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Fluoride removal from groundwater using *Lagerfstroemia indica l.* Seed powder

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ABSTRACT

This study mainly dealt with removal of fluoride from groundwater sources by using crape myrtle *(Lagerstroemia indica* L.) seed powder. The removal of fluoride from groundwater is done against the process parameters particle size, adsorbent dosage, agitation speed and temperature. The process parameter range was done by Design of experiment approach. The result of the batch study indicated that the maximum removal fluoride by *Lagerstroemia indica* L. seed powder was obtained as 94.3 % (Run 4). This was achieved for the combination of process parameters particle size of 600 µm, adsorbent dosage of 4 g, agitation speed of 400 rpm and 60°C. Further, the maximum removal fluoride was obtained for the contact time of 60 min. The study was extended to fit the experimental data into isotherm models. The model result showed that the Langmuir isotherm model was fitted well with the experimental data on removal of fluoride in a groundwater. Thus, this study concluded that *Lagerstroemia indica* L. seed powder is used effectively for the removal of fluoride from groundwater.

KEY WORDS: *Lagerstroemia indica*, Fluoride, Groundwater, DOE, Process parameters

1. INTRODUCTION

The fluoride occurs mainly as sellaite (MgF_2), fluorspar (CaF_2), cryolite (Na_3AlF_6) and luorapatite [$3Ca_3(PO_4)_2$ $Ca(F,Cl_2)$]. As fluorsparit is found in sedimentary rocks and as cryolite in igneous rocks. These fluoride minerals are nearly insoluble in water. Hence fluorides will be present in groundwater only when conditions favour their dissolution or high fluoride containing effluents are discharged to the water bodies from industries.

Fluoride displaces hydroxide ions from hydroxyapatite, $Ca_5(PO_4)_3OH$, the principal mineral constituent of teeth (in particular the enamel) and bones, to form the harder and tougher fluoroapatite, $Ca_5(PO_4)_3F$. Up to a small level (1–1.5 mg/L), this strengthens the enamel. However, fluoroapatite is an order of magnitude less soluble than hydroxyapatite, and at high fluoride concentration (1.5–4 mg/L), the conversion of a large amount of the hydroxyapatite into fluoroapatite makes the teeth and (after prolonged exposure) the bones denser, harder and more brittle. In the teeth this causes mottling and embrittlement, a condition known as dental fluorosis. With prolonged exposure at higher fluoride concentrations (4–10 mg/L) dental fluorosis progresses to skeletal fluorosis.

Fluoride is thus considered beneficial in drinking water at levels of about 0.7 mg/L but harmful once it exceeds 1.5 mg/L, which is the World Health Organisation limit being followed in most of the Nations (WHO, 2011) and is also the Australian recommended limit (NHMRC, 2004). The difference between desirable doses and toxic doses of fluoride is ill-defined, and fluoride may therefore be considered as an essential mineral with a narrow margin of safety (WHO, 2011).

There are several studies carried out (Mohapatra, 2011; Akbar Eskandarpour, 2008) to provide precise information on the field of fluoride removal for drinking water using various conventional methods. Earlier works revealed the suitability of variety of agro-based materials like *moringa oleifera* seed, corncob, groundnut husk, rice husk, tea leaves carbon, saw dust to treat the industrial wastewater. The suggested treatment methods for the removal of pollutants from various dairy industries are adsorption (Sivasankar, 2010; Mansour, 2011; Sivakumar and Shankar, 2012; Sivakumar, 2013b, Sivakumar, 2014c, Sivakumar, 2014f, Sivakumar, 2014g, Sivakumar, 2014k; Shankar, 2014a; Ji-Lai Gong, 2015; Sivakumar and Nouri, 2015), ion exchange, chemical precipitation, bioremediation (Shankar, 2014b; Sivakumar, 2014d, Sivakumar, 2014e), constructed wetland (Sivakumar, 2015; Turker, 2014; Sivakumar, 2013a, Sivakumar, 2014h; Ingole and Bhole, 2003; Soltan and Rashed, 2003) and electro-dialysis (Sivakumar, 2014i, Sivakumar, 2014j) etc.

The cheap and economic alternative method is adsorption technique. Adsorption deals with various adsorbents such as: alumina/aluminium based materials, clays and soils, calcium based minerals, synthetic compounds, agro based materials and carbon based materials. This study is focused on removal of fluoride from groundwater using *Lagerstroemia indica L*. seed powder. The efficiency of removal has been discussed with the help of design of experiment approach. The adsorption study is also extended to isotherm points of view.

2. MATERIALS AND METHODS

The objective in fluoride removal is to treat the high level fluoride in groundwater to bring down fluoride levels into acceptable limit as per BIS standard.

