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Defluoridation of water using Alum impregnated brick powder and its comparison with brick powder

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Abstract- The batch study process was adopted to study the defluoridation of water using alum impregnated on brick powder. Different adsorption parameters, viz. effect of pH, effect of dose and contact time were selected and their optimization was done for the study. Alum and brick, when used individually are effective materials to cause defluoridation of water. But both the materials have certain limitations. Comparison of adsorption by Alum impregnated brick powder was made with adsorption by brick powder. In the optimum condition of pH and dose of adsorbents, the percentage defluoridation from synthetic sample for Alum impregnated on brick powder was found to be 4.0-5.0 % higher than the defluoridation due to brick powder. The test was performed on synthetic water prepared by using public supply water. The water contained very little other impurities. Presence of other ions in synthetic water did not significantly affect the defluoridation efficiency of Alum impregnated brick powder. The optimum pH range for the material was found to be 6.0–8.0 and adsorption equilibrium was found to be 60 min. These conditions make it very suitable for use in drinking water treatment. Defluoridation capacity of Alum impregnated brick powder can be explained on the basis of the chemical interaction of fluoride with the metal oxides under suitable pH conditions. The adsorption process was found to follow first order rate mechanism as well as Langmuir isotherm.

Index terms: Defluoridation; Adsorption; Brick powder (BP); Alum impregnated on brick powder (AIBP).

I. INTRODUCTION

Natural waters contain fluorides in varying amounts. Consumption of water that contains fluoride in a concentration of approximately 1 mg/liter is found to be effective against tooth decay. For the same reason, salts with Fluoride concentration are added to public water supplies which contain Fluoride less than the desired concentration. In some places, where Fluoride concentration in drinking water supply has been in the permissible limits, tooth decay has been found to be 65% less than in communities with less or zero Fluoride [1]. Most un-fluoridated waters contain less than 0.3 mg/l fluoride.

Excessive consumption of water with fluoride above 1.5 mg/L, however, may cause fluorosis. Fluorosis is condition in which the teeth become mottled, discolored, and pitted during their development. Skeletal fluorosis, characterized by increased bone density and abnormal bone growths, may result from long-term consumption of water containing 8 to 20 mg/l of fluoride [7]. The consumption of fluorides in excess of 20 mg/day over a period of 20 years or more could result in crippling fluorosis [11]. Dental health is of primary consideration for the control of fluoride in water. The USEPA National Interim Primary Drinking Water Standards have indicated that the allowable level of fluoride should not exceed 1.4 to 2.4 mg/l. This level is dependent upon the average maximum daily air temperature since the amount of water, and consequently the amount of fluoride ingested, is primarily influenced by the air temperature of the area. In general, most municipal water supplies contain less fluoride than the amount that is considered to be beneficial to dental health; however, many water supplies are found to exceed this limit.

Rajasthan is located in the North-western part of India. It is more than 40% desert. Groundwater hence, contains fluoride above permissible levels. People in many parts of Rajasthan consume water with fluoride above 10 mg/L. As a result people of all age groups have mottled teeth, skeletal fluorosis, painful and restricted joint, deformities in limbs and hunch back. Providing safe drinking water in interior parts of has been a challenge since the beginning. Efforts have been taken to avenge the problem. But most of the rural people still do not have access to safe drinking water [1].

Concern about elevated fluoride levels in drinking water is not based so much on acute toxicity effects, but rather on the long-term exposure to low levels of fluoride. In Rajasthan, a number of projects have been carried out on a variety of treatment methods for the removal of fluoride from potable water supplies.

In adsorption method, different types of adsorbents have been used till date for defluoridation, e.g. activated alumina [13], coconut shell carbon [2], chemically activated carbon [8], bone charcoal [9], natural zeolites [10], burnt clay [6] and other low-cost adsorbents. The shortcomings of most of these methods are, low fluoride



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removal capacity, high operational and maintenance costs, lack of selectivity for fluoride, generation of large amount of sludge, undesirable after effects on water quality and complicated procedure involved in the treatment. The most commonly adopted method in India is the Nalgonda technique of community defluoridation which is based on precipitation process. It is very efficient and cost effective. The major limitations of Nalgonda technique are large amount of sludge production, daily addition of chemicals, least effective with water having high hardness and high total dissolved solids. Besides, it converts a large portion of ionic fluoride (67–87%) into soluble aluminum complex and practically, removes only a small portion of fluoride in the form of precipitate (18–33%) [5]. Therefore, this technique is erroneous. Residual aluminum ranging from 2.01 to 6.86 mg l⁻¹ was also reported in Nalgonda technique [12], which is dangerous to human health as aluminum is a neurotoxin, concentration as low as 0.08 mg/L in drinking water may cause Alzheimer's disease [3] and has strong carcinogenic properties. Studies have been carried out on brick powder [4]. In the present study, an attempt has been made for defluoridation of drinking water using Alum impregnated on brick powder (AIBP) as a new, feasible, suitable and low-cost adsorbent.

