Defluoridation of Water by Murraya koenigii (curry leaves) - A Natural Bio-Adsorbent

Akshay Sharma, Sunder Lal Meena, Surbhi Benjamin, Dipti Soni, Rakshit Ameta and Paras Tak*

Department of Chemistry, PAHER University, Udaipur (Raj.)

ABSTRACT: The excess amount of fluoride in drinking-water causes harmful effects such as dental and skeletal fluorosis. In the fluoride endemic areas, especially small communities with staggered habitat, defluoridation of potable water supply is still a problem in this study, it is investigated that removal of fluoride at different pH and contact time the effect of this adsorbent on fluoride removal was compared with other available adsorbents in ground water by (Curry Leaves) Murraya *koenigii*. It is much better adsorbent with high removal efficiency at higher concentration (5 ppm) of fluoride in ground water. In this work effect of time for adsorbent and initial concentration of fluoride on the % removal was also studied.

Keywords: Fluoride, Adsorption, Low cost Adsorbents, Contact time, Environmental Impact, pH.

INTRODUCTION

Fluoride is known to be a natural contaminant for ground water resources globally. Fluorine, a fairly common element of earth crust, is present in the form of fluorides in a number of minerals and in many rocks soil, and fresh and ocean water (Chidambaram, S. *et al.* 2001). The fluoride may be beneficial or detrimental depending upon its concentration and total amount consumed by drinking water (Underwood E.J. 1997). Over the past few decades, scientific literature in fluoride research suggests that fluoride concentrations between 0.7–1.2 mg/L are beneficial, especially to infants for the prevention of dental caries or tooth decay, but concentrations above1.2 mg/L cause mottling of teeth in mild cases but fluorosis (dental or skeletal) and several neurological disorders in severe cases (Environmental Health Criteria, EPA &HSS website; Susheela A.K. 2001). India is among 23 countries in the world facing this chronic disease. It is estimated that around 62 million people in 19 states of India are affected with various forms of fluorosis, which include dental, skeletal and non-skeletal manifestations (Chauhan V.S. *et al*. 2007). In the drinking water has been set as 1.5 mg/ L Maximum permissible limit of fluoride by many regulatory authorities like WHO, US EPA, CPCB etc Since many other available methods of defluoridation are costlier, there is an urgent need of developing a low cost method. Adsorption, ion exchange, electrodialysis, coagulation/ precipitation, dialysis, reverse osmosis, nano-filtration, ultra-filtration, etc. many processes available for the removal of fluoride from water (Chubar N.I. *et al*. 2005; Haron M.J. and Yunus W.M. 2001; Haron M. J. *et al*. 2001; Hichour M. *et al*. 2000; Kabay *et al*. 2008 ; Meenakshi S. and Maheshwari R.C. 2006; Meenakshi S. and Vishwanathan N. 2007; Papat K.M. *et al*. 1994; Simons R. 1993; Sourirajan S. and Mastsura T., 1972; Sundaram C.S. *et al*. 2008; Sundaram C.S. *et al*. 2008; Tor A. *et al*. 2009). The study of Ground Water Fluoride in Chandrapur city in Maharashtra, and effects of fluoride on human health are dependent on the concentration of fluoride in water (Weginwar N. 2008). The observation of Ground Water Fluoride in Chandrapur city help that understand the problem of fluoride in the India (Varadajan N. and Purandara B.K. 2008).

* *To whom correspondence be made:*
E-mail: parastak2011@gmail.com

Table 1
Biological Effects on Human Health

Fluoride conc. (mg/lit)	*Source*	*Effects*
1	Water	Prevention of dental caries
2	Water	Effect dental enamel
3 to 6	Water	Osteoporosis
8	Water	10 % Osteoporosis
20 to 80	Air & Water	Crippling skeletal fluorosis

Permissible Limits for fluoride Concentration in beverage is following according

- Bureau of Indian Standards (BIS)-0.6 to 1.2 mg/lit
- World Health Organization (WHO-1984) for drinking water-1 to one.5 mg/lit
- Indian Council of Medical analysis (ICMR-1975)-1 mg/lit
- World Health Organization (WHO) European Standards-zero.7 to 1.7 mg/lit associated with temperature

When the fluoride level in a beverage is marginally on top of one.0 mg/l happens molted enamel. A relationship between fluoride concentration in potable water and patterned enamel was initially established in 1931. Typical manifestations of dental pathology area unit loss of shining and development of horizontal yellow streaks on teeth. The high halide concentration in or adjacent to developing enamel, dental pathology develops in youngsters born and referred to in endemic areas of pathology. Once shaped, the changes within the enamel area unit permanent. The molted enamel area unit seen in Associate in nursing adult, they clearly indicate that the person has been exposed to high halide levels throughout her or his childhood. Skeletal pathology affects each adults and youngsters and is mostly manifested when consumption of water with fluoride levels surpassing three mg/l. typical symptoms of skeletal pathology area unit pain within the joints and backbone. In severe cases, this may lead to unhealthful the patient. Recent studies have shown that excess intake of fluoride may have sure non-skeletal health impacts like gastro-intestinal issues, allergies, anemia and tract issues. The undesirable effects of fluoride cause by nutritionary deficiencies.

The Defluoridation ways divided into 3 classes on the idea of the most removal mechanism:

- Chemical additive ways
- Contact precipitation
- Adsorption/ion exchange ways

Adsorption/ion-exchange method

The material retains fluoride either by physical, chemical or ion exchange mechanisms. During this methodology the raw water is passed through a bed containing defluoridating Material. The good vary of materials has been tried for fluoride uptake. Bauxite, magnetite, kaolinite, serpentine, numerous varieties of clays and red mud ar a number of the present materials studied. the overall mechanism of fluoride uptake by these materials is that the exchange of metal lattice chemical group or alternative anionic teams with halide. The bound pre-treatments like acid laundry, calcinations, etc. is inflated fluoride uptake capability. Rajasthan is one state wherever halide in high level is current all told the thirty two districts and has become a serious health hazard in 18 of them.

According to the survey conducted by the general public Health Engineering Department within the recent past, the drinking water sources in 9741 out of 37889 villages, 25.7%, and 6819 out of 45311, 15%, habitations were found to contain fluoride over one.5 mg/L within the absence of perennial rivers, surface sources and canal systems, groundwater, that usually contains high fluoride concentrations, several natural and inexpensive materials like red mud (Cengeloglu Y. *et al*. 2002; Tor A. *et al*. 2009). The Zr fertilized coconut shell carbon (Sai Satish R. *et al*.2007), edible nut shell carbon (Alagumuthu G. and Rajan M. 2010), ground nut shell carbon and clays (Tor A. 2006) are used as adsorbents for halide removal from beverage. Recently, amorphous corundum supported on carbon nanotubes (Li Y.H. *et al*. 2001), aligned carbon nanotubes (Li Y.H. *et al*. 2003), natural action compound fiber (Ruxia *et al*. 2002), and an particle exchanger supported a double hydrous chemical compound of Al and Fe ($Fe_2O_3Al_2O_3.xH_2O$) have been assayed for removing fluoride from drinking water additionally as industrial wastewater.

