

Adsorption of Fluoride from Aqueous Solution Using *Limonia Acidissima* as Adsorbent

G. Anusha¹, J. Raja Murugadoss² and P. Raju³

^{1&2}Department of Civil Engineering, Bannari Amman Institute of Technology, Sathyamangalam - 638 401
Erode District, Tamil Nadu, India.

³Department of Civil Engineering, Vetrivinaiyaga College of Engineering and Technology,
Trichy - 621 215, Tamil Nadu, India.

(Received on 01 October 2011 and accepted on 18 March 2012)

Abstract - In recent years the problem of water contaminant is drastically increasing due to the disposal of industrial wastewater containing iron, fluoride, mercury, lead, cadmium, phosphorous, silver etc into water bodies. Due to which, the contamination leads to serious health disorders and in some worst cases it causes permanent disabilities or even casual losses. In the present work an attempt has been made to remove such contaminants particularly fluoride and to study the efficiency of the removal of fluoride using a new material *Limonia Acidissima* which is commonly referred as wood apple. Accordingly a set of experiments have been conducted using batch and column processes, with the help of activated carbon prepared from the shell of wood apple. Experiments reveal that the adsorption capacity of the shell of wood apple is significant as 64% to yield promising solutions.

Keywords: Fluoride, Activated Carbon, Wood Apple Shell, Batch Study, Column Study

1. INTRODUCTION

Fluoride is a natural constituent of the Earth's crust and is present in varying concentrations in all ecosystems. It has drastically changed the biogeochemical cycles and in few cases it balances few classes of metals. The main anthropogenic sources of fluoride are industrial sources viz. tooth powder manufacturing industries and groundwater. In addition it also induces adverse effects on the environment and human health and moreover they are highly toxic even at low concentration to human beings and other living beings [1].

The removal of fluoride from synthetic solution can be done by adsorption technique using powdered activated carbon. Through the importance of this treatment is felt, its cost keeps the industrialists away from adopting the same [2,3,4,5]. It is also a fact that till date no material proves to be a better adsorbent than commercial activated carbon. Since the cost of activated carbon and its reactivation costs are high, this particular technology remains at an inaccessible distance and its application does not find a significant place in the field of environmental engineering. Hence in this present work an effort has been made to introduce a new class of activated carbon, prepared from the shell of wood apple for the removal of such fluoride content from wastewater.

During adsorption, the atoms within the structure of the adsorbent are attracted in all directions relatively equally, whereas the atoms at the surface exhibit an imbalanced attractive force, which the adsorbate molecules help to satisfy [6,7]. The significant parameter, which popularizes an adsorbent, is the presence of a great amount of surface area; normally via the wall area or slots, capillaries of pores permeating its structure, in a very small volume and weight.

II. MATERIALS AND METHODS

A. Adsorption with Activated Carbon

Certain organic compounds in wastewater are resistant to biological degradation and many others are toxic or nuisance (odor, taste, color forming) even at low concentration. Low concentration materials are not readily removed by conventional treatment methods. Activated carbon has an affinity for organics and its use for organic contaminant removal from wastewater [8],[9],[10].

The larger surface area, a critical factor in the adsorption process, enhances the effectiveness of the activated carbon for the removal of organic compounds from wastewater by adsorption [11]. The surface area of activated carbon typically can range from 500 - 1400 m²/g. The effect of chemical nature of the carbon's surface is less significant than the surface area. This chemical nature or polarity varies with the carbon type and can influence attractive force between molecules.

B. Experimental Methodology: Preparation of Adsorbent Using Wood Apple

The shell of wood apple as in Fig. 1 is a natural material which is thrown out as a waste. These wastes were collected and their size was reduced by breaking it into small size. Then they were dried in an oven at a temperature of 170°C for 24 hrs. It was then packed in an air tight cylindrical iron container with top completely sealed with an iron cover to prevent the entry of air during the process of charring. The sealed iron container was heated in a muffle furnace by slowly raising the temperature up to 600°C and subsequently it is maintained for the duration of one hour. The activated carbon thus prepared was subsequently washed with distilled water and oven dried.