II. MATERIALS AND METHODS

Bricks utilized as adsorbents were manufactured in a brick kiln, situated near DTU Campus (New Delhi). These bricks were washed with distilled water, dried and ground properly to obtain fine powder. The brick powder was washed several times with distilled water till clear water was obtained and was dried in oven at 105 °C for 12 h. The dried material was sieved to separate less than 150 micro meter size of particles for the present study. Lab grade Alum used in the study was taken from laboratory and was used in powdered form. 0.5 grams of Alum was mixed in 1 liters of distilled water and the brick powder was put in the solution for 24 hours. The brick containing Alum was dried under sunlight. The comparison of the defluoridation capacity of AIBP was made with that of the BP.

The stock solution of 100 mg/L fluoride was prepared by dissolving 221 mg of anhydrous NaF in 1 liter of distilled water. Test solution of 10 mg/L F⁻ concentration was prepared from fresh stock solution. All the experiments were carried out in 250 ml conical flasks with 100 ml test solution at room temperature (25°C). These flasks, along with test solution and adsorbent, were shaken in horizontal shaker to study the various control parameters. At the end of desired contact time, the conical flasks were removed from shaker and allowed to stand for 2 min for settling the adsorbent. Then, samples were filtered using Whatman no. 42 filter paper and filtrate was analyzed for residual fluoride concentration by using Fluoride meter as mentioned in the Standard Methods of Examination of Water and Wastewater. Batch study was conducted to determine the optimum conditions and to study the effect of pH, adsorbent dose and contact time on test solution. The effect of pH on fluoride was studied by adjusting the pH of test solution using 0.1 N HCl or 0.1 N NaOH on fixed quantity of adsorbent, while effect of adsorbent dose and contact time were studied by varying dose and contact time, respectively. Optimum conditions were selected for further studies. Water sample collected from a surface water source in New Delhi the details of which is given in the table 1.

Table 1: Water Quality parameters of Public supply water used for experimental work.

Parameter	Value
1. Total Dissolved Solids	230 mg/L
2. Fluoride	0.3 mg/L
3. Nitrate	4 mg/L
4. Chloride	40 mg/L
5. Total Hardness	120 /L



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III. RESULTS AND DISCUSSION

A. Effect of pH

The effect of pH on removal of fluoride was studied in the pH range of 2.0–11.0 and results are shown in Fig. 1

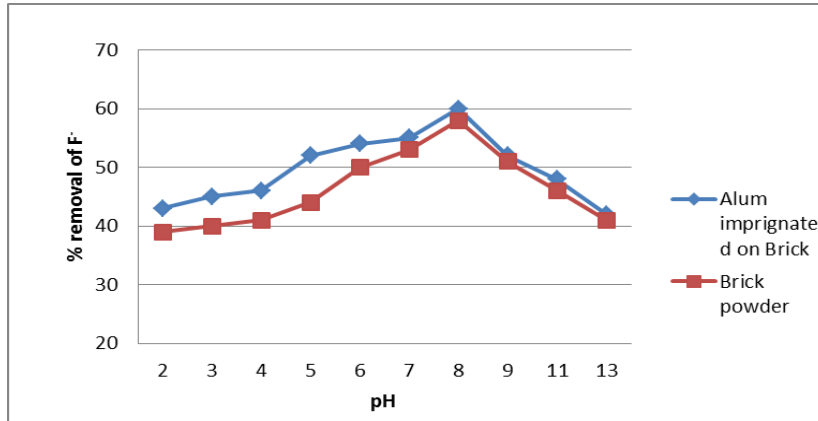


Fig 1: Effect of pH on fluoride removal: Fluoride concentration is 10 mg/L, pH is maintained at 7 and contact time is 60 minutes.

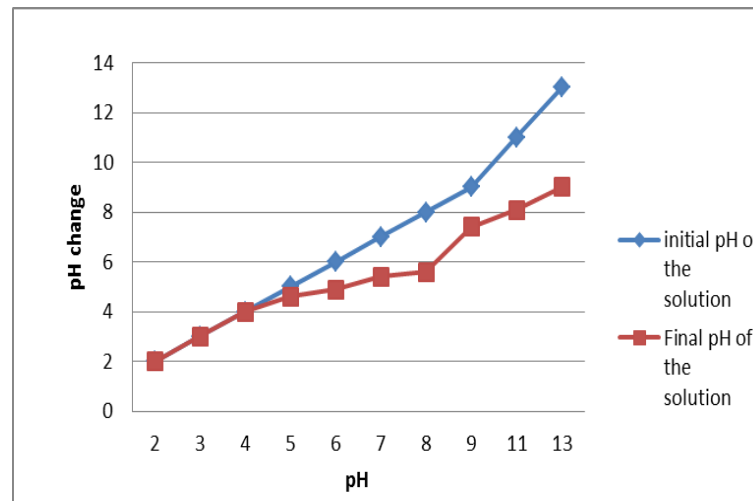


Fig 2: Initial and Final pH of the solution using Alum impregnated brick as the adsorbent.

