifolia* fruits-based activated carbon [29], *Moringa indicia* bark, chitin/cellulose composite [30], chitosan-coated silica [31], corn cobs [32], rice husk [33], tamarind seed and *Moringa indicia*-based activated carbon [34, 35] and bonemeal [36]. Cost effective natural

adsorbents for the removal of fluoride: a green approach [37] adsorption of fluorides in drinking water by palm residues [38] removal of fluoride from ground water by using bio-adsorbents like *aegle marmelos* (bilvepatra) [39] removal of fluoride from ground water by using bio-adsorbent like *lantana camera* (jamri) [40] defluoridation of water by *Murraya koenigii* (curry leaves) a natural bio-adsorbent [41]. Adsorption of Fluoride from Aqueous Phase by *Bombax Malabaricum* Carbon (Kaza's Carbon)[42]. A study on removal of fluoride ions using *Aloe barbadensis* as a low-cost natural adsorbent [43].

The defluoridation techniques employed so far have been applied to adsorption, ion exchange, precipitation, electrochemical and membrane techniques depending on the nature of the processes. However these techniques are not widely used due to their main cost, inefficiency, or failure in large scale application. Therefore, the discovery of a suitable low cost and environmentally friendly method is important for the efficient removal of fluoride in drinking water. In this work the leaves of the Gular plant (*Ficus racemosa*) were used as bio-adsorption.



Figure 1. *Ficus racemosa* Linn fruits.

*Ficus racemosa* Linn (Family; Morasi) in the treatment of many disorders in traditional medicine is used. It is one of all the ancient scriptures mentioned in the herbs of Ayurveda, Siddha, Unani and Homeopathy. Various parts of plants such as bark, root, leaf, fruit and latex are used as astringent, carminative, vermifuge and anti-dysentery. Eating it in extreme hunger is a good remedy. Fruit extracts are used in diabetes, leucoderma and menorrhagia. It has been used locally for a long time to relieve inflammation in skin lesions, lymphadenitis, sprains and fibrositis.

It is found in the northern parts of India, its length is 30-50 feet. It has an auspicious status in religious festivals and worship. Ficus fruits have a diameter of 2 inches and are green in color when they are apical and are found in clusters. The period is from March to June. Ayurveda medicines are used in the tree as astringent, antidiuretic and leucorrhea and menstrual disorders. Ficus fruit is used in the treatment of anemia and gastrointestinal disorders [44]. Its leaves are rich in flavonoids, triterpenoids (originally lanosterol), alkaloids and tannins. A new triterpene namely gluanol acetate and desmosic acid was isolated from the same part [45].

## MATERIALS AND METHODS

**Preparation of Adsorbent Ficus tree:** *Ficus racemosa* (Gular Plant) leaves are easily available at a tree these are collect and washed with distilled water several times and then dried in sunlight for 3-4 days. Now grind this dry leaves with mixer and sieved with suitable mesh size (30 BSS size) taken for analysis.

**Measurement of Fluoride Removal:** Fluoride Ion Meter Panamax (Model PX/IMC/321) was used to observe fluoride Removal from water sample. First it is calibrate with 100 and 10 ppm solutions prepared by the stock solution. Plastic beaker is washed with tap water and then distilled water. The 100 mL of 5 ppm solution of water was taken in 250 mL plastic beaker and 1.0 g of adsorbent hold

for 30 min. After 30 min filter the solution take reading of this sample at different time intervals and using various parameters like pH, dose of adsorbent and contact time of adsorbent for the further observations.

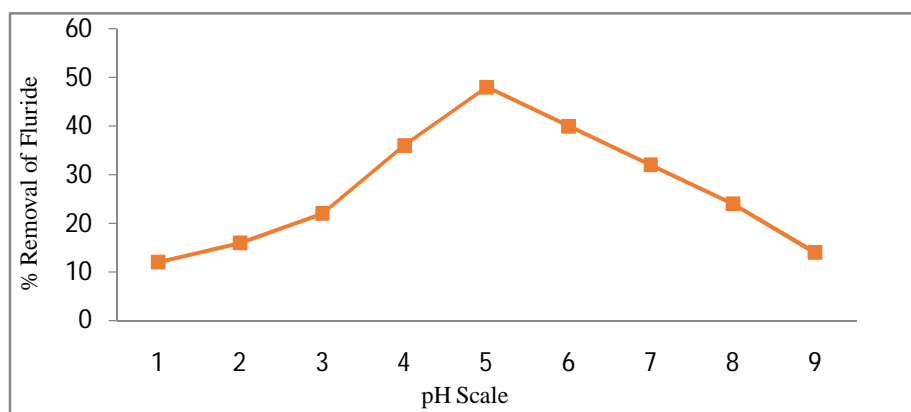
## RESULTS AND DISCUSSION

**Effect of pH:** 1 gm adsorbent is kept in 5 ppm fluoride solution of 100 mL water sample for 4 h. The change in pH is made from pH 2 to 10, it is observed that maximum fluoride removal was 48 percent at pH 5.5. The pH of the Fluoride solution was adjusted by the addition of 0.1(M) HCl and NaOH solutions as per requirements.

**Table 1.** Effect of pH

Time interval	pH-1.5	pH-2.5	pH-3.5	pH-4.5	pH-5.5	pH-6.5	pH-7.5	pH-8.5	pH-9.5
Initial	5	5	5	5	5	5	5	5	5
after 2 hour	4.4	4.2	3.9	3.2	2.6	3	3.4	3.8	4.3
ppm Removal	0.6	0.8	1.1	1.8	2.4	2	1.6	1.2	0.7
% Removal	12	16	22	36	48	40	32	24	14

1 gm adsorbent taken in 100 mL 5.0 ppm water



**Figure 2.** Effect of pH.

The pH can influence the surface charge of the adsorbent, the degree of ionization also the species of the adsorbate. The effect of pH on the removal of fluoride was studied in the range of 1.5-9.5 and results are illustrated in figure 2 pH plays an important role in the adsorption process on bio adsorbents. The results confirm a strong dependence between the adsorption of fluoride and pH, whereby adsorption appears to increase with increasing pH, within a pH range of 1-6. Maximum adsorption was observed at a pH of 5.5 for *Ficus recemosa* (Gular Plant) in 4 h.

