

ABSTRACT

High fluoride concentration is a worldwide problem in drinking water due its health effects. In India large population is mainly belong to rural areas which depend on ground water for their drinking purpose. The fluoride concentration in ground water varies from place to place. The data show that the fluoride distribution in ground water varies from 0.01mg/l to 48 mg/l [2]. The fluoride comes into ground water by various ways, for example, weathering of rocks, industrial effluents and geochemical reactions. Traces of fluoride in minute amounts is an essential component for bones and for the formation of dental enamel in animals and human but its high concentration more than 1.5 mg/l causes irreversible demineralization of bones and tooth tissues i.e. skeletal and dental fluorosis, damage to the brain, harmful effects on kidney and liver, headache, skin rashes, bone cancer and even death in extreme cases. Various treatment technologies for removing fluoride from groundwater have been investigated. Adsorption process for defluoridation has favoured for the most part in developing countries as it is techno-economical viable method, environmental friendly and straight forwardness in operation. Therefore it is require to observe defluoridation by cheap and easy method. This paper review about the defluoridation by a natural absorbent i.e. coconut husk which is cheap and effective and gives perfect removal of fluoride in drinking water upto 86%. For higher removal percentage efficiency of fluoride, optimal dose of adsorbent was also determined. From the results obtained it can be concluded that Coconut husk can become a cheap alternative for defluoridation of water.

KEYWORDS: Defluoridation, adsorption, low-cost adsorbent, treatment and reuse of water, eco-friendly.

INTRODUCTION

Water is an essential natural resource for sustaining life and environment that we have always thought to be available in abundance and free gift of nature. However, chemical composition of surface or subsurface is one of the prime factors on which the suitability of water for domestic, industrial or agricultural purpose depends. Freshwater occurs as surface water and groundwater. Though groundwater contributes only 0.6% of the total water resources on earth, it is the major and the preferred source of drinking water in rural as well as urban areas, particularly in the developing countries like India because treatment of the same, including disinfection is often not required. It caters to 80% of the total drinking water requirement and 50% of the agricultural requirement in rural India. But in the era of economic growth, groundwater is getting polluted due to urbanization and industrialization.

Fluoride contaminated ground water is creating problems sixty-two million people including 6 million children in the country in 17 states are affected with dental, skeletal and non-skeletal fluorosis [3]. The extent of fluoride contamination in the ground water varies from 1.0 to 48.0 mg/l in Rajasthan.[4] Fluoride is one of the most important elements for both human and animal health. Within the permissible limit of 0.5 – 1.5mg/L, fluoride serves to maintain healthy teeth and bones. On the other hand, elevated levels of fluoride (>1.5mg L⁻¹) can cause long-term diseases such as dental (1.5 – 4.0mg/L) and skeletal fluorosis (4.0 – 10.0 mg/L). The most seriously

affected states in India are Andhra Pradesh, Punjab, Haryana, Rajasthan, Gujarat, Tamil Nadu and Uttar Pradesh. The highest concentration observed to date in India is 48 mg/l in Rewari District of Haryana[10].

DE fluoridation of water was done by several technique such as coagulation and precipitation, membrane separation, ion exchange and electrolytic deposition, but found expensive and non-suitable for developing countries [6-7]. Adsorption is a low cost technique used for effective removal of fluoride from drinking water through local adsorbent materials. The biomass from plants and agricultural wastes by-products can be used for efficient fluoride uptake as well as solving their disposal problem. These low cost materials are helpful in replacing the expensive commercial adsorbent like activated carbon that has again problem in regeneration.

EFFECTS OF FLUORIDE

Fluorine is the most electronegative element and it is always present in a combined state as fluoride because of its high chemical reactivity. The fluoride is a great calcium-seeking element and it can disturb the calcified structure of bones and teeth in the human body at higher concentration resulting dental fluorosis or skeletal fluorosis similar kinds of effects are found in animal body parts also

ADVANTAGES OF ADSORPTION PROCESS

Batch adsorption experiments were conducted to examine time-dependent sorption behavior and the effects of temperature and solution pH on adsorption performance.