pH played a major role in fluoride adsorption on AIBP. Comparison has been made with the results of Fluoride adsorption on BP. Maximum adsorption of fluoride was found to be 55.0–59% for AIBP in pH range between 6.0 and 8.0. Percentage removal fell sharply as pH decreased below 6.0 and above 8.0. The lower adsorption efficiency of fluoride in acidic medium might be due to the formation of weakly ionized hydrofluoric acid, which reduces availability of free fluoride for adsorption. In alkaline conditions, lower adsorption may be due to the competition of OH⁻ ions with F⁻ ions for adsorption because of similarity in F⁻ and OH⁻ in charge and ionic radius [4]. Overall, Alum impregnated on brick powder showed 4-5 % increase in Fluoride adsorption. Alum makes the solution acidic and hence the final pH of the solution was on the left side of the pH scale. The pH dropped by 1-2 units in each sample. But maximum removal was obtained in pH range 6-8. pH 7 was adopted for further studies.



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B. Effect of dose

The effect of adsorbent dosage on adsorption of fluoride at pH 7.0, for Alum impregnated on brick powder for 60 minutes, was studied. The results are presented as percentage removal of fluoride from water versus adsorbent dosage in Figure 3. The fluoride removal increased by 4-5 % in this case. Earlier obtained values were from 41 to 58 % for brick powder which increased to 46-64 % for alum impregnated on brick powder for 0.4–4.4 g/100 ml dosages for both. However, it is hereby observed that after dosage of 1.6 and 2 g/100 ml there was

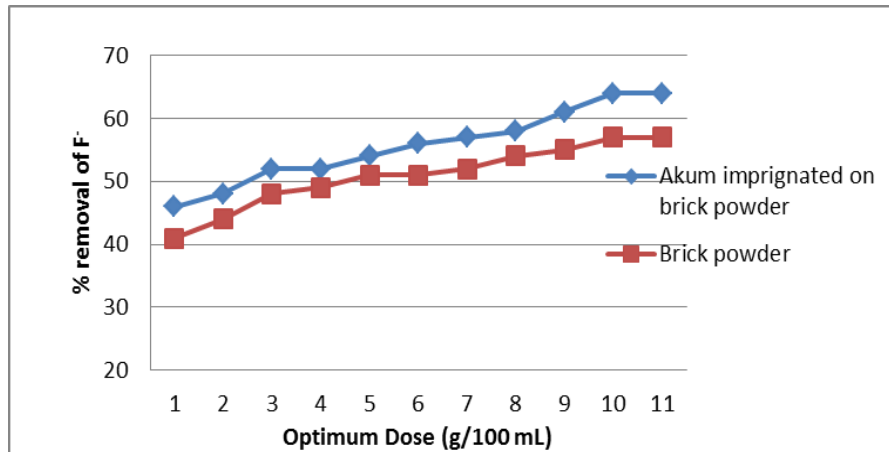


Fig 3: Effect of dose on fluoride removal: Fluoride concentration is 10 mg/L, pH is maintained at 7 and contact time is 60 minutes.

no significant change in percentage removal of fluoride. It was due to the overlapping of active sites at higher dosage, thus, reducing the net surface area [9]. Hence, 2 grams per 100 mL was taken for further study.

C. Effect of contact time

As contact time was increased, initially, percentage removal also increased, but after some time, it gradually approached an almost constant value, denoting attainment of equilibrium. It was assumed that the equilibrium time is that at which curves appear nearly asymptotic to the time axis. In the present case, the equilibrium time was obtained at 60 min for both AIBP and BP. The changes in the rate of removal might be due to the fact that initially all adsorbent sites were vacant and the solute concentration gradient was high. Later, the fluoride uptake rate by adsorbent had decreased significantly, due to the decrease in number of adsorption sites. Decreased removal rate, particularly, towards the end of experiment, indicates the possible monolayer of fluoride ions on the outer surface, pores of both the adsorbents and pore diffusion onto inner surface of adsorbent particles through the film due to continuous shaking maintained during the experiment. [4]