The some agriculture waste materials (bio-adsorbents) are inexpensive, available in large quantities and remain unused; they can be disposed without concerning expensive regeneration process. Furthermore, plant materials like Citrus limonum (lemon) leaf (Tomar V. *et al*. 2014), Ficusreligiosa (Peepal Leaf Powder) (Dwivedi S. *et al*. 2014), Banana peel dust (Bhaumik and Mondal N.K. 2014), Devdaru

(Polyalthia Longifolia) leaves (Bharalia R.K. and K.G. Bhattacharyya 2014), Neem charcoal (Chakrabarty S. and Sharma H.P. 2008). Neem (Azadirachta indica) and Kikar (Acacia arabica) leaves (Kumar S. *et al*. 2008) are reported as fluoride sequestration and hence applicable for defluoridation agents. Apart from these numerous waste, biomass sources on which some experimental adsorption properties have been reported e.g. Sawdust (Mann S. and Mandal A. 2014), Tea Ash (Mondal *et al*. 2012), Maize Husk Fly Ash (Jadhav A.S. and Jadhav M. V. 2014), Eggshell Powder (Bhoumik R. *et al*. 2012), Algal Spirogyra Sp. I02 (Venkata M. *et al*. 2007b), Babool Bark (Mamilwar B.M. *et al*. 2012), Tamarind seed (Murugan M. and Sbramanian E. 2006), Banana peel, groundnut shell and sweet lemon peel (Mohammad A. and Majumdar C.B. 2014). Reports on extraction of fluoride using different biomass are also available in recent literatures.

2. MATERIAL AND METHODS

Preparation of Standard fluoride solution (5ppm solution)

The stock solution of 100mg/ liter (100 ppm) of fluoride was prepared by dissolving 221 mg of anhydrous sodium fluoride (NaF) in 1000 ml distilled water. Now 5 ppm solution prepare from stock solution by suitable dilution.

The **curry tree** (*Murraya koenigii*) is a tropical to sub-tropical tree in the family Rutaceae (the rue family, which includes rue, citrus, and satinwood), which is native to India and Sri Lanka. Its leaves are used in many dishes in India, Sri Lanka, and neighbouring countries. Often used in curries, the leaves are generally called by the name 'curry leaves', although they are also literally 'sweet neem leaves' in most Indian languages (as opposed to ordinary neem leaves which are very bitter and in the family Meliaceae, not Rutaceae).

Preparation of adsorbent *Murraya koenigii*

Murraya koenigii leaves easily available at plant collect and washed with distilled water several times then dried in sunlight for 3-4 days. This dry leaves grind with mixer & sieved with suitable mesh size (30 bss size) taken for analysis

Measurement: To observe fluoride Removal used Fluoride Ion Meter Panomex Model PX/IMC/321. First Calibrate with 100 ppm & 10 ppm solution prepared by stock solution. Now check 5 ppm fluoride solution for system response Washed Plastic beaker with tap water then distilled water Take 100 ml of 5 ppm solution of water in 250ml plastic beaker weigh 1.0 gm of adsorbent hold for 30 minutes after 30 min filter the solution take reading using various parameter like pH, dose of adsorbent and contact time of adsorbent.

3. RESULTS & DISCUSSION

(i) Effect of pH

The result of the pH on the removal of fluoride was studied by varied the pH from pH 2 to 11. The pH plays a vital role in adsorption method on bio adsorbents. The removal efficiency of the adsorbents is concluded that it's looking on the pH of the test sample of fluoride, as shown in Fig.1. The results are presented, wherever it will be seen that most removal of about 56 % is found at pH-8 & dose of 10 g/L and thereafter the percent removal became more or less constant.

(ii) Dose of Adsorbent (*Murraya koenigii*)

The adsorbent dose on the removal of fluoride was studied by varied the adsorbent dose from 0.25 to 2 g/100ml. The results are presented, wherever it can be seen that maximum removal of about 84 is found at a dose of 2 g/100ml and thereafter the % removal became more or less constant. At the beginning, removal of fluoride will increase as increasing the dose until some extent at the moment very slightly modification within the removal of fluoride it means, the curve lapse as flat indicating the higher fluoride adsorption occurs at their maximum dose and the removal remains constant. a higher availability of surface and pore volume in adsorption because of this surface assimilation increases after that adsorption of fluoride is constant at higher dose because of saturation of pore volume and surface.

(iii) Effect of Contact Time

It is observed that at optimum pH and dose fluoride ions increases with increase in contact time to some level. The optimum percentage removal of fluoride by three considered bio adsorbents at different contact times explain by Fig. 3. The adsorption of metal ion by adsorbent also depends on the interactions of functional groups between the solution and the surface of adsorbent. Adsorptions can be assumed to be complete when equilibrium is achieved between the solute of solution and the adsorbent. However, it progressively approached an almost steady value, denoting accomplishment of equilibrium, these similar trends also observed in Effects of stirring rate and temperature on fluoride removal by fishbone charcoal (Kiledar D.J. and Bhargava D.S. 1993)

(iv) Effect of Temperatur

The using neem charcoal adsorbent as the defluoradation of contaminated drinking water by kinetics and equilibrium studies (Chakrabarty S. and Sharma H.P. 2012).

(v) Kinetics Studies

(a) Psuedo-First Order

The adsorption kinetics data can be described by pseudo-first order kinetic equation. The linear form of this equation is-

$$\text{Log}\,(Q_e\text{-}Q_t) = \log Q_e\text{-}\ k\text{-}_1 \times t \qquad (1)$$

where, Q_e and Q_t are amounts adsorbed at equilibrium and at time t, respectively. k_1 is the rate constant ofpseudo-first order equation.