2.1. Study Area: Dharmapuri district, which came into existence from 02.10.1965 is situated in the North western Corner of Tamil Nadu and is bounded by Tiruvannamalai and Villupuram Districts on the east, Salem District on the South, Krishnagiri District on the north and Kaveri river on the west. It is located between latitudes N 11° 47' and 12° 33' and longitudes E 77° 02' and 78° 40'. The climate of the Dharmapuri District is generally warm. The hottest period of the year is generally from the months of March to May, the highest temperature going up to 38 °C in April. The Climate becomes cool in December and continuous so up to February, touching a minimum of 17 °C in January. On an average the District receives an annual rainfall of 895.56 mm. The Soil type ranges from black to mixed loam; Red sandy soils are seen in Harur Taluk. Black and loam soil are found in Dharmapuri Taluk.

2.2. Collection of Water Samples and Analysis: To know the exact conditions of the groundwater collected from Dharmapuri, it is very much essential to go for water sampling and testing for the various parameters such as like pH was measured with the help of pH meter, electrical conductivity (EC) was measured with the help of an electrical conductivity meter which in turn used to calculate the TDS, anions like Ca, Mg, Na and K and cations like HCO_3, Cl_2, SO_4, and NO_3 and F were measured as per the standard procedure stipulated by APHA, AWWA, and WEF, 2005.

The values of these physico-chemical parameters obtained from groundwater of different areas are used to determine the suitability of groundwater for drinking purpose. But, this study was mainly focused on removal of fluoride from the groundwater rather than other parameters. In order to know the fluoride removal using the *Lagerstroemia indica L*. seed powder, groundwater was collected from Dharmapuri, Tamil Nadu with the help of air tight sterilized bottles, took to the laboratory and then they were stored at 4°C to avoid contamination and do the analysis in later stage. The initial concentration of fluoride was found to be 3.36 mg/L with the pH of 6.8.

2.3. Preparation of Adsorbent: Material used for the removal of fluoride is *Lagerstroemia indica L*. seed powder. The adsorbent was collected by grinding the *Lagerstroemia indica L*. seed for the experimental purpose. *Lagerstroemia indica L*. crape myrtle is a family of Lythraceae. It is having more adaptable, highly ornamental and long flowering, *Lagerstroemia indica L.* is an excellent choice for parks and gardens, around buildings, carparks and shopping areas, since, the tree size is small and hence, it increases the landscape value. The height is around 6m and having width around 4m.

The main habit of *Lagerstroemia indica L* is upright, open, multi-stemmed small tree. The foliage of *Lagerstroemia indica L* is leathery, dark green leaves turning yellowish-orange in autumn. The colour of flower is mid-lavender flowers in panicles mid-summer to mid-autumn. The fruit is insignificant and bark is sinuous bark, exfoliating (after about three years) to a light silvery grey under bark. It can adaptable to a range of conditions, heat and some dryness but requires adequate moisture during establishment. It can tolerate to best in moist, well drained, slightly acidic soils in a position receiving full sun. Transplants easily provided adequate moisture levels are maintained. It will help to promote spring growth by allowing the plant to first establish a strong root system.

2.4. Adsorption Experiment: The Magnetic stirrer apparatus was used for evaluating and optimizing the fluoride removal process. This method consists of batch experiments involving different controlling parameters like particle size, adsorbent dosage, agitation speed and temperature. The 100 ml of groundwater sample is taken in a 250 ml conical flask, and then placed on a magnetic stirrer for reducing the fluoride in a groundwater sample against the different process parameters. The experiments were performed at different particle size (600, 500, 425, 300 µm), adsorbent dosage (1, 2, 3, and 4g), agitation speed (100, 200, 300, and 400 rpm) and temperature (30, 40, 50, and 60 °C).

2.5. Design of Experiment: This method is used to reduce the numbers of experiments for the selected process parameters. According to this method, the selected numbers of experiments for this present study in only 16 (selected process parameter are 4 and selected levels are 4) instead of doing 256 experiments. All the experiments were carried out under natural conditions. The selected experimental runs are represented by 1 to 16 for 16 experiments and are presented in Table 1.

Table.1.Design of Experimental Runs (Orthogonal array)

Runs	Particle size, µm	Adsorbent dosage, g	Agitation speed, rpm	Temperature, °C
1	1	1	1	1
2	1	2	2	2
3	1	3	3	3
4	1	4	4	4
5	2	1	2	3
6	2	2	1	4
7	2	3	4	1
8	2	4	3	2
9	3	1	3	4
10	3	2	4	3
11	3	3	1	2
12	3	4	2	1
13	4	1	4	2
14	4	2	3	1
15	4	3	2	4
16	4	4	1	3

3. RESULTS AND DISCUSSION

The experiments were performed at different particle size (600, 500, 425, 300 µm), adsorbent dosage (1, 2, 3, and 4 g), agitation speed (100, 200, 300, and 400 rpm) and temperature (30, 40, 50, and 60 °C) for removing fluoride from groundwater, Dharmapuri. The selected experimental runs for the process parameters are represented by 1 to 16 and are presented in Table 2.