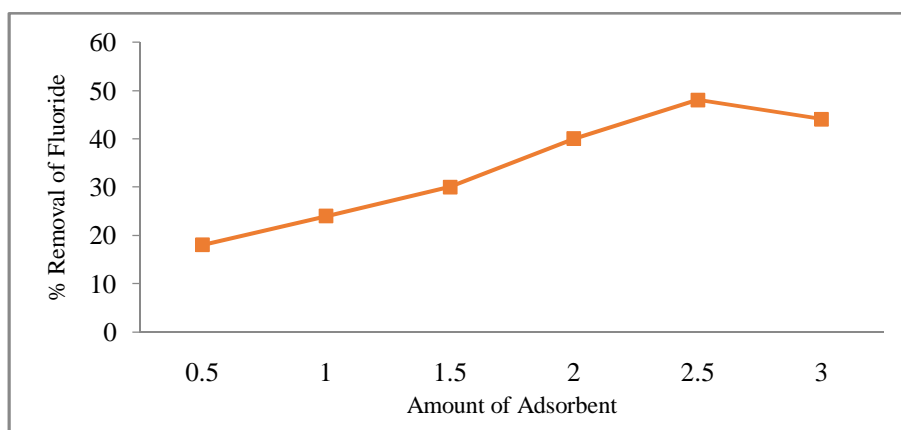
Where it shows that the overall charge on the surface of bio adsorbent is positive. Positive charge binds the negatively charged fluoride ions. In the case of *Ficus recemosa* (Gular Plant). When anionic exchange sorption occurs than the surface of the adsorbent gets positively charged and sorption of fluoride ion at the lower value of pH (< 7). Relative sorption inhibition occurred at basic pH (> 7) range, might be assigned to the increase of hydroxyl ion leading to the formation of aqua-complexes; thereby, desorption occurred. According to the study of pH optimization, adsorption of bio adsorbent mostly observed in the acidic range of the pH, which is more beneficial and cost-effective for the removal.

**Effect of Adsorbent Dose:** To investigate the effect of dose were conducted by varying adsorbent dose in the range of 0.25 g to 3.0 g L<sup>-1</sup> at constant initial fluoride concentration of 5 mg L<sup>-1</sup> and

optimum pH 5.5. For this purpose we take four sample of 0.25 g, 0.50 g, 1.5 g, 2.0g, 2.5 g and 3.0 g adsorbent in 100 mL 5 ppm fluoride solution and kept for 4 h. In this observation 48 % fluoride is removal at 2.5 g adsorbent dose.

**Table 2.** Amount of Adsorbent

S.No	Wt of Adsorbent (Gm)	Amount of Water 5.0 ppm ( ML)	Initial Conc. Before Treatment (ppm)	After 3.0 hrs (ppm)	ppm Removal	% Removal
1	0.5	100 ml	5.00	4.1	0.90	18
2	1	100 ml	5.00	3.8	1.20	24
3	1.5	100 ml	5.00	3.5	1.50	30
4	2	100 ml	5.00	3	2.00	40
5	2.5	100 ml	5.00	2.6	2.40	48
6	3	100 ml	5.00	2.8	2.20	44



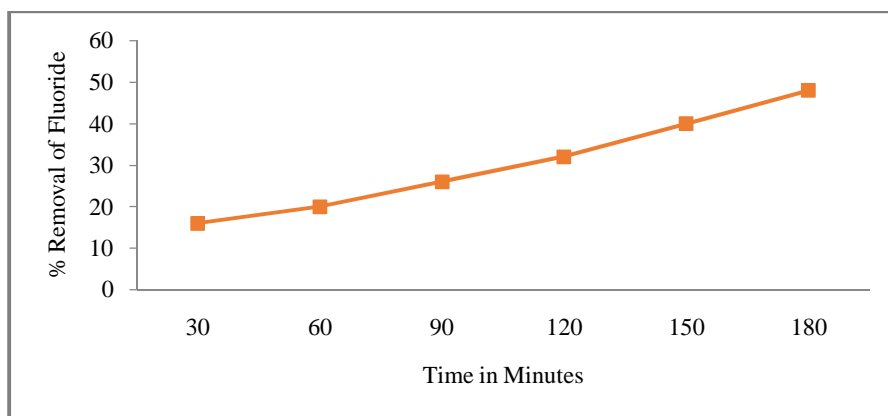
**Figure 3.** Effect of Adsorbent Dose.

The removal efficiency of fluoride is strongly dependent on the concentration of adsorbent dose in a test sample. Removal of fluoride increases as the increasing dose of adsorbent in the sample as shown in figure 3. At the starting, removal of fluoride increases as increasing the dose until some extent after that very slight change in the removal of fluoride it means, the curve indicating that the removal remains constant at the higher fluoride concentration occurs when their maximum dose. Adsorption increases after that adsorption of fluoride is constant at a higher dose because of saturation of pore volume and surface. Efficiency for *Ficus recemosa* (Gular Plant) leaves powder from 4.1 ppm to 2.6 ppm, for the dose range 0.25-3.0 g 100 mL<sup>-1</sup>.

**Effect of Contact Time:** To investigate the effect of contact time, experiments were conducted by varying adsorbent dose in the range of 0.25 to 3.0 g 100 mL<sup>-1</sup> at constant initial fluoride concentration of 5 mg L<sup>-1</sup>. The dosage range was selected based on adsorption test [46]. The residual fluoride concentrations were measured by taking samples at different contact time (30 to 180 min).