Cheap: The cost of adsorbent is low since they are often made from locally, abundantly and easily available materials.

No Sludge Generation: Unlike the problems in other techniques (ex: precipitation), there is no issue of sludge generation in adsorption process.

Competitive Performance: Performance of adsorption process in terms of efficiency and cost is comparable with the other methods available.

ABOUT THE STUDY

The aim of the present study was to examine the feasibility of using low cost natural absorbents such as coconut husk, in Fluoride removal using adsorption technique. Adsorption characterization was done by using spectrophotometer. The effect of adsorbent dosage, contact time, pH, initial fluoride concentration and shaking speed were determined.

PRACTICAL EXPERIENCES OF THE RELATED WORK

The Nalgonda technique, based on precipitation processes, is also a common defluoridation technique. The limitations of the process are, daily addition of chemicals, large amount of sludge production, and low effectiveness for water having high total dissolved solids and hardness. Ion exchange methods are efficient for fluoride removal, but a tedious and difficult process of preparation of resins as well as the high cost necessitates a search for an alternative technique. Adsorption methods can be implemented for the removal of fluoride due to physical, chemical, or ion exchange interactions with the adsorbents. Two types of contacting systems of adsorption are usually encountered, namely, the batch and fixed-bed processes. Batch type processes are usually limited to the treatment of small volumes of effluents whereas the bed column systems have the advantage of continuous operation up to the point of saturation. Adsorbent is a porous substance that has a high surface area and has the ability to absorb or adsorb other substances using intermolecular forces onto its surface.

Mohammad Mehdi Mehrabani Ardekani, Roshanak Rezaei Kalantary, Sahand Jorfi, Mohammad Nurisepehr (2013) compared the efficiency of Bagasse, Modified Bagasse and Chitosan for fluoride removal from water by adsorption. The pH value of 7, contact time of 60 min and adsorbent dose of 2 g/L were determined as optimum conditions for all three adsorbents. Chitozan and bagasse did not show good capability for fluoride removal, but modified bagasse showed more than 90% removal at optimized conditions, including the pH value of 7, contact time of 60 min and adsorbent dosage of 2 g/L. Both Langmuir and Freundlich isotherms show good correlation for description of results, but the Langmuir model with the correlation value of 0/99 is superior.

Joshi *et al.* (2012) have studied fluoride removal capability of zirconium impregnated lapsi seed stone activated carbon (ZILSSSAC). The optimal condition of fluoride removal was achieved at pH 3-4, contact time of 180 minutes and adsorbent dosage of 2.0 g/L for initial fluoride concentration of 10 mg/l. At pH 3, the maximum uptake of fluoride was observed to be 3.25 mg/g and at neutral condition the adsorption capacity was 1.6 mg/g.

Rajan *et al.* (2013) have studied the fluoride removal capacity of zirconium impregnated walnut shell carbon (ZIWSC). The fluoride removal of ZIWSC and WSC were 94% and 81% at pH 3 respectively. The Langmuir maximum adsorption capacity of ZIWSC was 3.19 mg/g at 303K for optimum condition of 180 minutes of contact time size < 53 µm and dosage of 1.5 gm/100 ml. The pseudo-second order was best fitted as per kinetic study. The thermodynamics study revealed that the process of adsorption was endothermic and bicarbonate ions reduced the removal efficiency by 85.67% to 54.94%.

S. T. Ramesh, R. Gandhimathi, P. V. Nidheesh and M. Taywade (2012) investigated the adsorption potential of bottom ash for defluoridation of drinking water using batch and continuous fixed bed column studies. The optimum contact time for fluoride was found to be 105 minutes with the maximum efficiency of 73.5 % at 70mg/100ml bottom ash dosage. The optimum pH was found to be pH 6 with the maximum efficiency of 83.2 %. During the column studies; increase in fluoride ion uptake with an increase in the bed height was due to an increase in the contact time. A high degree of linearity of the BDST plot indicates the validity of the BDST Model when applied to continuous column studies.

