



## REMOVAL OF FLUORIDE IN DRINKING WATER BY GREEN CHEMICAL APPROACH

**R. N. YADAV<sup>\*</sup>, RAJDEEP YADAV, NAVIN KUMAR DAGAR, PRIYANKA GUPTA, O.  
P. SINGH and M. P. S. CHANDRAWAT<sup>a</sup>**

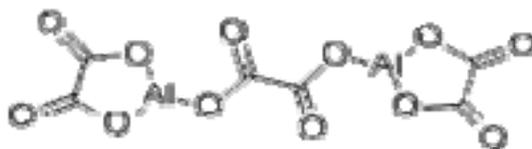
Department of Chemistry, Govt. R. R. (P.G.) Autonomous College, ALWAR (Raj.) INDIA

<sup>a</sup>School of Chem. Sci. and Tech., Eternal University, SIRMOUR (H.P.) INDIA

(Received : 14.10.2011; Revised : 29.10.2011; Accepted : 01.11.2011)

### ABSTRACT

Aluminum oxalate as adsorbent was used by authors for preparing traditional soil pots to minimize fluoride content in drinking water samples. The adsorbent dose was varied in the range of 2 g to 8 g per 500 g soil. Langmuir and Freundlich isotherms curves were also plotted. pH, TDS, hardness, fluoride and aluminum parameters were also examined in residual water samples. Maximum fluoride removal efficiency of adsorbent was studied.



Aluminum oxalate<sup>8</sup>

**Key words:** Fluorosis, Defluoridation, Adsorption.

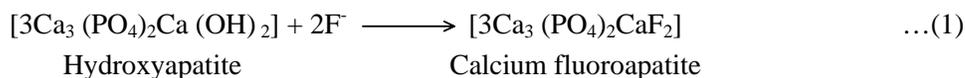
### INTRODUCTION

The prolonged intake of water containing excess of fluoride (> 1.5 ppm WHO limit) causes the crippling disease called fluorosis. The first case of endemic fluorosis in India was reported from Andhra Pradesh in 1937<sup>1</sup>. At least 17 Indian states including Rajasthan have been identified as having excess fluoride in drinking water. All the 33 districts in Rajasthan are endemic for fluorosis. According to WHO 20% villages out of total fluoride affected village in the world are found in India and out of total affected villages in India 52% are in Rajasthan<sup>4,6</sup>. It is to be noted that not only India but also several other nations in the world are severely affected by the problem of fluorosis.

Fluorosis, a disease caused by excessive intake of fluoride, Fluorosis disease is mainly of two types: Dental and Skeletal Fluorosis.

- Dental fluorosis (Fluoride intake > 1.5 ppm)
- Skeletal fluorosis (Fluoride > 3.0 ppm)

Chemical change of bone & teeth minerals is shown in Eq. 1.



Commonly used domestic defluoridation processes various defluoridation methods are used for removal of fluoride from drinking water. These exiting methods for defluoridation of drinking water is expensive, slow, in efficient, unhygienic and highly technical.

1. Nalgonda technique (Flocculation and Sedimentation)
2. Activated alumina process (Adsorption)
3. KRASS Process
4. Other processes (Bio-remedial, Ion exchange, R.O. etc.)

### **New defluoridation method by- green chemical approach**

A comparative study of degree of toxicity of NaF, NaSiF<sub>6</sub>, CaF<sub>2</sub>, CaSiF<sub>6</sub>, MgF<sub>2</sub>, ZnF<sub>2</sub>, AlF<sub>3</sub> and CuF<sub>2</sub> showed that calcium and aluminum fluoride are less toxic than other fluoride<sup>3</sup>. Therefore, in present paper authors used aluminum oxalate as defluoridating agents in soil pots and developed a new defluoridation method.

## **EXPERIMENTAL**

The following investigations were conducted to find out the fluoride minimizing capacity of aluminum oxalate in the water samples kept in the soil pots.