(b) Pseudo-Second Order

The kinetics can also be described by pseudo-second order model. The linear form of pseudo-second order is expressed as -

$$T/Q_t = 1/k_2/Q_e^{\,2} + 1/Q_e \times t \qquad (2)$$

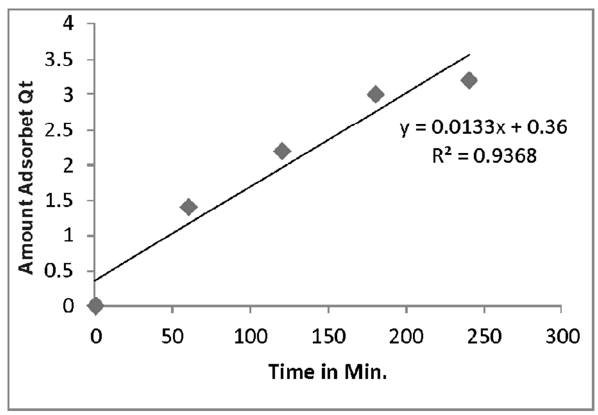
where, k_2 is pseudo-second order constant. A plot of T/Q_t against t gives a linear relationship Q_e and k_2 can be determined from the slope and intercept of this plot.

Table (V)
Kinetics models

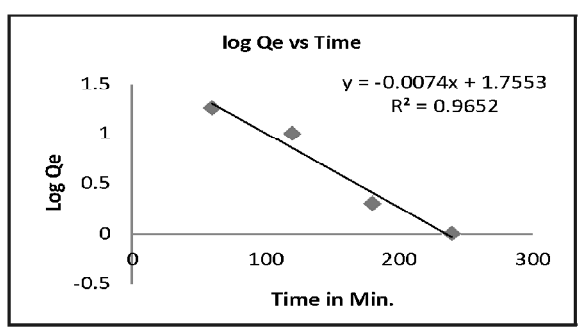
Q_{max}	*Time (t) (min)*	$Q_e = (Q_{max}\text{-}Q_t)$	*1+log* Q_e	*T/Qt*
	0.0	3.2	1.3802	0
	60.0	1.8	1.255273	42.8571
3.2	120.0	1	1	54.5454
	180.0	0.2	0.301	60
	240.0	0	0.0	75

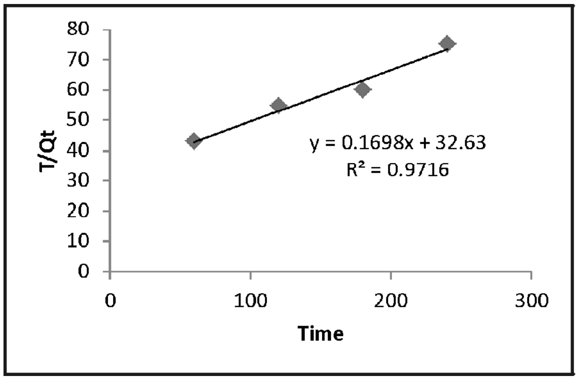
The values of R^2 in plots (Figs. 2 and 3) are 0.965 and 0.971 for pseudo-first order and pseudo-second order, respectively, which indicates that this adsorption process followed both the kinetic models, but pseudo-second order model was more suitable than the pseudo-first model.

Time	*Amount Adsorbed*	*T/Qt*
0	0	0
60	1.4	42.8571
120	2.2	54.5454
180	3	60
240	3.2	75



Time	*Qe*	*1+log*
0	3.2	1.3802
60	1.8	1.255273
120	1	1
180	0.2	0.301
240	0	0.0





Time	*T/Qt*
0	0
60	42.8571
120	54.5454
180	60
240	75

(vi) Adsorption Isotherm

The plot of the mass of gas adsorbed per gram of adsorbent (x / m) versus equilibrium pressure at constant temperature is given by equations (i).

In the Langmuir adsorption isotherm, the degree of adsorption is directly

Proportional to e, i.e., the fraction of surface area occupied.

$$x / m \ \alpha \ \theta = k\theta \qquad \text{(i)}$$

The relationship between the quantity of gas adsorbed by unit mass of solid adsorbent and pressure at particulate temperature. It can be expressed by the equation (ii).

$$x / m = kp^{1/n} \qquad \text{(ii)}$$

(a) Langmuir Adsorption Isotherm

In the Langmuir equation, the degree of adsorption is directly proportional to e, i.e., the fraction of surface area occupied by the equation (iii).

$$x / m \ \alpha \ \theta = k\theta \qquad \text{(iii)}$$

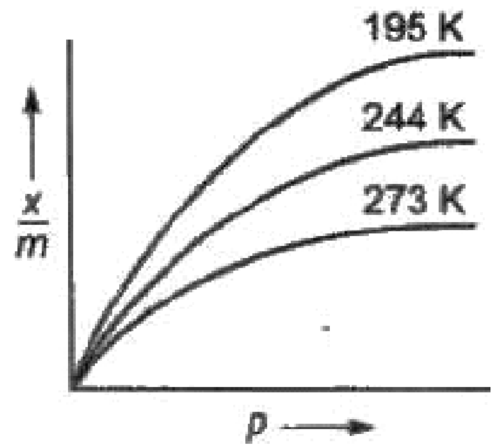
this effect explain by the table no. v and graph.

(b) Freundlich Adsorption Isotherm

It gave an empirical relationship between the quantity of gas adsorbed by unit mass of solid adsorbent and pressure at a particular temperature. It can be expressed by the equation.

$$x / m = kp^{1/n} \qquad \text{(i)}$$

Where, x is the mass of the gas adsorbed on mass m of the adsorbent at pressure p, k and n are constants which depend on the nature of the adsorbent and the gas at a particular temperature.



At low pressure, $n = 1$, i.e., $x / m = kp$

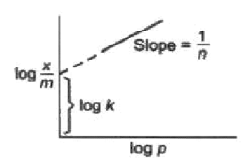
At high pressure, $n > 1$, i.e., $x / m = k$ (independent of p)

Taking logarithm of Eq. (i)

$$\log \frac{x}{m} = \log k + \frac{1}{n} \log p$$

Plot of $\log \frac{x}{m}$ *vs* $\log p$ is a straight line with slope $\frac{1}{n}$ and intercept on *y*-axis = $\log k$.

The factor $\frac{1}{n}$ can have values between 0 and 1.



(v) Physical characterization

The ***Murraya koenigii*** leaf powder before (untreated) and after (treated) batch adsorption was Characterized using SEM (Scanning electron microscopy), in this experiment under the given conditions: initial fluoride concentration = 5 mg/L, temperature = 30ºC, shaking speed = 150 rpm and contact time = 24h respectively, for the preparation of SEM samples.

The SEM images of the *Murraya koenigii* leaf powder before treatment of fluoride ions shows a cluster of very fine particles with a surface coating while the SEM images of the *Murraya koenigii* leaf powder after treatment of fluoride ions shows small irregularly shaped fine particles mostly fluoride ions adhering to the surfaces of the adsorbent. Fig. 1.1 &1.2.