Alagumuthu *et al.* (2010) have investigated fluoride removal capacity of zirconium impregnated groundnut shell carbon (ZIGNSC) from water. The fluoride evacuations of ZIGNSC and ground nut shell carbon (GNSC) were 84% and 63.67% respectively. The optimum contact time for maximum removal was 180 minutes for initial fluoride concentration of 3 mg/l at dosage of 2.0 mg. The adsorption capacity of ZIGNSC was 2.32 and 2.50 mg/g at temperature of 303K and 333K respectively. The kinetic study followed the pseudo-second-order equation. The bicarbonate ions was responsible for decrease of fluoride removal from 84% to 74.6% [15].

EXPERIMENTAL WORK

MATERIALS AND METHODS

Preparation of Adsorbent:

Adsorbent of Coconut husk:

The adsorbent was prepared as described by Hanafiah *et al.* (2006). The coconut husk was cut into small pieces and blended, washed with distilled water to remove dirt and colour and air-dried for 24 h to avoid thermic deactivation of the adsorbent surface. It was sieved to pass through a 2 mm stainless steel endecott sieve and a portion of this was stored in clean polyethylene containers (labeled as unmodified adsorbent) prior to analysis. For the modification of the adsorbent, about 400 g of the washed adsorbent was mixed with 600 ml of 0.1 mol dm⁻³ NaOH. The mixture was heated at 120 °C for 30 min with occasional stirring. The powdered coconut husk was separated using a Buckner funnel and a vacuum pump and washed with distilled water until the washings were free of color and the pH of wash solution was 7. The washed adsorbent was air-dried for 24 h and labeled as NaOH- modified coconut husk. The HCl modification was also carried out using the same procedure as the NaOH modification but with 0.1 mol dm⁻³ HCl.

Effect of Adsorbent dose:

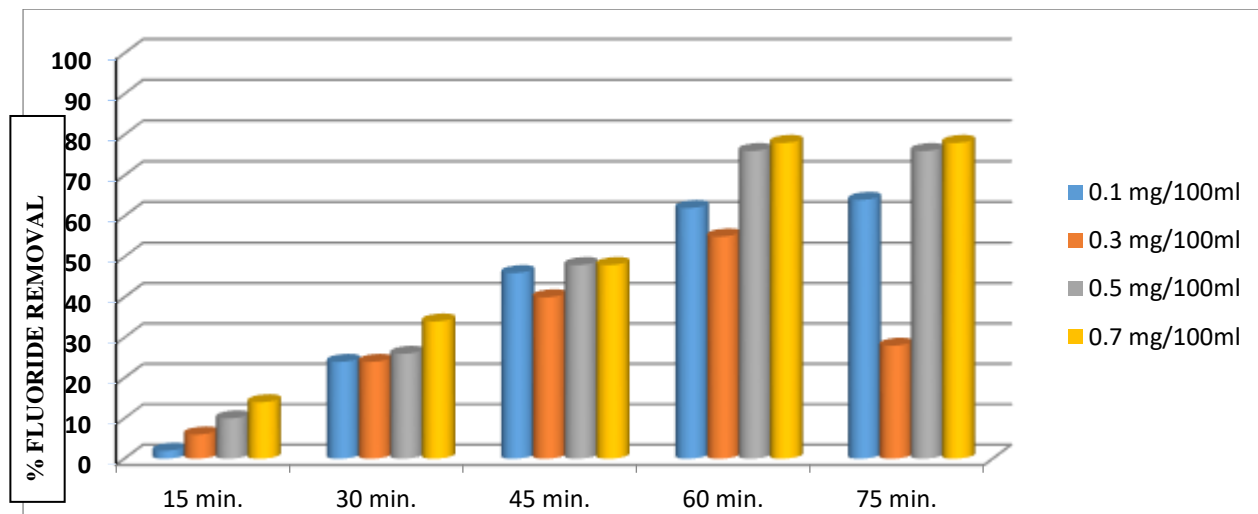
The effect of the adsorbent dose on the removal of fluoride was studied by varying the dose from 0.1g/100ml to 0.7g/100ml. The results are presented, where it can be seen that maximum removal of about 78% is found at an adsorbent dose of 0.7 g/100ml and thereafter the percent removal became more or less constant.