Fig. 1a Shell of Wood Apple



Fig. 1b Activated Carbon from Shell of Wood Apple

C. Experimental Methodology: Batch Adsorption Experiments and Column Adsorption Studies

Batch adsorption study was made by employing arbitrary shakers. 150 ml of desired concentration were agitated continuously with known amount of activated carbon by varying the dosage, Samples were taken and their residual fluorides were monitored at regular intervals. After a lapse of a prefixed contact time, 50 ml sample was drawn from the samples, filtered and analyses for residual fluoride concentration.

To assess the influence of optimum pH, optimum dosage, time interval and isotherm, four different set of experiments were carried out, viz. a) Bottles with different pH ranging from 1 to 12 were varied and conditions were analysed for residual fluoride concentration. b) Bottles with increasing doses ranging from 0.5g to 5 g of the adsorbent with fixed contact time were analysed. c) Bottles with optimized pH and sorbent dosage were agitated for varied timings ranging from 5 to 40 minutes and then analysed for residual fluoride concentration. d) Langmuir and Freundlich isotherms were plotted and the isotherm satisfies the equilibrium condition of the adsorbent.

Further the laboratory experiments were conducted with 45 mm diameter and 300 mm length column setup. The studies were carried out for a fixed flow rate of 2 mL/min and for which the depths of carbon are taken as 2.5 and 5 cm. The column was packed with adsorbents at various depths. The influent and effluent were adjusted to be 2 mL/min. Samples were collected at regular interval of 15 min and analyzed for residual concentration. The percentage removal was calculated till the carbon gets exhausted. The experiments were conducted for all the adsorbents and the results obtained were compared. The results were plotted taking volume treated in mL as abscissa and residual metal concentration in mg/L as ordinate.

III. RESULTS AND DISCUSSIONS

With reference to the discussion in the above section nine experiments were successfully conducted to study the influence of pH on the efficiency of fluoride removal. Accordingly the pH was varied with an increment of 2 for a range of 9 to 12. From the experiments it is found that the hydrogen ion concentration plays a critical role on the adsorption capacity of wood apple shell. It is found that for a pH of 7, the efficiency was found to be better as in Fig. 2. Further another set 9 experiments were conducted to determine the influence of dosage on the removal efficiency. Dosage was varied from 0.025g to 0.6g with an interval of 0.025g and it has been found that the dosage variation plays a significant role in determining the removal efficiency. It is found that for a dosage of 1.5g, the removal efficiency has an appreciable increase as in Fig.3 Similarly 8 set of experiments were conducted and the influence of time interval for fluoride removal was determined by varying the time interval from 5 minutes to 40 minutes with an interval of 5 minutes. It has been found that time also plays an important role in addition to pH and dosage as in Fig. 4. By varying the time interval it was found that at the contact time of 15 minutes, the removal efficiency has an appreciable increase. From the results obtained from the laboratory experiments a Langmuir isotherm and a Freundlich isotherm was plotted to generate the adsorption pattern as in Figs. 6 and 7.