**Table 3.** Effect of Contact Time at 2.5 g

Dose g 100mL <sup>-1</sup>	Contact time (min.)	Initial Concentration	Final Fluoride (mg L <sup>-1</sup> ) ppm	Reduction of Fluoride	% Removal
2.5	30	5 ppm	4.2	0.8	16
2.5	60	5 ppm	4	1	20
2.5	90	5 ppm	3.7	1.3	26
2.5	120	5 ppm	3.4	1.6	32
2.5	150	5 ppm	3	2	40
2.5	180	5 ppm	2.6	2.4	48



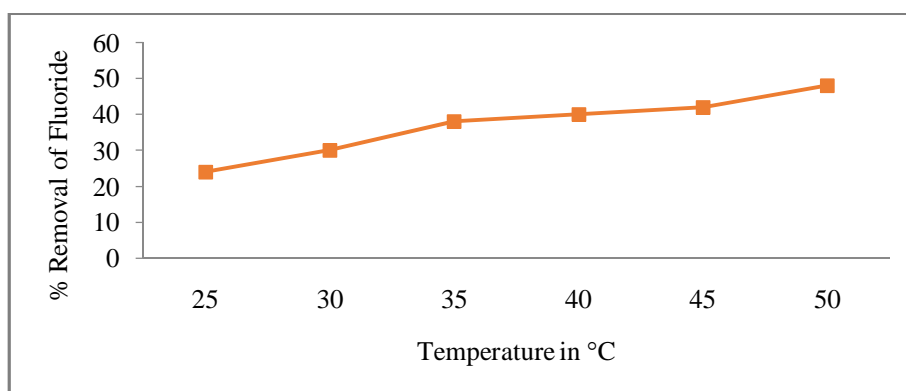
**Figure 4.** Effect of Contact Time at 2.2g.

It is found that at optimum pH and dose, the exclusion of fluoride ions increases with an increase in contact time to some level. Further increase in contact time does not increase the Fig-3.8 explains the optimum percentage removal of fluoride by three considered bio adsorbents at different contact times. The adsorption of the fluoride ion by adsorbent also depends on the interactions of the solution and the surface of the adsorbent. It is assumed that Adsorptions can be completed when equilibrium is achieved between the solute of the solution and the adsorbent.

**Effect of Temperature:** In this process 1 g adsorbent was placed in 100 mL of 5 ppm solution at 22°C, 30°C, 40°C and 50°C temperature for 3 h the maximum fluoride removal was 48% at 50°C.

**Table 4.** Effect of Temperature

Temperature °C	Initial Concentration	After 3 h	Removal amount	% Removal of Fluoride
25	5	3.8	1.2	24
30	5	3.5	1.5	30
35	5	3.1	1.9	38
40	5	3	2	40
45	5	2.9	2.1	42
50	5	2.6	2.4	48



**Figure 5.** Effect of temperature.

The influence of temperature on the removal of adsorption capacity of the fluoride ion was measured at different temperatures ranging from 25 to 50°C figures 5. Display the percent removal for fluoride ion increased with temperature, from 24 to 48% for fluoride ion. When temperature was increased from 25 to 50°C and removal almost reached equilibrium at 50°C and then almost constant. This explains why at very high temperature fluoride ion increase and then constant its adsorbent power through denaturation [47].

The defluoridation methods depend on hot climates, for sorption capacities attained under room temperature conditions may be higher than in the field as a result of increased temperatures [48]. Temperature can impact the physical binding processes of fluoride to a sorbent. However, temperature also can have a direct impact on the physical properties of a sorbent, if thermally treated prior to exposure, so that sorption capacities can be significantly altered.

**Effect of Mesh Size of Adsorbent:** Adsorbent was sieved with 30 BSS, 36 BSS, 52BSS, 60BSS Sieve. After this 1 g of Adsorbent was placed in 100 mL of 5 ppm. Fluoride solution for 4 h it gets maximum 48% removal of fluoride at 30 BSS Mesh size.

Table 5. Size of Adsorbent

Fluoride concentration	60 BSS	52 BSS	36 BSS	30 BSS
initial	5	5	5	5
after 3 hour	3.5	2.9	2.9	2.6
ppm Removal	1.5	2.1	2.1	2.4
% Removal	30	36	42	48

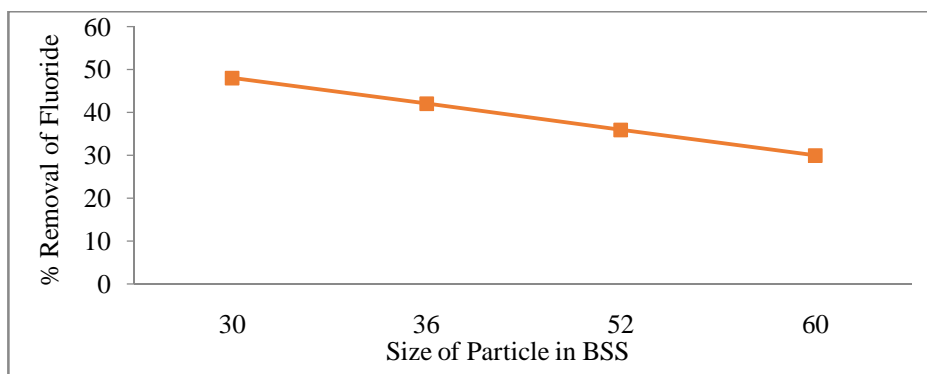


Figure 6. Effect of particle size.

### Isotherm Studies

**Adsorption isotherms:** Adsorption studies are necessary to adequately explain the adsorption process. Several models have been used for adsorption isotherms to describe experimental data. However, the most suitable for this study are the Langmuir and Freundlich isotherms models.

The parameters obtained from different models provide some important information on the adsorption mechanism as well as the affinity of the adsorbent. Linear regression is frequently used to determine the best fitting isotherm by judging its correlation coefficient. The parameters obtained from different models provide some important information on the adsorption mechanism as well as the affinity of the adsorbent. Linear regression is frequently used to determine the best fitting isotherm by judging its correlation coefficient.