### **Determination of different physical and chemical parameters of water samples**

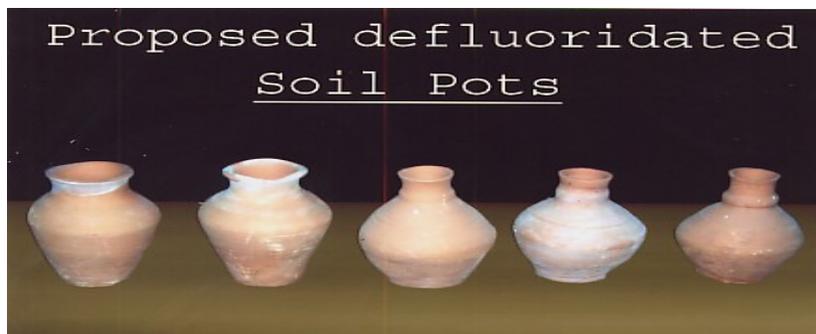
The pH, TDS and Al ion concentration were determined by the standard procedures. Result of these parameters shows that the values of all parameters in water samples are in their desirable limits.

### **Preparation of soil pots**

Four soil pots (A, B, C and D) were prepared after incorporation of aluminum oxalate (2 g., 4 g., 6 g and 8 g.) in 500 g. of soil respectively as shown in flow chart given below<sup>2,5-7</sup>.

### **Determination of fluoride concentration**

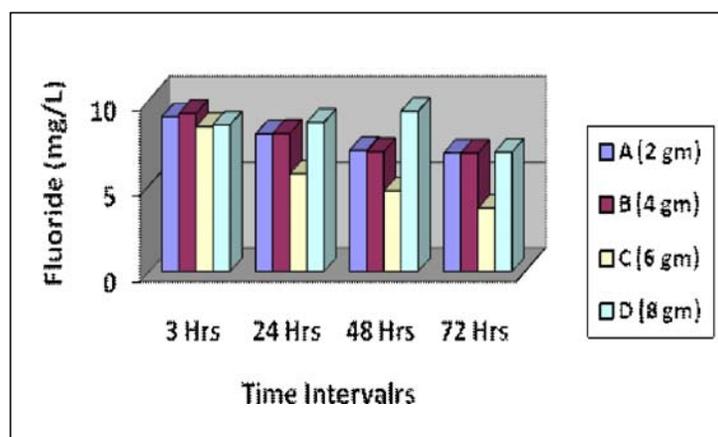
Fluoride concentration of untreated sample and the treated fluoride water samples was determined as per the standard procedure by ion selective method by Orion 720<sup>+</sup> after time interval of 3, 24, 48 and 72 hours. Results are given in Fig. 1



**Fig. 1: Proposed soil pots**

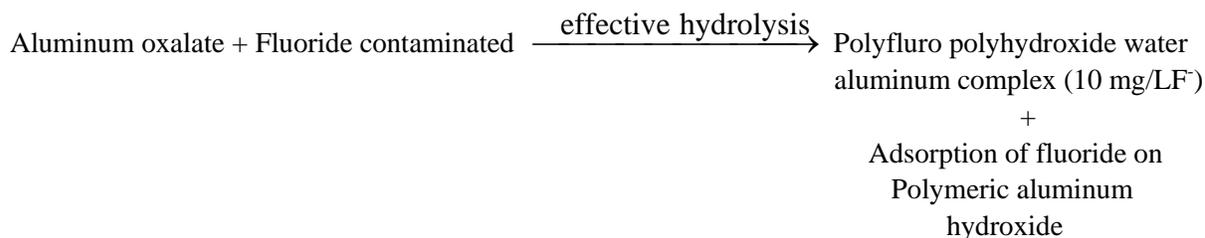
## RESULTS AND DISCUSSION

Investigation pertaining to the effect of increasing of aluminum oxalate in the soil pot on the fluoride concentration in Fig. 2 shows that the fluoride concentration of water sample (10 mg/L fluoride) decreases in the all soil pots with increasing amounts of aluminum oxalate at contact time periods. It is noticed that soil pot No-1 (having 2 g  $C_6Al_2O_{12}$ ), decreases the fluoride concentration of the water sample about 20% but in case of soil pot No. 3 (having 6 g  $C_6Al_2O_{12}$ ), the concentration of fluoride decreases about 70% at time interval 72 hours. This can be explained on the basis of surface chemistry. It is a general phenomena of surface chemistry that more the surface area of adsorbent more the adsorbate are adsorbed on the surface of adsorbent to form a unimolecular layer (Langmuir isotherm limitation) of adsorbent during chemisorption process. Result shows that a certain amount of aluminum oxalate reduces the fluoride concentration in the water sample.