Table.2.Design of Experimental Runs for Process Parameters

Runs	Particle size, µm	Adsorbent dosage, g	Agitation speed, rpm	Temperature, °C
1	600	1	100	30
2	600	2	200	40
3	600	3	300	50
4	600	4	400	60
5	500	1	200	50
6	500	2	100	60
7	500	3	400	30
8	500	4	300	40
9	425	1	300	60
10	425	2	400	50
11	425	3	100	40
12	425	4	200	30
13	300	1	400	40
14	300	2	300	30
15	300	3	200	60
16	300	4	100	50

The top clear water from the conical flask of 16 experiments after settlement of 60 min. was collected and further, filtered with Whatman filter paper for removing impurities and then the cleared wastewater was taken for analyzing fluoride as per standard procedure given by APHA (2005). The adsorption removal percentage of fluoride in a groundwater using *Lagerstroemia indica L*. seed powder was calculated by using the following formula:

$$\text{Percentage Removal} = \frac{(C_1 - C_2)}{C_1} \text{x } 100$$

in which C_1 is the concentration of fluoride in mg/l before treatment with *Lagerstroemia indica L*. seed powder and C_2 is the concentration of fluoride in mg/l after treatment with *Lagerstroemia indica L*. seed powder. The results of the 16 experiments for removing fluoride from groundwater by *Lagerstroemia indica L*. seed powder are given in Table 3.

Table.3.The Percentage Removal of Fluoride from Groundwater using *Lagerstroemia indica L.* seed powder

Runs	Initial value, mg/L	Final value, mg/L	Percentage Removal
1	3.36	0.40	88.10
2	3.36	0.78	76.90
3	3.36	0.86	74.50
4	3.36	0.19	94.30
5	3.36	0.25	92.50
6	3.36	0.26	92.30
7	3.36	0.27	91.90
8	3.36	0.47	86.00
9	3.36	0.55	83.70
10	3.36	0.70	79.20
11	3.36	0.29	91.40
12	3.36	0.24	92.80
13	3.36	0.55	84.20
14	3.36	1.87	44.30
15	3.36	0.62	81.50
16	3.36	0.66	80.30

From the Table 3, it may be found that the maximum removal of fluoride in groundwater by *Lagerstroemia indica L*. seed powder is about 94.3 % (Run 4). This was achieved for the combination of process parameters particle size of 600 µm, adsorbent dosage of 4 g, agitation speed of 400 rpm and 60°C. Further, the maximum removal fluoride was obtained for the contact time of 60 min.

Equilibrium Study: Adsorption isotherms are mathematical models that describe the distribution of the adsorbate species among liquid and adsorbent, based on a set of assumptions that are mainly related to the heterogeneity/homogeneity of adsorbents, the type of coverage and possibility of interaction between the adsorbate species. Adsorption data are usually described by adsorption isotherms, such as Langmuir and Freundlich isotherms. These isotherms relate metal uptake per unit mass of adsorbent, q_e, to the equilibrium adsorbate concentration in the bulk fluid phase C_e. In this study, Langmuir model only described in details.

The Langmuir model is based on the assumption that the maximum adsorption occurs when a saturated monolayer of solute molecules is present on the adsorbent surface, the energy of adsorption is constant and there is no migration of adsorbate molecules in the surface plane. The Langmuir isotherm is given by:

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e}$$

The constants in the Langmuir isotherm can be determined by plotting ($1/q_e$) versus ($1/ C_e$) and making use of above equation rewritten as:

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{q_m K_L}\frac{1}{C_e}$$

where, q_m and K_L are the Langmuir constants, representing the maximum adsorption capacity for the solid phase loading and the energy constant related to the heat of adsorption respectively.

The combination of optimum process parameters particle size of 600 µm, adsorbent dosage of 4 g, agitation speed of 400 rpm and 60°C (Run 4) was used for conducting the equilibrium study against the different concentration of fluoride in a groundwater for the different contact time. The effect of concentration on reduction of fluoride in a groundwater against the different contact time is shown in Fig. 1.

From the Fig. 1, it may be observed that the adsorption capacity of *Lagerstroemia indica L.* seed powder for removing fluoride in a groundwater increased with decreasing of initial concentration. The maximum removal percentage was achieved for the dilution ratio of 4. Further, it is found that adsorption capacity of fluoride by *Lagerstroemia indica L.* seed powder occurred very rapidly within the first 20 min., but attained the equilibrium at 60 min. for all dilution factors.