**Langmuir isotherm:** As Reported by Langmuir adsorption isotherm occurs at homogenous sites and forms a monolayer. The values of separation factor RL can be summarized as shown in table 6.

$$RL = 1 + aLC_0$$

Table 6. Separation factor

Value of RL	Types of isotherm
RL > 1	Unfavorable
RL = 1	Linear
0 < RL < 1	Favorable
RL = 0	Irreversible

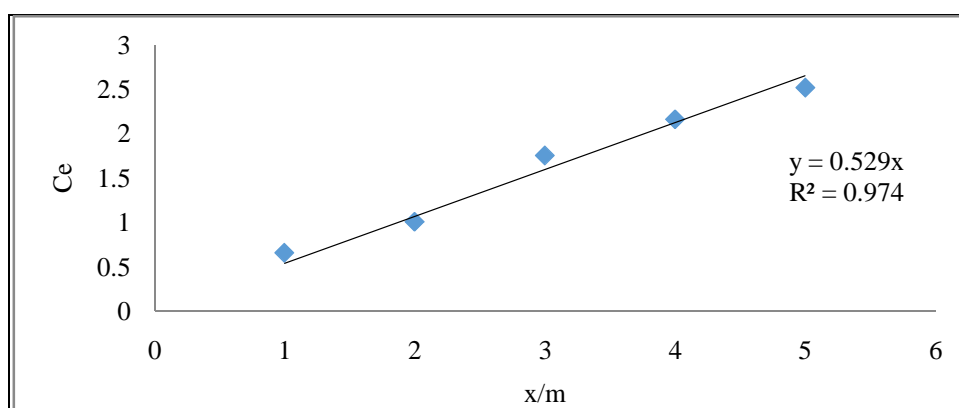
The equation for Langmuir isotherm is

$$1/q_e = 1/q_m \times K_L \times C_e + 1/q_m \quad \dots (1)$$

where  $q_e$  is amount of fluoride adsorbed per unit weight of adsorbate,  $C_e$  is unadsorbed fluoride concentration at equilibrium,  $q_m$  is maximum adsorption capacity and  $K_L$  is the Langmuir constant. A linear plot of  $1/q_e$  v/s  $1/C_e$  suggest the applicability of Langmuir isotherm (Figure 7).

**Table 7.** Langmuir isotherm

Ci (mg L <sup>-1</sup> )	Ce (mg L <sup>-1</sup> )	X (mg)	m (g)	X/m (mg g <sup>-1</sup> )	Ce / (X/m)	log Ce	log X/m
1	0.35	0.65	1.0	0.65	0.535	-0.4559	-0.187
2	1	1	1.0	0	1.000	0	0
3	1.25	1.75	1.0	1.75	0.714	0.09669	0.203
4	1.84	2.16	1.0	2.16	0.852	0.2648	0.3344
5	2.48	2.52	1.0	2.52	0.984	0.3944	0.4014



**Figure 7.** Langmuir isotherm.

The  $R^2$  values of linear correlation plot of  $1/q_e$  v/s  $1/C_e$  is **0.974**, which suggest that the Langmuir isotherm provides a good model for this adsorption system.

**Freundlich Isotherm:** The Freundlich isotherm gives the relationship between equilibrium liquid and solid phase capacity based on the multilayer adsorption properties consisting of the heterogeneous surface of the adsorbent.

The Freundlich equation is based on adsorption on a heterogeneous surface and it is given as-

$$q_e = KF \times C_e^{1/n} \quad \dots (2)$$

It can be written in a linear form as-

$$\log q_e = KF + 1/n \log C_e \quad \dots (3)$$

Here, KF is Freundlich constant and n is the adsorption intensity of the dye onto adsorbent or surface heterogeneity. A linear plot of  $q_e$  v/s  $\log C_e$  suggests that Freundlich isotherm is also applicable in this case (Figure 8).



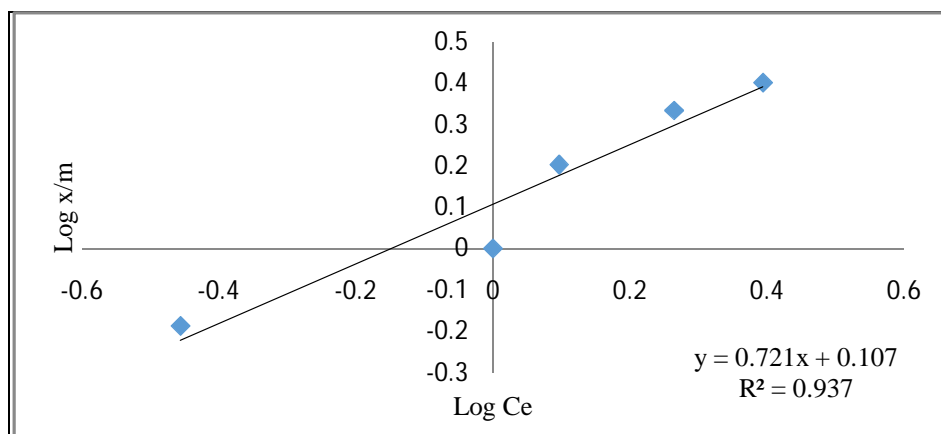


Figure 8. Freundlich adsorption isotherm.

### Kinetic Study

**Adsorption kinetic studies:** A study of chemical kinetics is very important for any adsorption system to determine the rate constants for a reaction and to determine how quickly or slowly the reaction is proceeding. Thus it is revealed under this study system that is more aptly described by the pseudo-second-order kinetics in figure 7 which was based on the assumption that the rate-limiting step may be physisorption due to the presence of weak forces of attraction between adsorbent and adsorbate.