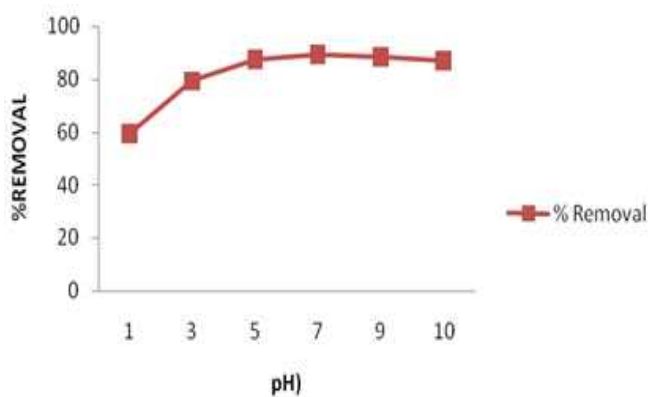


Fig. 2 Studies on pH Value

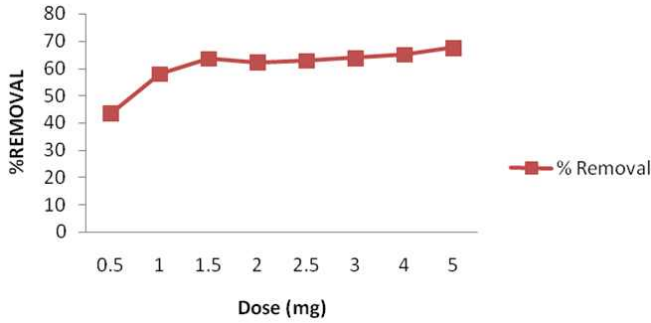


Fig. 3 Studies on Optimum Dosage of Activated Carbon prepared from Shell of Wood Apple