Dose(g/100ml)	Contact time (min)	Initial fluoride (mg/l)	Final fluoride(mg/l)	Reduction in fluoride (mg/l)	% removal efficiency
0.1	15	5	4.9	0.1	02
0.1	30	5	3.8	1.2	24
0.1	45	5	2.7	2.3	46
0.1	60	5	1.9	3.1	62
0.1	75	5	1.8	3.2	64

Dose(g/100ml)	Contact time (min)	Initial fluoride (mg/l)	Final fluoride(mg/l)	Reduction in fluoride (mg/l)	% removal efficiency
0.3	15	5	4.7	0.3	06
0.3	30	5	3.8	1.2	24
0.3	45	5	3.0	2.0	40
0.3	60	5	2.3	2.7	55
0.3	75	5	1.6	1.4	28

Dose(g/100ml)	Contact time (min)	Initial fluoride (mg/l)	Final fluoride(mg/l)	Reduction in fluoride (mg/l)	% removal efficiency
0.5	15	5	4.5	0.5	10
0.5	30	5	3.7	1.3	26
0.5	45	5	2.6	2.4	48
0.5	60	5	1.2	3.8	76
0.5	75	5	1.2	3.8	76

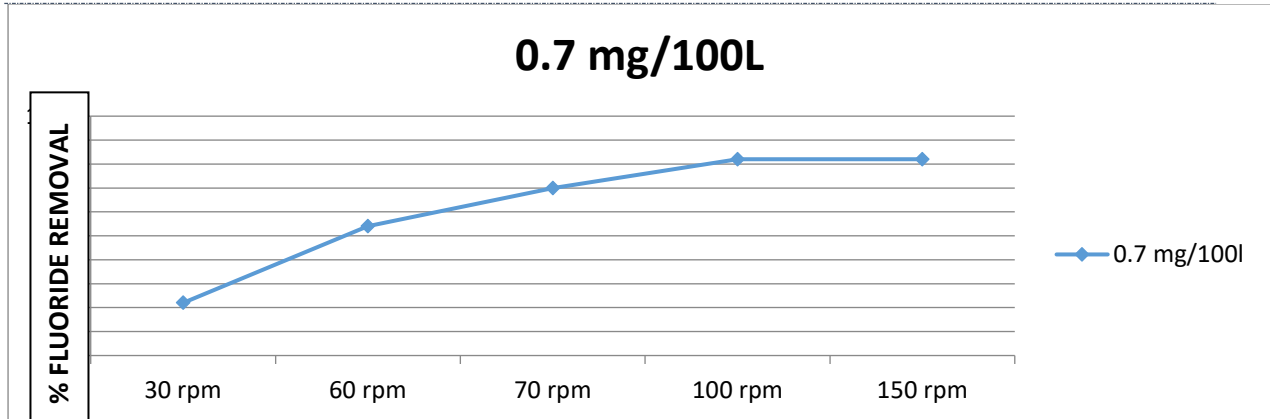
Dose(g/100ml)	Contact time (min)	Initial fluoride (mg/l)	Final fluoride(mg/l)	Reduction in fluoride (mg/l)	% removal efficiency
0.7	15	5	4.3	0.7	14
0.7	30	5	3.3	1.7	34
0.7	45	5	2.6	2.4	48
0.7	60	5	1.1	3.9	78
0.7	75	5	1.1	3.9	78



Effect of Stirring rate

The effect of the stirring rate on the removal of fluoride was studied by varying the stirring rate from 30 to 150 rpm. The results are presented, where it can be seen that maximum removal of about 82% is found at a stirring rate of 150 rpm and thereafter the percent removal became more or less constant.