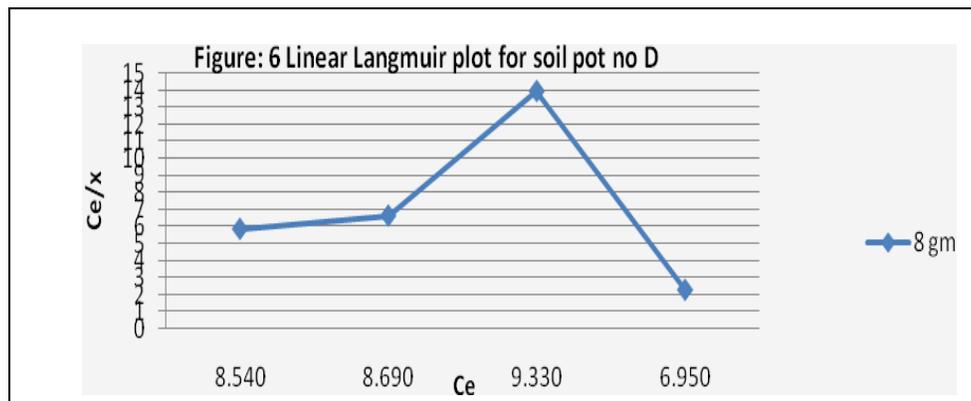
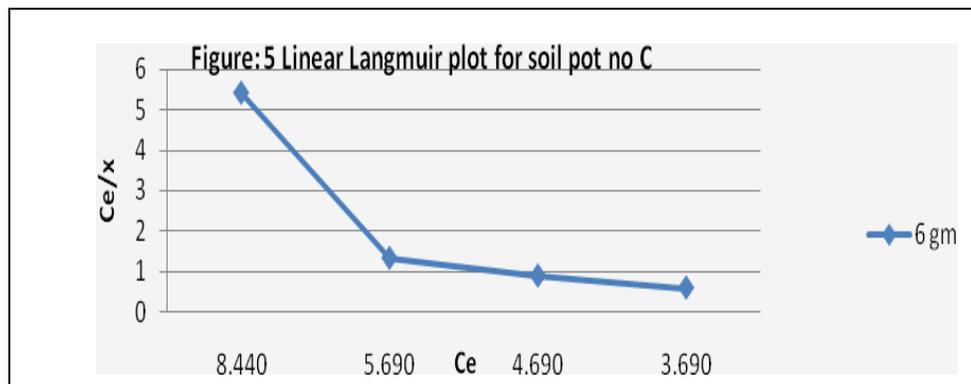
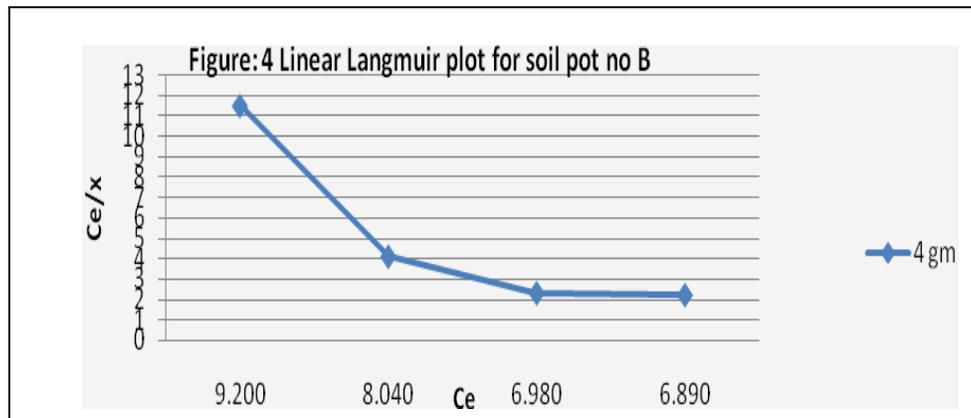
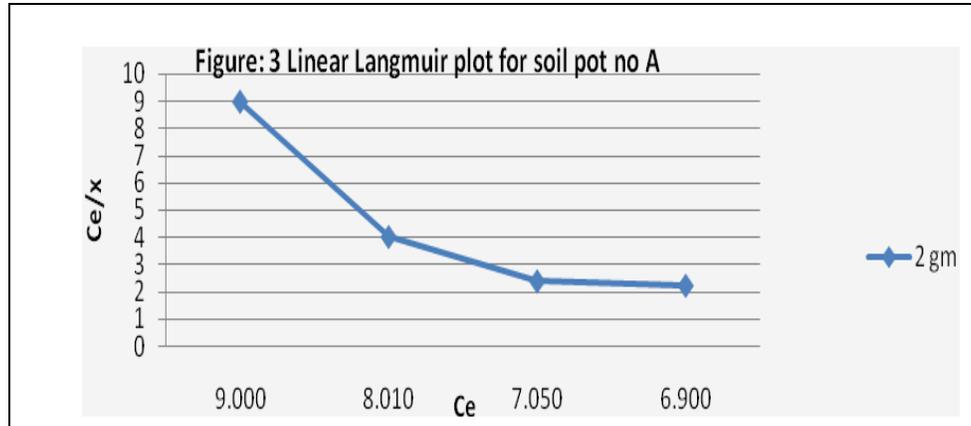