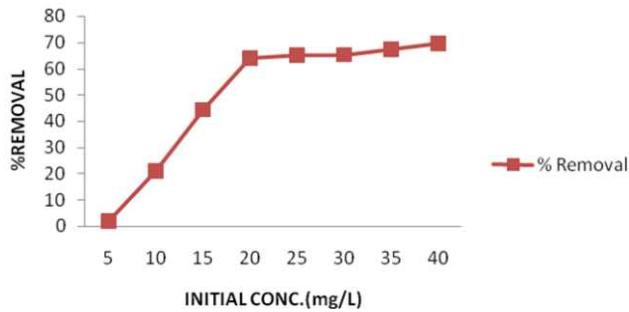


Fig. 4 Studies on Optimum Contact Time of Adsorbent with Adsorbate

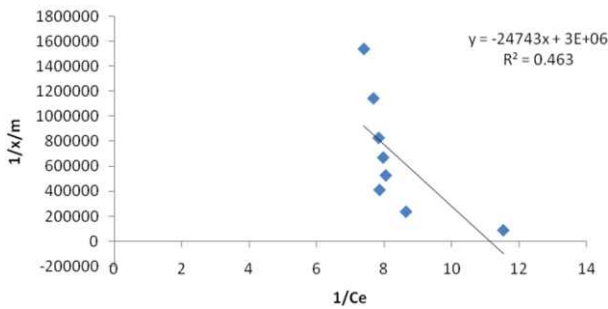


Fig. 5 Langmuir Isotherm

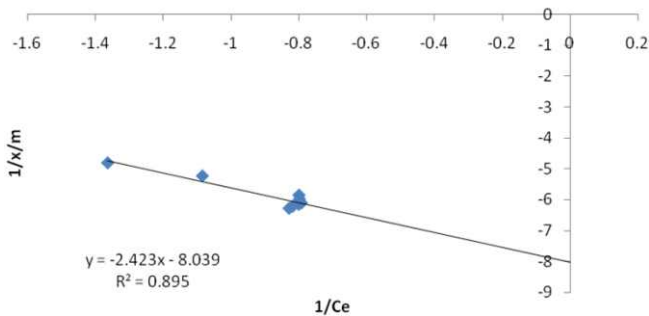


Fig. 6 Freundlich Isotherm

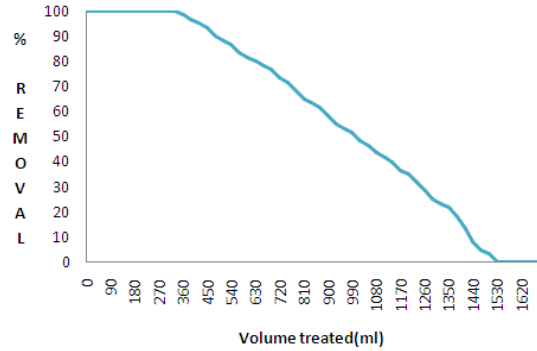


Fig. 7 Column Break through Curve for 2.5cm bed depth of activated carbon

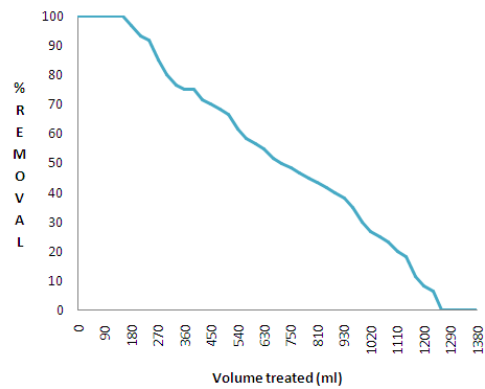


Fig. 8 Column Break through Curve for 5.0 cm bed depth of activated carbon

IV. CONCLUSION

The efficiency of a new variety of adsorbent prepared from the seeds of Limonia Acidissima has been successfully demonstrated through series of laboratory experiments by varying the values of pH, dosage and time for the synthetic solution. The experiments were carried out for fluoride removal and the results reveal that the shellof Limonia Acidissima has much potential for a pH of 7, dosage of 1.5g and contact time of 15 minutes.

ACKNOWLEDGMENT

The authors would like to thank the Management of Bannari Amman Institute of Technology for providing the necessary laboratory facilities to carry out this research work.

REFERENCES

- [1] D.P. Tiwari, D. K. Singh and D. N. Saksena, "Hg (II) Adsorption from Aqueous Solutions using Rice Husk Ash", *Journal of Environmental Engineering*, pp.479-481, June 1995.
- [2] K.P. Rakhi Gopalan, "Removal of Hexavalent Chromium- Chrome Plating Effluent Treatment using Tapioca Peel Carbon", *Enviro*, pp.51-58, Sep-Nov 2007.
- [3] F. Hernainz, M. Calero, G. Blazquez, G. Tenorio and M. A Martin-Lara., "Effect of the Presence of Chromium (III) on the Removal of Lead (II) from Aqueous Solutions by Agricultural Wastes", *Journal of Environmental Engineering*, pp.1348-1356, Dec. 2009.
- [4] Akbar Eskandarpour, Maurice S. Onyango, Aoyi Ochieng, Shigeo Asai, "Removal of fluoride ions from aqueous solution at low pH using schwertmannite", *Journal of Hazardous Materials*, Vol.152, pp.571-579, 2008.
- [5] Feng Shen, Xueming Chen, Ping Gao, Guohua Chen, "Electrochemical removal of Fluoride ions from industrial wastewater", *Chemical Engineering Science*, Vol.58, pp.987-993, 2003.
- [6] H. S. Ashoka, S. S. Inamela, "Adsorption Removal of Methyl Red from Aqueous Solution with Treated Sugarcane Bagasse and Activated Carbon", *Environmental Pollution Control Journal*, Vol 14, Nov-Dec 2010.
- [7] Monika Dakeshene, "Adsorption Removal of Tartiazine (E102) Over Alkali Activated Fly Ash", *Asian Journal of Chemistry Asian Journal of Chemistry*, Vol 22, pp.7718-7732, 2010.
- [8] Sarita Sinha, Kavita Pandey, Dinesh Mohan, and Kunwar P. Singh, "Removal of Fluoride from Aqueous Solutions by Eichhornia crassipes Biomass and Its Carbonized Form", *Ind. Eng. Chem. Res.*, Vol.42, pp.6911-6918, 2003.
- [9] P.I. Dergunov, A.V. Tvardovski, A.V. Klinger, A.A. Fomkin, "Modeling of the Adsorption Deformation of Microporous Adsorbents Interacting with Gases and Vapours", *Russian Physics Journal*, Vol. 48, pp.11, 2005.
- [10] Datsko. T. Ya., V.I. Zelentsov, "Dependence of the Surface Charge and the Fluorine Adsorption by γ -Aluminum Oxide on the Solution Temperature", *Surface Engineering and Applied Electrochemistry*, Vol. 45 No. 5, pp.404-410, 2009.
- [11] Ramanathan Natarajan, Jatin Sharma, Inderjit Nirdosh., "Adsorption of N- hydrocinnamoyl-N-phenyl hydroxylamine on pure minerals", *Adsorption*, Vol.16, 541-548, 2010