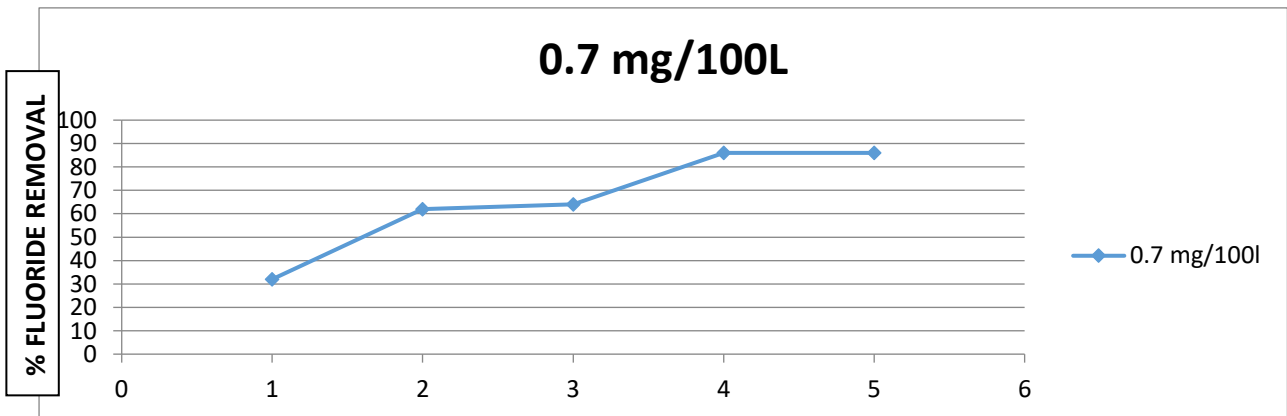
Dose(g/100ml)	Stirring rate (rpm)	Initial fluoride (mg/l)	Final fluoride(mg/l)	Reduction in fluoride (mg/l)	% removal efficiency
0.7	30	5	3.9	1.1	22
0.7	60	5	2.3	2.7	54
0.7	70	5	1.7	3.3	70
0.7	100	5	0.9	4.1	82
0.7	150	5	0.9	4.1	82



Effect of Temperature

The effect of the temperature on the removal of fluoride was studied by varying the temperature from 283 to 313 K. The results are presented, where it can be seen that maximum removal of about 86% is found at a dose of 0.9 g/L at a temperature of 313K and thereafter the percent removal became more or less constant.

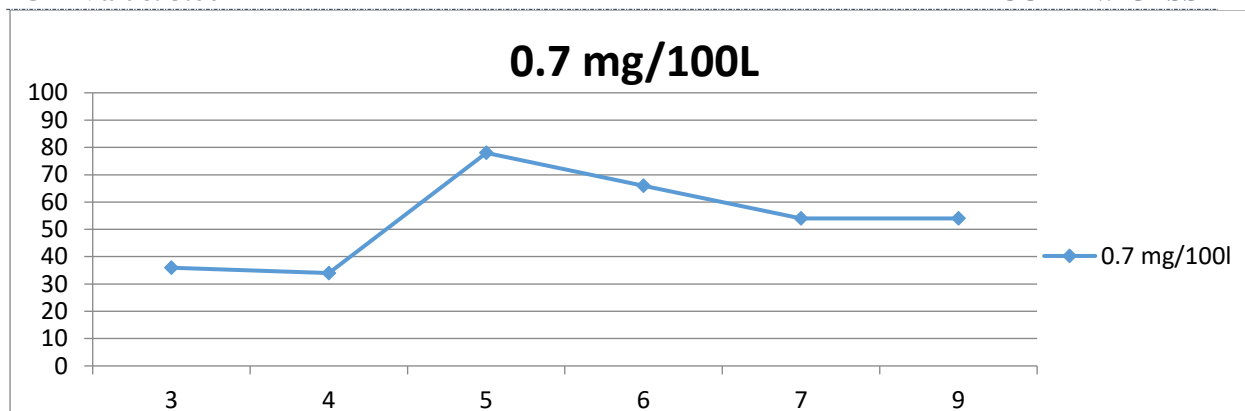
Dose(g/100ml)	Temperature (in K)	Initial fluoride (mg/l)	Final fluoride(mg/l)	Reduction in fluoride (mg/l)	% removal efficiency
0.7	283	5	3.4	1.6	32
0.7	293	5	2.9	3.1	62
0.7	303	5	1.8	3.2	64
0.7	313	5	0.7	4.3	86
0.7	323	5	0.7	4.3	86