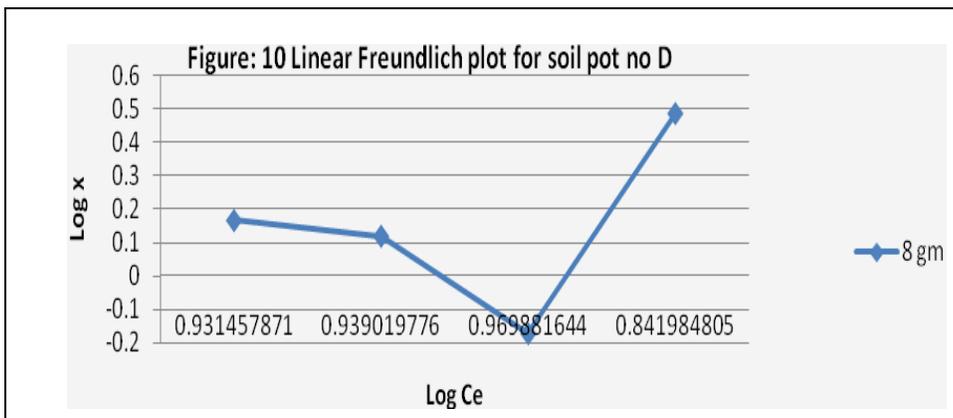
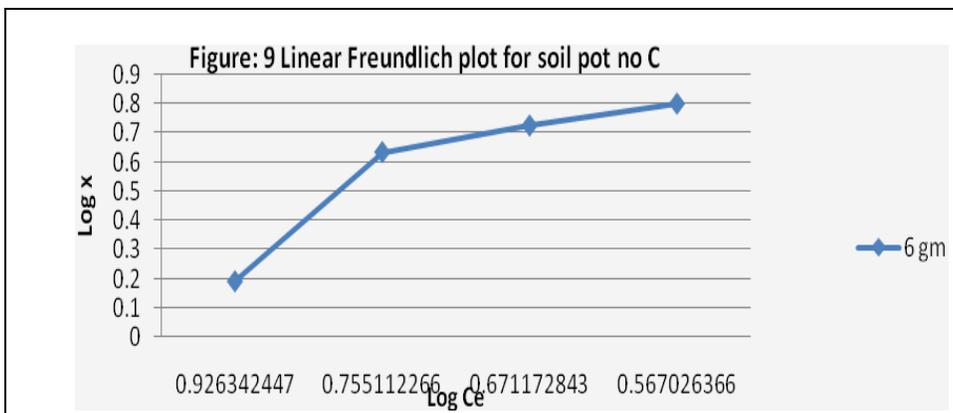
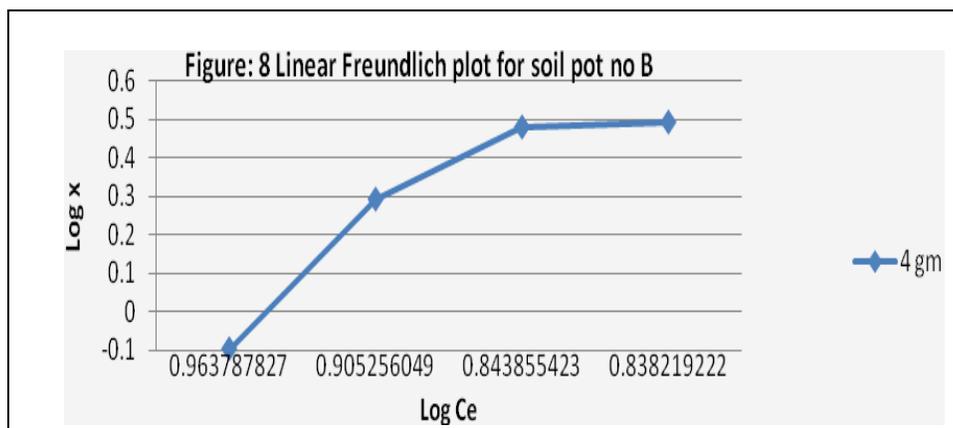
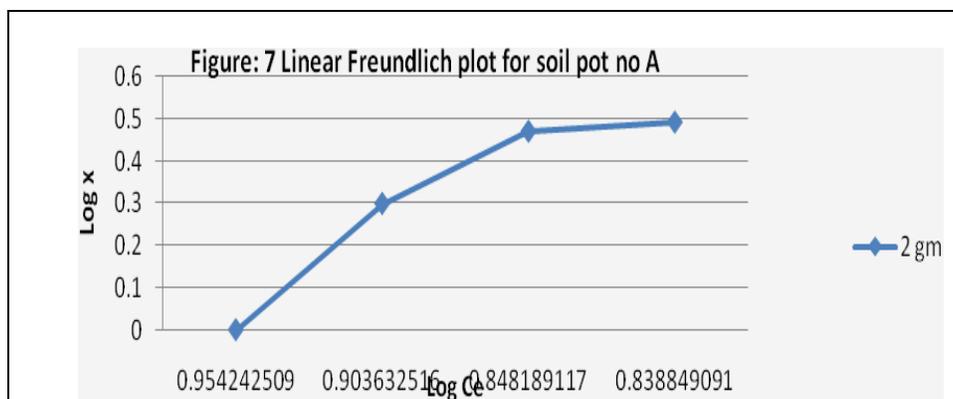
**Fig. 2: Concentration of fluoride ions in different soil pots**