4. CONCLUSIONS

The paper 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. Observation of various low cost adsorbents presented here shows a great potential for the fluoride removal. The use of commercially available adsorbents can be replaced by the inexpensive and effective low cost adsorbents. There is need for more studies to better understand the process of low-cost adsorption and to demonstrate the technology effectively.

Acknowledgements

This research work is the account of devoted work fluoride removal from ground water using Bio-Adsorbent in the field of Chemistry at PAHER University, Udaipur, India, which would not have been possible without the help of many. A few lines are too short to make a complete account of my deep appreciation for my advisor Dr. Paras Tak. I also thankful to our dean Prof. Suresh C. Ameta, I would like to thank him for his valuable guidance and constant encouragements which I have received during the research work.

I acknowledge my sincere regards to Prof. B.P. Sharma, President PAHER University and Mr. Sharad Kothari, Registrar, Prof. Hemant Kothari, Dean P.G. Studies, PAHER University, Udaipur to provide me laboratory and funding facilities for analysis, special thanks to all Faculty members, Department of Chemistry, PAHER University, Udaipur, India, for their dedication towards students. I am also thankful to my lab mates' Sunderlal Meena who worked with me during the research work.

References

[1] Alagumuthu G and Rajan M (2010) Equilibrium and Kinetics of Adsorption of Fluoride onto Zirconium Impregnated Cashew Nut Shell Carbon. *Chem. Eng. J.* 158:451–457.

[2] Bharalia R K and Bhattacharyya K G (2014) Kinetic and Thermodynamic studies on fluoride Biosorption by Devdaru (Polyalthia longifolia) Leaf powder. Octa Journal of Environmental Research Vol. 2(1): 22-31.

[3] Bhaumik R and Mondal N K (2014) Optimizing adsorption of fluoride from water by modified banana peel dust using response surface modelling approach. Appl Water Sci DOI 10.1007/s13201-014-0211-9.

[4] Bhaumik R, Mondal N K, Das B, Roy P, Pal K C, Das C, Banerjee A, and Datta J K (2012) Eggshell Powder as an Adsorbent for Removal of Fluoride from Aqueous Solution: Equilibrium, Kinetic and Thermodynamic Studies, E-Journal of Chemistry. 9(3):1457-1480.

[5] Cengeloglu Y., Kir E, Ersoz M (2002) Removal of Fluoride from Aqueous Solution by Using Red Mud. *Sep. Purif. Technol.,* 28(1):81–86.

[6] Chakrabarty S and Sarma H P (2012) Defluoridation of contaminated drinking water using neem charcoal adsorbent: kinetics and equilibrium studies. *Int. J. Chem.Tech.Res.* 4:511–516.

[7] Chakrabarty S and Sarma H P (2012) Defluoridation of contaminated drinking water using neem charcoal adsorbent: kinetics and equilibrium studies. Int J ChemTech Res 4(2):511–516.

[8] Chauhan V S, Dwivedi P K and Iyengar L (2007) Investigations on activated alumina based domestic defluoridation units. *J. Hazard. Mater.* B 139**:** 103–107.

[9] Chidambaram S Ramanathan AL, Vasudevan S, (2001) Fluoride removal studies in water using natural material. Water, air and soil pollution, 29: 0378-473.

[10] Chubar N I, Samanidou V F, Kouts V S, Gallios G G, Kanibolotsky V A, Strelko V V, Zhuravlev I Z (2005) Adsorption of fluoride, chloride, bromide, and bromated ions on a novel ion exchange, *J. Colloid Interface Sci.* 29:167–74.

[11] Dwivedi1 S, Mondal P, and Balomajumder C (2014) Bioadsorption of Fluoride by Ficusreligiosa (Peepal Leaf Powder): Optimization of process Parameters and Equilibrium study, Research Journal of Chemical Sciences Vol. 4(7):52-60.

[12] Environmental Health Criteria, EPA &HSS website.

[13] Haron M J, Yunus W M (2001) Removal of fluoride ion from aqueous solution by a cerium–poly (hydroxamic acid) *J. Environ. Sci. Health* A. 36:727–734.

[14] Jadhav A S, Jadhav M V, (2014) Use of Maize Husk Fly Ash as an Adsorbent for Removal of Fluoride from Water, International Journal of Recent Development in Engineering and Technology . 2(2):41-45.

[15] Kabay N, Arar O, Samatya S, Yuksel U, Yuksel M, (2008) Separation of fluoride fromaqueous solution by electrodialysis: effect of process parameters and other ionic species, *J. Haz. Mater*. 153:107–113.

[16] Killedar D J and Bhargava D S (1993) Effects of stirring rate and temperature on fluoride removal By fishbone charcoal. *Ind. J. Environ. Health.* 35:81–87.

[17] Kumar S, Gupta A and Yadav J P (2008) Removal of fluoride by thermally activated carbon Prepared from neem (Azadirachta indica) and kikar (Acacia arabica) leaves. Journal of Environmental Biology. 29(2):227-232.

[18] 25. Li Y H, Wang S, Cao A, Zhao D, Zhang X, Wei J, Xu C, Luan Z, Ruan D, Liang J, Wu D, Wei 26. B (2001) Adsorption of fluoride from water by amorphous alumina supported carbon 27. nanotubes. *Chem. Phys. Lett.,* 350:412–416.

[19] Li Y H, Wang S, Zhang X, Wei J, Xu C, Luan Z, Wu D (2003) Adsorption of fluoride from water by aligned carbon nanotubes, *Mater. Res. Bull.* 38:469–476.

[20] Mamilwar B M, Bhole A G, Sudame A M (2012) Removal of Fluoride From Ground Water By Using Adsorbent Int. J. Eng. Res. Appl. 2,(4):334-338.

[21] Mann S and Mandal A (2014) Removal of Fluoride from Drinking Water Using Sawdust, Int. Journal of Engineering Research and Applications ISSN : 2248-9622, Vol. 4, 7 (2):116-123.

[22] Meenakshi S and Maheshwari R C (2006) Fluoride in drinking water and its removal, *J. Haz. Mater.* 137: 456-463 (2006).

[23] Meenakshi S and Viswanathan N (2007) Identification of selective ion-exchange resin for fluoride sorption, *J. Colloid Interface Sci.,* 308:438–450.

[24] Mohammad A, and Majumder C B (2014) Removal of fluoride from synthetic waste water by using Bio-Adsorbents , IJRET: International Journal of Research in EngineeringandTechnology.3(4):776-785.

[25] Mondal N K, Bhaumik R, Baur T, Das B, Roy P and Datta J K (2012) Defluoridation of Water by Tea Ash: An Unconventional Biosorbent, Chem Sci Trans. 1(2):239-256.