Effect of pH

The effect of the pH on the removal of fluoride was studied by varying the pH from pH 3 to 9. The results are presented, where it can be seen that maximum removal of about 78% is found at a dose of 5 g/L at 5pH of sample and thereafter the percent removal became more or less constant.

Dose(g/100ml)	pH	Initial fluoride (mg/l)	Final fluoride(mg/l)	Reduction in fluoride (mg/l)	% removal efficiency
0.5	3	5	3.2	1.8	36
0.5	4	5	3.3	1.7	34
0.5	5	5	1.1	3.9	78
0.5	6	5	1.7	3.3	66
0.5	7	5	2.3	2.7	54
0.5	9	5	2.3	2.7	54



CONCLUSION

Based on the results of this study, it can be concluded that coconut husk have good performance to adsorb fluoride from drinking water especially for high concentration of fluoride and had given an excellent results. The biosorbent was successful in removal of fluoride ions from aqueous solution of 0.7mg/l fluoride concentration with about 86% efficiency at 323K temperature. It was also observed that the adsorption was pH dependent with maximum adsorption achieved at pH 5.0 with 78% efficiency.

REFERENCES

- [1] Mohammed Arif tikki (2014) Fluoride Removal from water – a review International journal of scientific & engineering research volume -5 issue 1, January.
- [2] Priya goswami et al (2015) defluoridation of water using low cost adsorbent International journal of chemical studies.
- [3] Kumar S., Gupta A. and Yadav J.P. (2008): 29(2) 227-232 Removal of fluoride by thermally activated carbon prepared from neem (*Azadirachta indica*) and kikar (*Acacia Arabica*) leaves Journal of Environmental Biology.
- [4] Vahid. Khatibikamal, Ali, Torabian, Fatemch Janpoor, Cholamali Hoshyaripur (2010), Fluoride removal from industrial waste water using electrocoagulation and its adsorption kinetics. Journal of Hazardous Materials, B179, 276-280.
- [5] 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, 227-232.
- [6] Murugan, M. and Subramanian, E. (2006) Studies on de- fluoridation of water by Tamarind seed, an unconventional biosorbent. Journal of Water and Health, 4, 453- 461.
- [7] Srimurali, M.(2001), Removal of color removal from textile dye wastes by chemical coagulation, Ph.D. Thesis, S.V. University, Tirupati, India.
- [8] A. Ramdani, S. Taleb, A. Benghalem, N. Ghaffour, (2010) Removal of excess fluoride ions from Saharan brackish water by adsorption on natural materials, Desalination. 250 408–413.
- [9] A. Saxena, M. Fujiki, R. Rai, S. Kim, G. Kwak,(2004) Highly sensitive and selective fluoride ion chemosensing, fluoro alkylated poly silicane, Macromol. Rapid Commun. 25 1771–1775.
- [10] T. Rango, A. Vengosh, G. Dwyer, G. Bianchini, (2013) Mobilization of arsenic and other naturally occurring contaminants in groundwater of the main ethiopian rift aquifers, Water Res. 47 5801–5818. doi:10.1016/j.watres.2013.07.002
- [11] M. Srimurali and J. Karthikeyan (2008) Activated alumina : defluoridation of water and household application – a study twelfth international water technology conference IWTC12, Alexandria, Egypt.