The removal of fluoride from water sample (10 mg/L) is expected due to the formation of polyhydroxide aluminum complex (e.g.  $[Al(H_2O)_3(OH)_3]$   $[Al(H_2O)_2(OH)_4]$  etc.) with fluoride and adsorption of fluoride on polymeric aluminum oxalate.



The linear Langmuir plots between  $C_e/x$  and  $C_e$  are shown in the Fig. 3-6. The linear Freundlich isotherm models are shown in the Fig. 7-10 by plotting  $\log x/\log C_e$ . The constant values of the both isotherms for each soil pot are given in the Table 1. It is observed from the curves and the correlation coefficient data that the fluoride adsorption follows neither Langmuir isotherm nor Freundlich isotherm in a perfect way. However, it follows Langmuir isotherm in a better way. The value of  $n$  is always less than unity, which indicates that, none of the soil pots have completely energetically homogeneous surface. This can be explained that all the four soil pots are associated with certain amounts of aluminum oxalate at certain specific sites having different activation energy. The adsorption involves attractive electrostatic interaction between the negative sites created by the ionization of the sodium fluoride and the positively charged  $Al^{3+}$  cations.





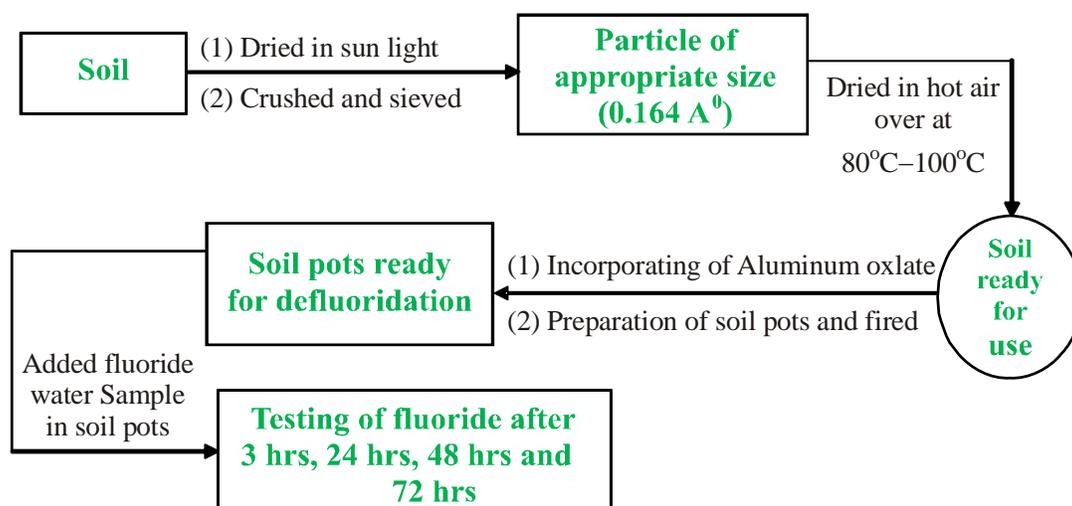


Fig. 11: Flow chart of adsorbent (soil pots) preparation and fluoride adsorption on to soil pots

Table 1: Langmuir and Freundlich equation constants for adsorption of fluoride ions on different soil pots

Soil Pots No.	$C_6Al_2O_{12}$ in soil pots (g/500 g soil)	Langmuir equation constants		Freundlich equation constants	
		$x_m$ (mg/g)	k	N	$k_f$
1	2 g	0.487075	-0.17016	-3.82577	3.736795