[26] Murugan M and Subramanian E (2006) Studies on defluoridation of water by Tamarind seed, an unconventional biosorbent, J Water Health. 4: 453-461.

[27] Popat K M, Anand P S and Dasare B D (1994) Selective removal of fluoride ions from water by the aluminium form of the aminomethylphosphonic acid-type ion exchanger, *React. Polym.* 23:23– 32.

[28] Ruixia L, Jinlog G, Hongxiao T (2002) Adsorption of fluoride, phosphate, and arsenate ions on a new type of ion exchange fiber *J. Colloid Interf. Sci.* 248:268–274.

[29] Sai sathish R, Raju N S R, Raju G S, Nageswara Rao G, Anil Kumar K, Janardhana C (2007) Equilibrium and kinetic studies for fluoride adsorption from water on zirconium impregnated coconut shell carbon. *Sep. Sci. Technol.,* 42(4):769–788.

[30] Simons R (1993) Trace element removal from ash dam waters by nanofiltration and diffusion dialysis, *Desalination.* 89:325–341.

[31] Sourirajan S and Matsurra T(1972) Studies on reverse osmosis for water pollutioncontrol. *Water Res.* 6:1073–1086.

[32] Sujana M G, Thakur R S, Das S N and Rao S B, (1997) Defluorination of wastewaters,*Asian J. Chem.* 4:561–570.

[33] Sundaram C S Viswanathan N and Meenakshi S (2008) Uptake of fluoride by Nano- hydroxyapatite/ chitosan, a bioinorganic composite, *Bioresour. Technol.* 99:8226–8230.

[34] Sundaram C S, Viswanathan N and Meenakshi S (2008) Defluoridation chemistry of synthetic hydroxyapatite at nano scale: equilibrium and kinetic studies, *J. Haz. Mater.* 155:206–215.

[35] Susheela A K (2001) Treatise on Fluorosis, Fluorosis Research and Rural Development Foundation, India.

[36] Tomar V, Prasad S, Kumar D (2014) Adsorptive removal of fluoride from aqueous media using Citrus limonum (lemon) leaf, Microchemical Journal 112:97–103.

[37] Tor A (2006) Removal of fluoride from an aqueous solution by using montmorillonite. *Desalination,* 201:267–276.

[38] Tor A Danaoglu N Arslan G Cengeloglu Y (2009) Removal of fluoride from water by using granular red mud: batch and column studies. *J. Hazard. Mater.* 164: 271–278.

[39] Underwood E J (1997) Trace Elements in Human and Animal Nutrition. Academic Press, New York, 545.

[40] Varadajan N and Purandara B K (2008) Fluoride Contamination in Ground water ofMalaprabha Sub Basin, *Journal of Environ Science and Engg.* 50(2):121-126.

[41] Venkata M S, Ramanaiah, S V, Rajkumar, B, Sarma, P N (2007b) Removal of fluoride from aqueous phase by biosorption onto algal biosorbent Spirogyra Sp. I02: sorption mechanism elucidation. J. Hazard. Mater. 141:465–474.

[42] Weginwar N, Takarkhede A G and Kamble R K (2008) Ground Water Fluoride in Chandrapur city in Maharashtra, *Indian Journal of Environmental Protection*, 28(7): 648- 653.

(1) Effect of pH

pH	*Initial concentration (ppm)*	*Final concentration (ppm)*	*Removal in ppm*	*% Removal*
2.0	5.0	4.2	0.8	16
3.0	5.0	3.9	1.1	22
4.0	5.0	4	1.0	20
5.0	5.0	3.5	1.5	30
6.0	5.0	2.8	2.2	44
7.0	**5.0**	2.3	**2.7**	54
8.0	5.0	2.2	**2.8**	56
9.0	5.0	2.4	2.6	52
10.0	5.0	3.2	1.8	36
11.0	5.0	3.1	1.9	38

(ii) Dose of Adsorbent (*Murraya koenigii*)

S. No.	*wt of adsorbent (gm)*	*Amount of water 5.0 ppm (ml)*	*initial concentration before treatment (ppm)*	*after 5.0 hrs (ppm)*	*ppm removal*	*% Fluoride Removal*
1	0.2500gm	100 ml	5.00	2.8	2.20	44
2	0.5006 gm	100 ml	5.00	2.4	2.60	52
3	1.008 gm	100 ml	5.00	2	3.00	60
4	2.0118 gm	100 ml	5.00	1.7	3.30	66

(iii) Effect of Contact Time

(a)

Dose Gm/100ml	*Contact Time (Min)*	*Initial Conc*	*Final Fluoride (Mg/L)*	*Reduction of Fluoride*	*% Removal Efficiency*
0.251	30	5.00	4.4	0.60	12
0.251	60	5.00	3.5	1.50	30
0.251	120	5.00	3.2	1.80	36
0.251	180	5.00	3	2.00	40
0.251	240	5.00	2.8	2.20	44
0.251	300	5.00	2.7	2.30	46

(b)

Dose Gm/100ml	*Contact Time (Min)*	*Initial Conc.*	*Final Fluoride (Mg/L)*	*Reduction of Fluoride*	*% Removal Efficiency*
0.5012	30	5.00	4.2	0.80	16
0.5012	60	5.00	3.2	1.80	36
0.5012	120	5.00	3	2.00	40
0.5012	180	5.00	2.7	2.30	46
0.5012	240	5.00	2.5	2.50	50
0.5012	300	5.00	2.5	2.50	50

(c)

Dose Gm/100ml	*Contact Time (Min)*	*Initial Conc.*	*Final Fluoride (Mg/L)*	*Reduction of Fluoride*	*% Removal Efficiency*
1.0118	30	5.00	4	1.00	20
1.0118	60	5.00	2.8	2.20	44
1.0118	120	5.00	2.5	2.50	50
1.0118	180	5.00	2.2	2.80	56
1.0118	240	5.00	2.2	2.80	56
1.0118	300	5.00	2.1	2.90	58

(d)

Dose Gm/100ml	*Contact Time (Min)*	*Initial Conc.*	*Final Fluoride (Mg/L)*	*Reduction of Fluoride*	*% Removal Efficiency*
2.01	30	5.00	3.8	1.20	24
2.01	60	5.00	2.6	2.40	48
2.01	120	5.00	2.4	2.60	52
2.01	180	5.00	2	3.00	60
2.01	240	5.00	2	3.00	60
2.01	300	5.00	1.9	3.10	62