Table 8. Adsorption kinetic studies

Contact time (min)	Reduction of fluoride
0	0.7
30	0.9
60	1.2
90	1.5
120	1.8
150	2.2
180	2.6

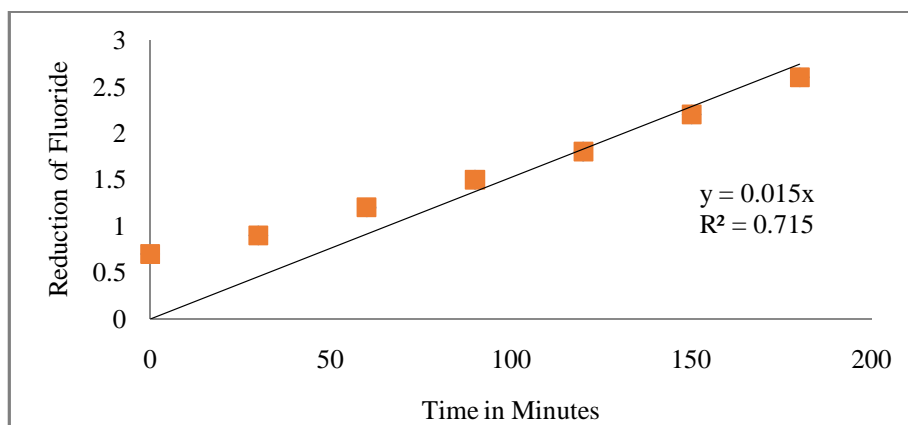


Figure 9. Amount of Adsorbent.

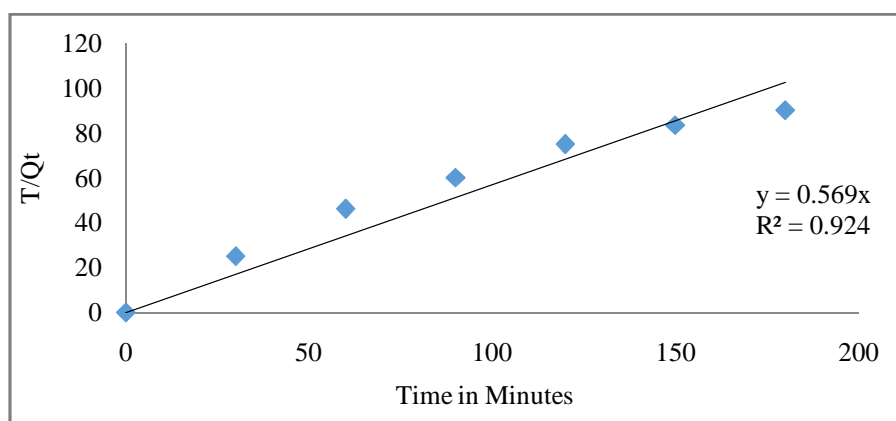
**Pseudo-First Order:** The adsorption kinetics data can be described by pseudo-first order kinetic equation. The linear form of this equation is-  $\log (q_e - q_t) = \log q_e - k_1 \times t$  Where,  $q_e$  and  $q_t$  are amounts adsorbed at equilibrium and at time  $t$ , respectively.  $k_1$  is the rate constant of pseudo-first order equation.

**Pseudo-Second Order:** The kinetics can also be described by pseudo-second order model. The linear form of pseudo-second order is expressed as  $-t/q_t = 1/k_2/q_e^2 + 1/q_e \times t$  Where,  $k_2$  is pseudo-second order constant. A plot of  $t/q_t$  against  $t$  gives a linear relationship.  $q_e$  and  $k_2$  can be determined from the slope and intercept of this plot.

The values of  $R^2$  in plots (Figure 9 and 10) are 0.95 and 0.998 for pseudo-first order and pseudo-second order, respectively, which indicates that this adsorption process followed both the kinetic models, but pseudo-second order model was more suitable than the pseudo-first model.

**Table 9. Pseudo second order parameter**

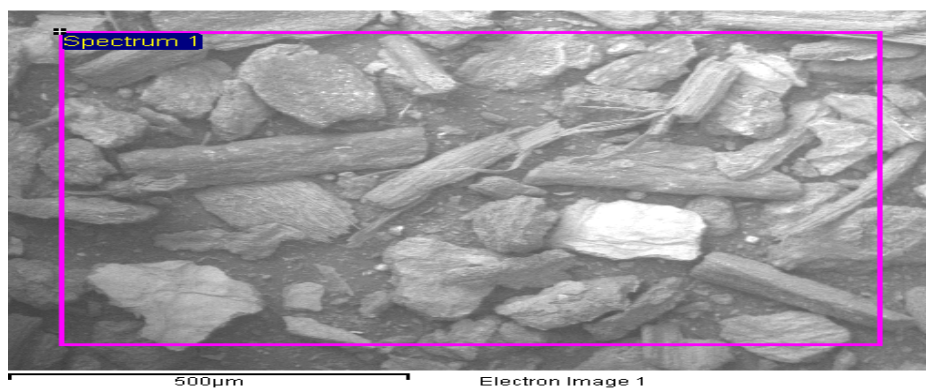
Contact time (min)	T/Q <sub>t</sub>
0	0
30	25
60	46.15
90	60
120	75
150	83.33
180	90

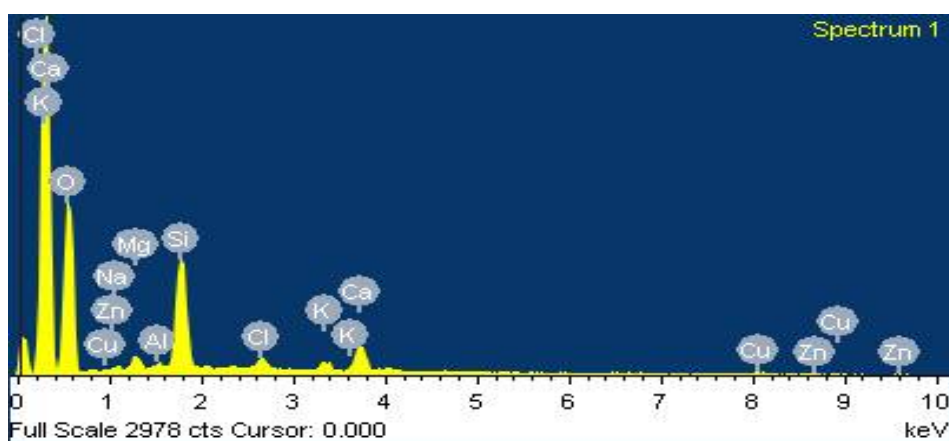


**Figure 10. T/Q<sub>t</sub> V/s Time.**

It is second order because  $R^2$  value is near to one (**0.924**)

**EDX Analysis:** EDX analysis of the sample of Gular Plant Leaves (*Ficus recemosa*) powder after treated with fluoride water attached to this file in figure form.