2	4 g	0.46465	-0.16922	-4.07778	3.954843
3	6 g	1.807318	-0.36847	-1.5051	1.736619
4	8 g	0.203617	-0.13747	-6.16173	5.881287

## CONCLUSION

- (i) Adsorption isotherm of fluoride ions follows the mixed model of the Langmuir and Freundlich isotherm. The adsorption does not depend on the BET surface area of the pots and takes place on certain specific site.
- (ii) Aluminum oxalate can be used as defluoridating agent in soil pots without effecting the environment as a Green Chemical Approach.

## REFERENCES

1. Rajiv Gandhi National Drinking Water Mission, Prevention and Control of Fluorosis in India, Ministry of Rural Development, New Delhi, **Vol. I**, 24-40 (1993).
2. Standard Methods for the Examination of Water and Waste Water, 17<sup>th</sup> Ed., American Public Health Association, APHA, New York, U.S.A. (1989).
3. Vogel's Qualitative Inorganic Analysis, 6<sup>th</sup>, Revised by 7G Suehla, Reprinted Published by Orient Longmann Ltd, 1/24, Asafali road, New Delhi (1989) pp. 180.
4. R. N. Yadav and Rajdeep, Aluminum Chloride as Defluoridating Agent in Drinking Water Soil Pots (Vessels of Earthenware), Rec. Trd. Ph. Sci. Env. (2008) pp. 13-26.

5. R. N. Yadav and Rajdeep, Aluminum Nitrate ( $\text{Al}(\text{NO}_3)_3$ ) as Defluoridating Agent in Drinking Water Soil Pots (Vessels of Earthenware), NEERI, (2011).
6. R. N. Yadav and Rajdeep, Defluoridation of the Potable Water by Aluminum Sulphate, *Int. J. Chem. Sci.*, (2009) p. 760-774.
7. R. N. Yadav and Rajdeep, Study of the Aluminum Ammonium Sulphate as Defluoridated Agent in Drinking Water Earthenware, *Archives of Appl. Sci. Res.*, (2010) p. 11-22.
8. <http://en.wikipedia.org>.Wikipedia the Free Encyclopedia.