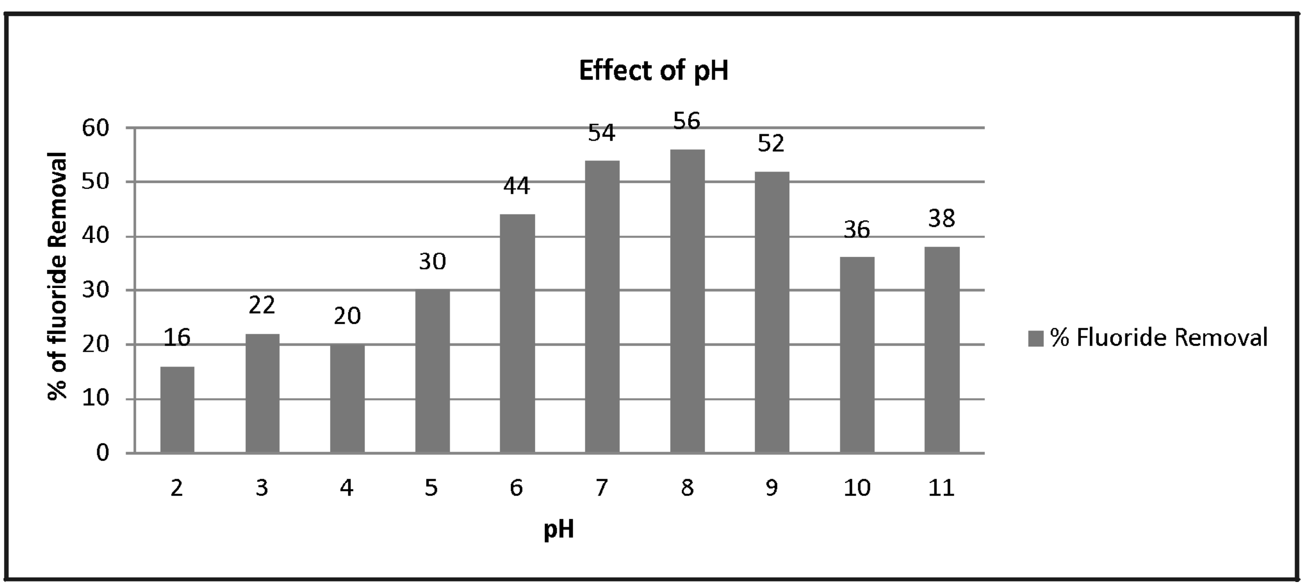
(iv) Effect of Temperature

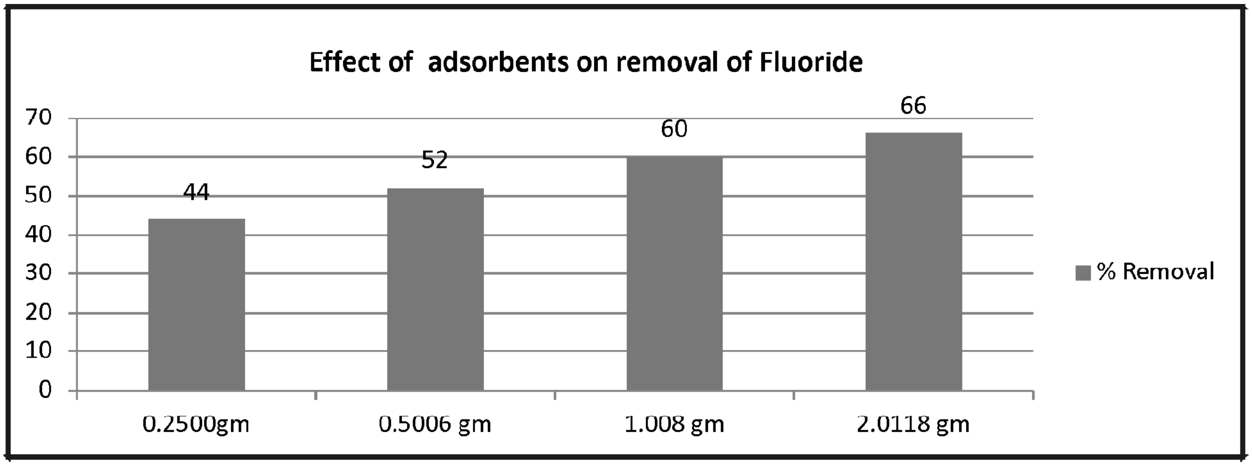
Dose (g/100ml)	*Temperature (in C)*	*Initial fluoride (mg/l)*	*Final fluoride (mg/l)*	*Reduction in fluoride (mg/l)*	*% removal efficiency*
1.0	22	5.0	2.4	2.6	52
1.0	30	5.0	2	3.0	60
1.0	40	5.0	1.9	3.1	62
1.0	50	5.0	1.8	3.2	64