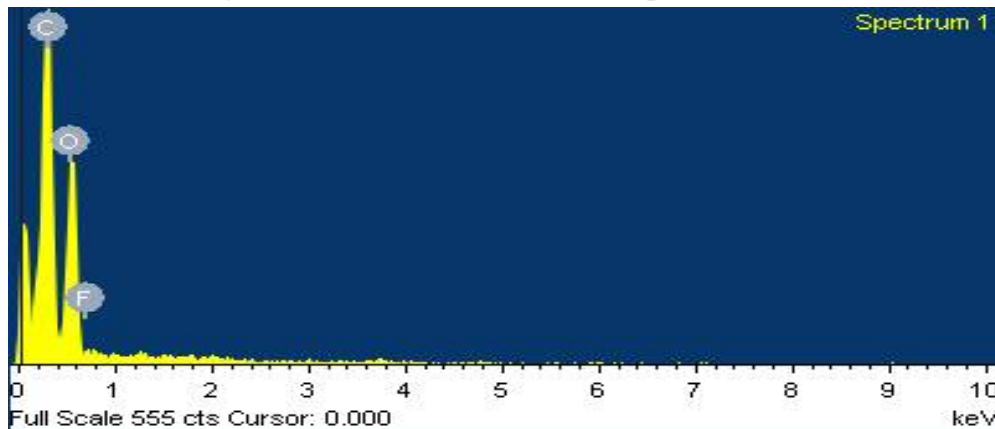
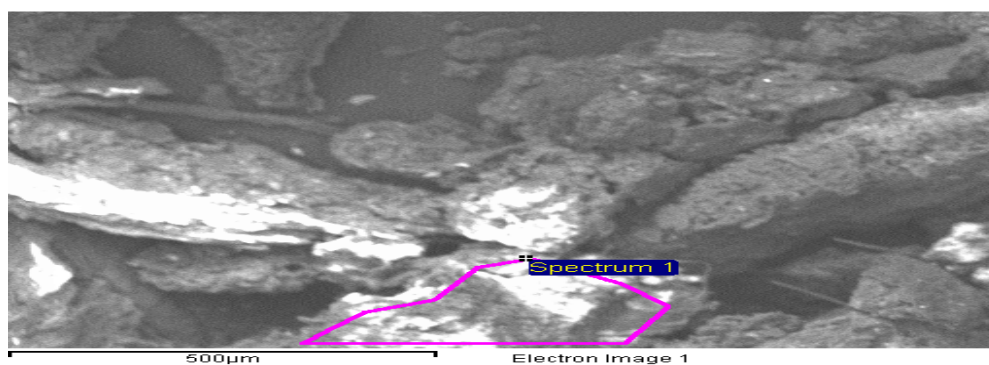




Spectrum processing:  
No peaks omitted  
Processing option : All elements analyzed (Normalized)  
Number of iterations =6

Element	Weight%	Atomic%
O	69.95	82.04
Na	1.11	0.91
Mg	2.35	1.82
Al	0.56	0.39
Si	15.11	10.10
Cl	1.83	0.97
K	1.59	0.76
Ca	4.75	2.23
Cu	1.34	0.39
Zn	1.40	0.40
Totals	100.00	

Figure 11. Before Treatment.



Processing option : All elements analyzed (Normalised)		
Number of iterations =4		
Element	Weight%	Atomic%
C	48.22	55.37
O	51.72	44.59
F	0.06	0.04
Totals	100.00	

Figure 12. After treatment

## APPLICATION

Fluorosis is an important public health problem in India. This technique is very useful for removing fluoride from ground water. A large ability to remove fluoride represents an overview of the various low-cost advertisers presented here. Use of commercially available adsorbents can be replaced by inexpensive and effective low-cost adsorbents. More studies are needed to better understand the process of low-cost adsorption and to demonstrate the technology effectively. In the bio-adsorption process we will also make nano-particles of bio-adsorption / waste material to defluoridation of drinking water.

## CONCLUSION

This research work briefly highlighted the importance of adsorption process and its benefits. Also the overview of various papers publishes in various journals on removal of fluoride ions from water or wastewater by adsorption using various low cost adsorbents instead of expensive commercial adsorbents. The ability of different adsorbents in the removal of fluoride depends on dose of adsorbate, characteristics of adsorbent, pH, temperature, contact time etc., some low cost adsorbents presented here shows a good efficiency for the fluoride removal. The commercially available adsorbents can be replaced by the cheap and effective low cost adsorbents.