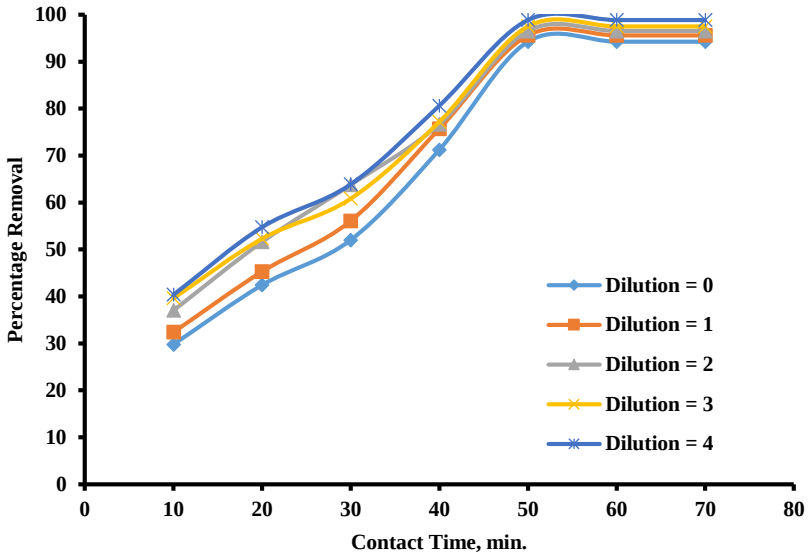

Fig.1.The effect of concentration on fluoride reduction in a groundwater with an optimum process parameters particle size of 600 µm, adsorbent dosage of 4g, agitation speed of 400 rpm and 60°C (Run 4)

The removal of fluoride in a groundwater by *Lagerstroemia indica L. seed* powder in Fig. 1 was used for fitting the Longmuir isotherm model. The results of Longmuir isotherm for the removal of fluoride in a groundwater is presented in Fig. 2.

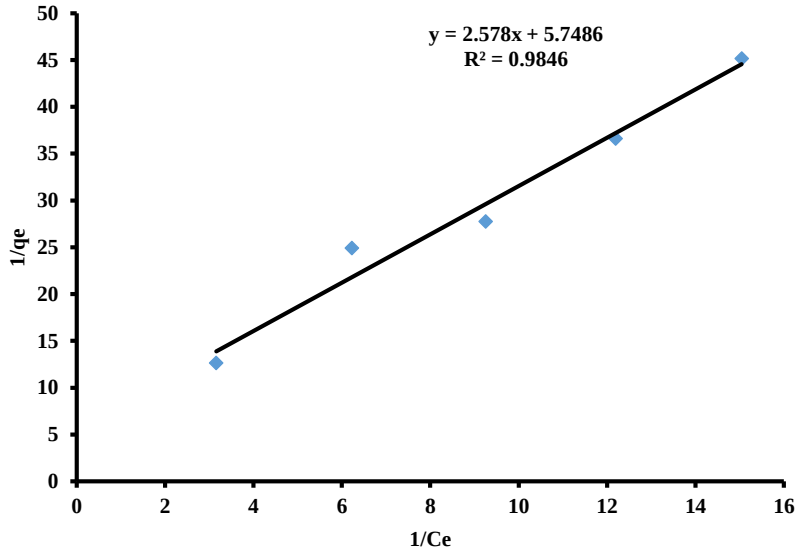

Fig.2.Longmuir Adsorption Isotherm for the removal of fluoride in a groundwater

It can be seen from Fig. 2 that the experimental data on the removal of fluoride in a groundwater is fitted well with the Langmuir equation (R^2=0.9846). From Fig. 2, the values of q_m and K_L were found to be 0.1739 mg/g and 2.229 L/mg for the parameter fluoride. Based on the results obtained from the isotherm model, Langmuir isotherm model could be reproduced the experimental results for the removal of fluoride in a groundwater.

4. CONCLUSION

Removal of fluoride was very successfully observed with the *Lagerstroemia indica L*. seed powder, which makes the treatment economical. This study demonstrated that *Lagerstroemia indica L*. seed powder as a low-cost filter was an effective adsorbent for the removal of fluoride in a batch method. The experimental parameters were discussed in detail are particle size, adsorbent dosage, agitation speed and temperature. Form this experimental parameter details it has been confirmed that there is a maximum removal of 94.3 % for the particle size of 600 µm, adsorbent dosage of 4 g, agitation speed of 400 rpm and 60°C. The experimental data on removal of fluoride in a groundwater were fitted to Langmuir isotherm model. Based on the results obtained from the isotherm model, the equilibrium data found fitted well with Langmuir isotherm model. Thus, this study concluded that *Lagerstroemia indica L*. seed powder is most economical and worthwhile adsorbent for removing fluoride in a groundwater.

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