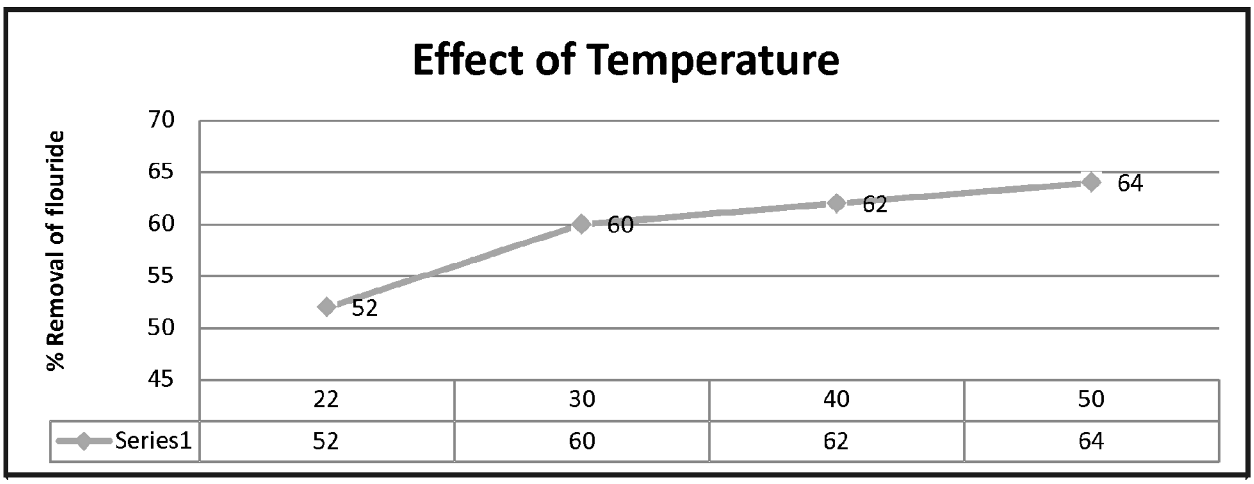
(v) Adsorption Isotherm

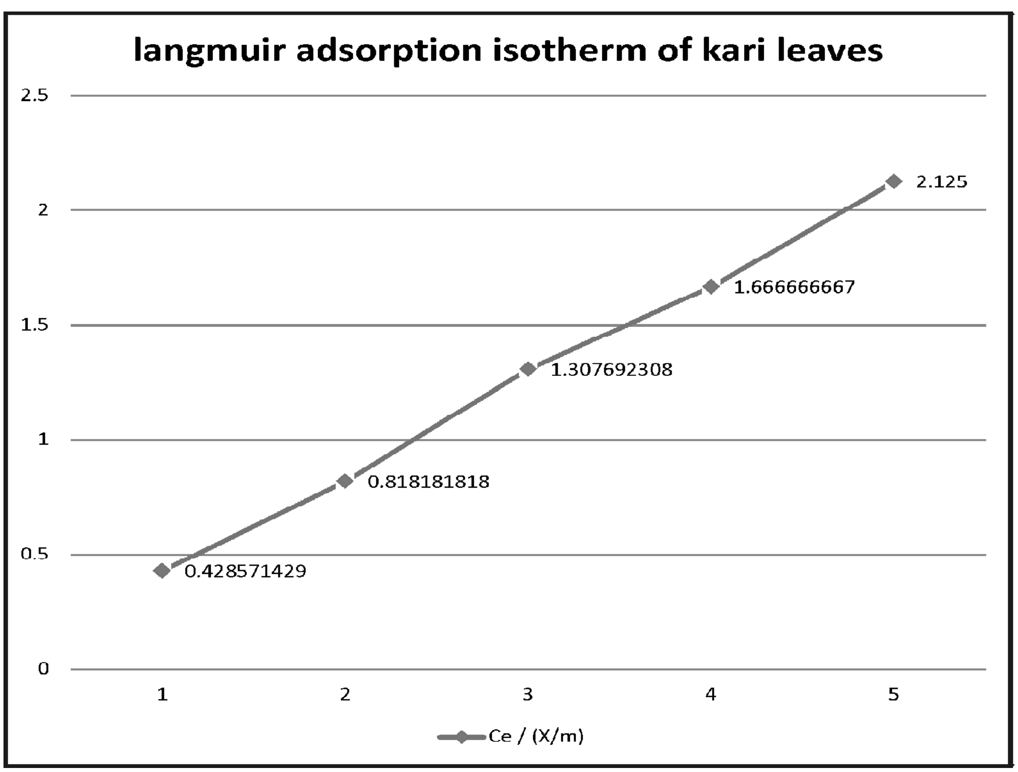
Adsorption isotherm of Curry leaves (Murraya koenigii)

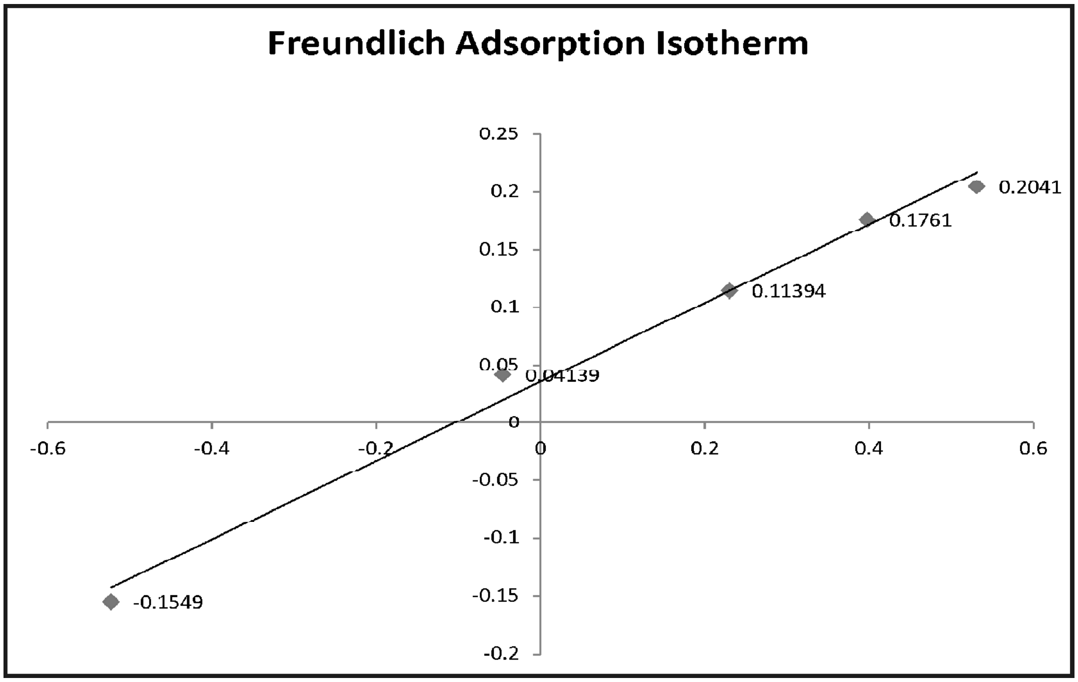
Ci(mg/l)	*Ce (mg/ l)*	*X (mg)*	*m (g)*	*X/m (mg/g)*	*Ce / (X/m)*	*log Ce*	*log X/m*
1	0.3	0.7	1.0	0.7	0.429	-0.5228	-0.1549
2	0.9	1.1	1.0	1.1	0.818	-0.04575	0.04139
3	1.7	1.3	1.0	1.3	1.308	0.2304	0.11394
4	2.5	1.5	1.0	1.5	1.667	0.3979	0.1761
5	3.4	1.6	1.0	1.6	2.125	0.5314	0.2041




Effect of adsorbents on removal of Fluoride
70
60
50
40
30
20
10
0
44
52
60
66
0.2500gm
0.5006 gm
1.008 gm
2.0118 gm
% Removal


Effect of Temperature
% Removal of flouride
70
65
60
55
50
45
52
60
62
64
22
30
40
50
Series1
52
60
62
64


langmuir adsorption isotherm of kari leaves
2.5
2
1.5
1
0.5
0
0.428571429
0.818181818
1.307692308
1.666666667
2.125
1
2
3
4
5
Ce / (X/m)