## REFERENCES

- [1]. M. Bishnoi and, S. Arora, Potable groundwater quality in some villages of Haryana, India: focus on fluoride, *J. Environ. Biol.*, **2007**, 28(2), 291-295.
- [2]. K. Shailaja, M. E. C. Johnson, Fluorides in groundwater and its impact on health, *J. Environ. & Biol.*, **2007**, 28(2), 331-333.
- [3]. A. Quintáns-Fondo, G. Ferreira-Coelho, R. Paradelo-Núñez, J. C. Nóvoa-Muñoz, M. Arias-Estévez, M. J. Fernández-Sanjurjo, E. Álvarez-Rodríguez, A. Núñez-Delgado, Promoting sustainability in the mussel industry: Mussel shell recycling to fight fluoride, *pollution. J. Clean. Prod.*, **2016**, 131, 485-490.
- [4]. J. Yang, Wang, M. Lu, J. Yang, K. Wang, K. Liu, M. Luo, H. Pang, L. Wang, B. Fluorine in the environment in an endemic fluorosis area in Southwest, *China Environ. Res.*, **2020**, 184, 109300.
- [5]. I. van der Veen, A. C. Hanning, A. Stare, P. E. G. Leonards, J. de Boer, J. M. Weiss, The effect of weathering on per- and polyfluoroalkyl substances (PFASs) from durable water repellent (DWR) clothing, *Chemosphere.*, **2020**, 249, 126100.
- [6]. S. Sorlini, M. C. Collivignarelli, M. Carnevale Miino, Technologies for the control of emerging contaminants in drinking water treatment plants, *Environ. Eng. Manage. J.*, **2019**, 18, 2203-2216.
- [7]. D. Barpaga, V. T. Nguyen, B. K. Medasani, S. Chatterjee, B. P. McGrail, R. K. Motkuri, L. X. Dang, Insight into Fluorocarbon Adsorption in Metal-Organic Frameworks via Experiments and Molecular Simulations, *Sci. Rep.*, **2019**, 9, 10289-10299.

- [8]. S. L. Choubisa, D. Choubisa, recover Status of industrial fluoride pollution and its diverse adverse health effects in man and domestic animals in India, *Environ. Sci. Pollut. Res.*, **2016**, 23, 7244–7254.
- [9]. A. Y. Bagastyo, A. D. Anggrainy, C. S. Nindita, Warmadewanthi, Electrodialytic removal of fluoride and calcium ions to phosphate from fertilizer industry wastewater, *Sustain. Environ. Res.*, **2017**, 27, 230–237.
- [10]. G. Dartan, F. Taspinar, I. Toroz, Analysis of fluoride pollution from fertilizer industry and phosphogypsum piles in agricultural area, *J. Ind. Pollut. Control.*, **2017**, 33, 662–669.
- [11]. Y. Yu, S. Cui, R. Fan, Y. Fu, Y. Liao, J. Yang, Distribution and superposed health risk assessment of fluorine co-effect in phosphorous chemical industrial and agricultural source, *Environ. Pollut.*, **2020**, 262, 114249.
- [12]. R. C. Meenakshi, Fluoride in Drinking Water and its Removal, Center for Rural Development and Technology, IIT, Delhi, **2006**.
- [13]. BIS 10500, Indian Standard Drinking Water Specification, Bureau of Indian Standards, New Delhi, **1991**.
- [14]. N. Bhat, S. Jain, K. Asawa, M. Tak, K. Shinde, A. Singh, V. V. Gupta, Assessment of fluoride concentration of soil and vegetables in vicinity of zinc smelter, Debari, Udaipur, Rajasthan, *Journal of clinical and diagnostic research.*, **2015**, 9(10), ZC63-ZC-66.
- [15]. V. Kazemi Moghadam, M. Yousefi, A. Khosravi, M. Yaseri, A. H. Mahvi, M. Hadei, A. A. Mohammadi, Z. Robati, A. Mokamel, High concentration of fluoride can be increased risk of abortion, *Biol. Trace Elem. Res.*, **2018**, 185, 262–265.
- [16]. M. Yousefi, M. Yaseri, R. Nabizadeh, E. Hooshmand, M. Jalilzadeh, A.H. Mahvi, Association of hypertension, body mass index, and waist circumference with fluoride intake; water drinking in residents of fluoride endemic areas, *Iran Biol. Trace Elem. Res.*, **2018**, 185, 282-288.
- [17]. A. Kumar, V. K. Singh, K. K. Singh, B. Singh, A. K. Nag, Physico-Chemical Study of Some Fluorosis Affected Child Dental Caries And Their Bmi Value In Rajauli Sub-Division of Nawada District of Bihar, *J. Applicable Chem.*, **2016**, 5 (4), 816-825.
- [18]. S. M. Habuda, R. M. Ergovic, F. Andrew, A Review on Adsorption of Fluoride from Aqueous Solution, *Materials*, **2014**, 7, 6317-6366.
- [19]. K. Somasekhara Rao, KAZA'S Carbons- Tools of Detoxification (Removal of Trace Elements like Lead, Manganese) of Aqueous Waste Industrial Effluent Water, *J. Applicable Chem.*, **2016**, 5 (6), 1251-1255.
- [20]. D. P. Chary, Adsorption-Remediation of Fluoride By Municipal Solid Waste Ashes, *J. Applicable Chem.*, **2013**, 2 (6), 1641-1647.
- [21]. Kaza. S. Rao, B. V. Rajeswara Reddy, Ch. Chakrapani, Activated KAZA's Carbons-Defluoridation of Potable Water, Lap Lambert Academic Publishing, GmbH & Co, Germany, USA, UK, **2011**
- [22]. S. Ligia, C. Carolina, C. Cristina, Fluoride Removal from Aqueous Solutions by Sorbtion-Flotation *U.P.B. Sci. Bull., Series B.*, **2012**, 74(4), 87-102.
- [23]. B. R. Gudipudi, S. Asif, B. Kumar and V. Rajesh-Defluoridation of water with environmental waste materials, *International Journal of Civil Engineering and Technology.*, **2019**, 10, 01, 2446–2452.
- [24]. G. Alagumuthu, V. Veeraputhiran, R. Venkataraman, Fluoride Sorption Using Cynodon Dactylon-Based Activated Carbon, *Hem. Ind.*, **2011**, 65(1), 23-35.
- [25]. S. Kumar, A. Gupta, J. P. Yadav, Removal of fluoride by thermally activated carbon prepared from neem (*Azadirachta indica*) and kinar (*Acacia arabica*) leaves, *J. Environ. & Biol.*, **2008**, 29(2), 227-232.
- [26]. A. Sivasamy, K. P. Singh, D. Mohan, M. Maruthamuthu, Studies on defluoridation of water by coal-based sorbents, *J. Chem. Technol. Biot.*, **2001**, 76, 7, 717-722.
- [27]. C. Janardhana, G. Nageswara Rao, R. Sai Satish, V. Sai Lakshman, Study on Defluoridation of Drinking Water by Impregnation of metal Ion in Activated Charcol, *Indian J. Chem. Techn.*, **2006**, 13, 414-416.

- [28]. R. P. Singh, Y. Singh, D. Swaroop, Defluoridation of Groundwater in Agra City Using Low Cost Adsorbents, *Bull. Environ. Contam. Toxicology*, **2000**, 65, 120-125.
- [29]. C. H. Chakrapani, S. Babu, K. N. K. Vani, Kaza. S. Rao, Adsorption kinetics for the removal of fluoride from aqueous solution by activated carbon adsorbents derived from the peels of selected citrus fruits, *E-J Chem.*, **2010**, 7, 419-427.
- [30]. G. Jayapriya, R. Ramya, X. R. Rathinam, P. N. Sudha, Equilibrium and kinetic studies of fluoride adsorption by chitin/cellulose composite, *Arch. Appl. Sci. Res.*, **2011**, 3(3), 415-423.
- [31]. Y. Vijaya, A. Krishnaiah, Sorptive response profile of chitosan coated silica in the defluoridation of aqueous solution, *E-J Chem.*, **2009**, 6(3), 713-724.
- [32]. S. P. Hemant, B. P. Jignesh, P. Sudhakar, V. J. Koshy, Removal of fluoride from water with powdered corn cobs, *J. Environ. Sci. Eng.*, **2006**, 48(2), 135-138.
- [33]. S. D. Waheed, S. J. Attar, M. D. Waghmare, Investigation on sorption of fluoride in water using rice husk as an adsorbent, *Nat. Environ. Pollut. Tech.*, **2009**, 8(2), 217-223.
- [34]. M. Murugan, E. Subramanian, Studies on Defluoridation of Water by Tamarind Seed, an Unconventional Biosorbent, *J. Water Health*, **2007**, 4(4), 453-461.
- [35]. G. Karthikeyan, S. S. Ilango, Fluoride sorption using *Moringa indica*-based activated carbon, *Iran J. Environ. Health Sci. Eng.*, **2007**, 4(1), 21-28.
- [36]. S. Gao, J. Cui, Z. Wei, Study on the fluoride adsorption of various apatite materials in aqueous solution, *J. Fluor. Chem.*, **2009**, 130, 1035-1041.
- [37]. S. Tamrakar, R. Verma, S. Kumar Sarand, C. Verma, Cost Effective Natural Adsorbents for the Removal of Fluoride: A Green Approach, *Rasayn J. Chem.*, **2019**, 12(2), 455-463.
- [38]. M. C. Collivignarelli, A. Abbà, M. C. Miino, V. Torretta, E. C. Rada, F. M. Caccamo, S. Sorlini, Adsorption of Fluorides in Drinking Water by Palm Residues, *Sustainability*, **2020**, 12, 3786-3798.
- [39]. A. Sharma, S. benjamin, D. Soni, R. Ameta, P. Tak, Removal of Fluoride from Ground Water by Using Bio-Adsorbents Like Aegle Marmelos (Bilve Patra), *International Journal of Current Advanced Research.*, **2018**, 7, 1(H), 9231-9235.
- [40]. A. Sharma, R. Ameta, S. Benjamin, D. Soni, S. Sharma, P. Tak, Removal of Fluoride from Ground Water by Using Bio-Adsorbent like *lantana camera* (Jamri), *International Journal of Science and Research.*, **2017**, 3, 442-446.
- [41]. Akshay Sharma, Sunder Lal Meena, Surbhi Benjamin, Dipti Soni, Rakshit Ameta, Paras Tak, Defluoridation of Water by *Murraya koenigii* (curry leaves) -A Natural Bio-Adsorbent, *Int. J. Pure Appl. Chem.*, **2018**, 13(3-4), 192-203.
- [42]. K. A. Emmanuel, A. Veerabhadrarao, T. V. Nagalakshmi, Ch. Sureshbabu, K. S. Rao, Adsorption of Fluoride from Aqueous Phase by Bombax Malabaricum Carbon (Kaza's Carbon), *J. Applicable Chem.*, **2015**, 4(1), 166-177.
- [43]. S. Rayappan, B. Jeyaprabha, P. Prakash, A study on removal of fluoride ions using *Aloe Barbadensis* as a low- cost natural adsorbent, *J. Applicable Chem.*, **2014**, 3(3), 1189-1201.
- [44]. C. V. Rao, A. R. Verma, M. V. Kumar, S. Rastogi, *J. Ethnopharmacol.*, **2008**, 115, (2)323-326.
- Title missed**
- [45]. A. R. Shiksharathi, S. Mittal, *Ficus racemosa*: Phytochemistry, traditional uses, and pharmacological properties: A review. *Int. J. Adv. Pharm. Res.*, **2011**, 4, 6-15.
- [46]. B. Shimelis, F. Zewge, B. S. Chandravanshi, Removal of excess flouride from water by aluminium hydroxide, *Bull. Chem. Soc. Ethiop.*, **2006**, 20 (1), 17-34.
- [47]. K. Sreenivasa, B. Narashimamurthy, K. N. Chandrashekara, Effect of pH and Temperature on defluoridation of drinking water by using activated alumina based adsorption, *IOSR J. Applied Chemistry*, **2015**, 8(7), 56-59.
- [48]. R. Kannan, M. Rajasimman, N. Rajamohan, B. Sivaprakash, Equilibrium and kinetic studies on sorption of malachite green using *Hydrilla verticillata* biomass, *International J. Environmental Research*, **2010**, 4(4), 817-824.