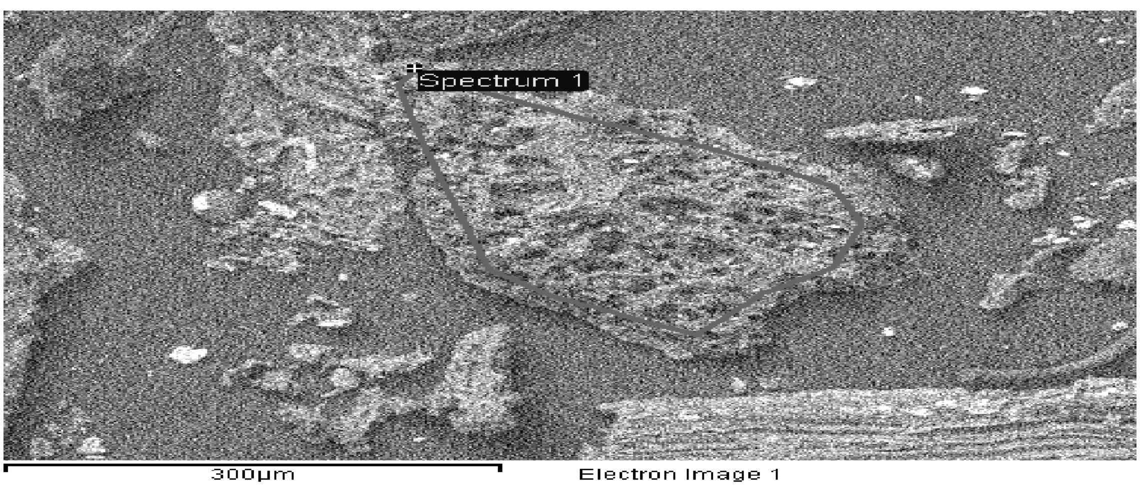


Figure 1.1

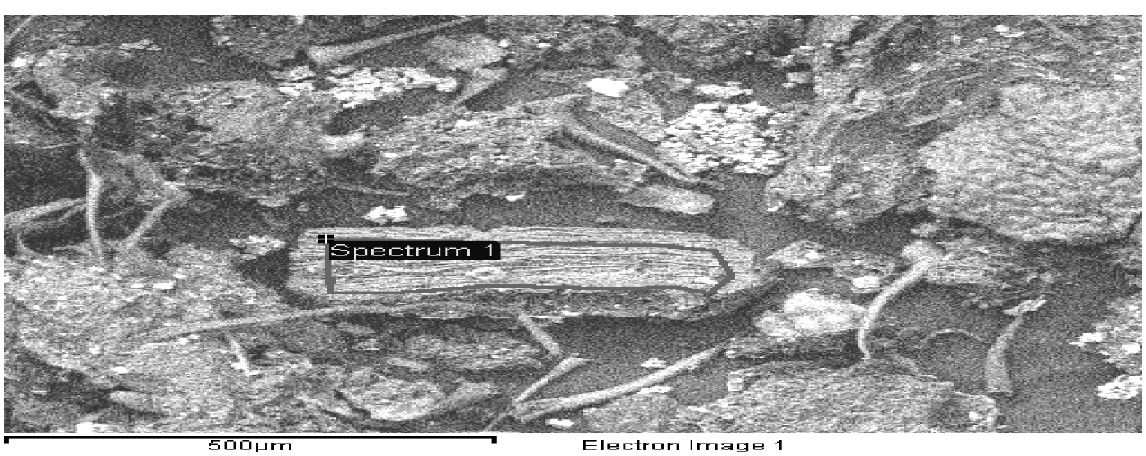


Figure 1.2

Spectrum processing : No peaks omitted

Processing option: All elements analyzed (Normalised)

Number of iterations = 5 Standard :

C CaCO3 1-Jun-1999 12:00 AM

O SiO2 1-Jun-1999 12:00 AM

F MgF2 1-Jun-1999 12:00 AM

Na Albite 1-Jun-1999 12:00 AM

Mg MgO 1-Jun-1999 12:00 AM

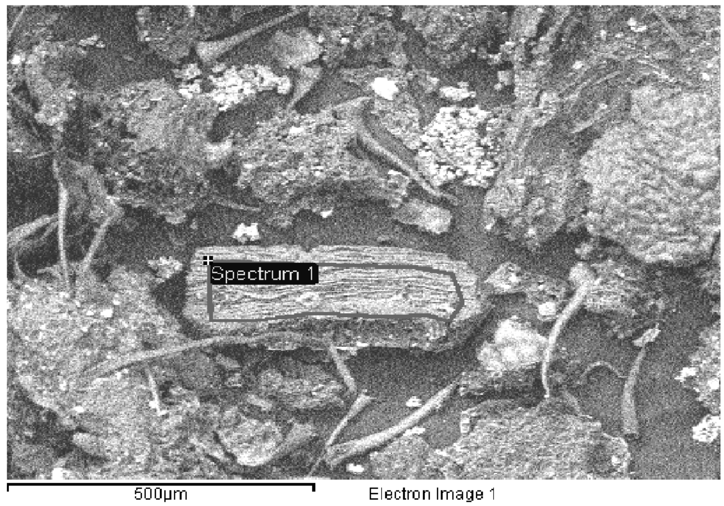
Al Al2O3 1-Jun-1999 12:00 AM

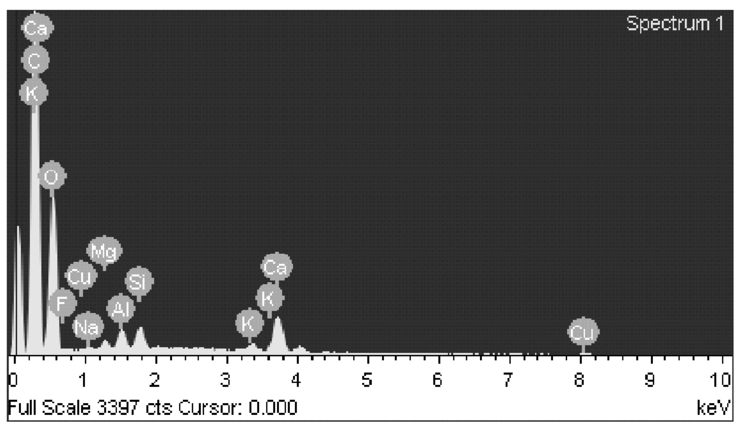
Si SiO2 1-Jun-1999 12:00 AM

K MAD-10 Feldspar 1-Jun-1999 12:00 AM

Ca Wollastonite 1-Jun-1999 12:00 AM

Comment: EDS on Sample C2





Element	Weight %	Atomic%
C K	49.67	58.18
O K	44.51	39.14
F K	0.69	0.51
Na K	0.18	0.11
Mg K	0.43	0.25
Al K	0.77	0.40
Si K	0.86	0.43
K K	0.35	0.13
Ca K	2.17	0.76
Cu K	0.35	0.08
Totals	100.0	

inca

Paras Tak EDS 21.7.16 7/25/2016 12:55:53 PM