D. Adsorption isotherms

The adsorption data were fitted to linearly transformed Langmuir and Freundlich isotherms. The linearized Langmuir isotherm is given by the Equation 1:

$$\frac{1}{q_e} = \frac{1}{Q_m} + \frac{1}{bQ_m C_e} \quad (1)$$

By plotting graph between $1/q_e$ and $1/C_e$. From the values of the Langmuir and Freundlich isotherms, we see that R^2 value is maximum for Langmuir isotherm which is 0.98. It can be concluded that the adsorption follow Langmuir isotherm and hence, assumptions by Langmuir shall be applicable. Langmuir isotherm assumes that each surface site can be singly occupied, There are no lateral interactions between adsorbed species, the enthalpy of adsorption is independent of surface coverage and energy of adsorption is constant thus creating homogeneity of energy on the surface (there is dynamic equilibrium between the adsorption and desorption processes).



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where Q_m is the maximum adsorption for a complete monolayer coverage. In the equation b and Q_m is obtained

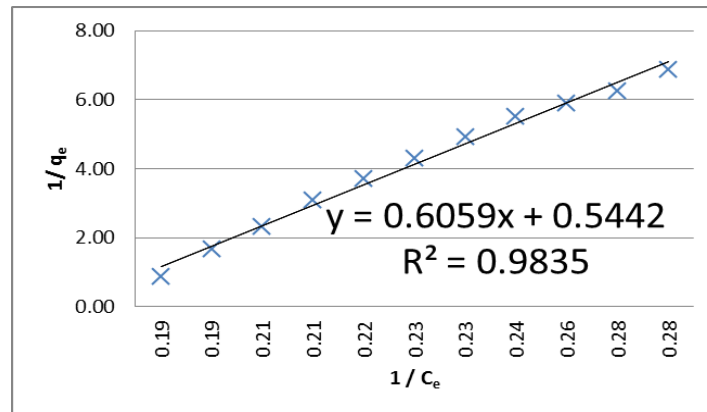


Fig 4: Plot of Langmuir isotherm for adsorption of Fluoride by Alum impregnated on brick powder

IV. CONCLUSIONS

In the present study, a new adsorbent AIBP has been studied for removal of fluoride from synthetic water samples of different fluoride concentrations. The conclusions drawn from the above study are given below:

- (1) Adsorption of fluoride on AIBP from aqueous solution is found to be first order reaction and mechanism of fluoride removal on adsorbent was found to be complex. It follows the same mechanism as in case of defluoridation by using brick powder but with enhanced results. The surface adsorption as well as intra-particle diffusion contributes to the rate-determining step.
- (2) The optimum pH was found to be in range of 6.0–8.0 for maximum adsorption of fluoride, which makes it very suitable for use in drinking water treatment, especially in rural areas. Using lime to balance the slight acidic medium of the effluent water shall make it suitable for use.
- (3) Alum and brick when used individually remove fluoride but has certain limitations. When Alum impregnated bricks were used, better results were obtained with 4-5% increased fluoride removal.
- (4) Langmuir isotherm was followed in the process with correlation value of 0.98.

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REFERENCES

- [1] Abhas Jain, S.K. Singh, "Prevalence of fluoride in ground water in Rajasthan state: Extent, contamination levels and mitigation", Scientific Online, July, 2014
- [2] A.J. Arulanantham, T. Ram Krishna, N. Balasubramaniam, "Studies on fluoride removal by coconut shell carbon", Indian J. Environ. Health 13, 531–536, 1992
- [3] A.M. Davison, "Water supply aluminum concentration: dialysis dementia and effect of reverse osmosis water treatment", Lancet 9, 785–787, 1982
- [4] Asheesh Kumar Yadav, C.P. Kaushik, Anil Kumar Haritash, Ankur Kansal, Neetu Rani, "Defluoridation of groundwater using brick powder as an adsorbent", Journal of Hazardous Materials B128 (2006) 289–293
- [5] B.V. Apparao, S. Meenakshi, G. Karthikayan, Nalgonada technique of defluoridation of water, Indian J. Environ. Protect. 10 (1990) 292–298.
- [6] G. Karthikeyan, M.N. Andral, S.G. Sundar, Defluoridation property of burnt clay, J. IWWA 31 (1999) 291.



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- [7] Hodge, H.C., and Smith, F.A., "Some Public Health Aspects of Water Fluoridation, Fluoridation as a Public Measure," A.A.A.S. (J.H. Shaw, Editor). Publ. No. 38, Washington, D.C. (1954).
- [8] K. Muthukumar, N. Balasubramaniam, T.V. Ramakrishna, Removal of fluoride by chemically activated carbon, Indian J. Environ. Protect. 12 (1995) 514–517.
- [9] Killedar, D.J., D.S. Bhargava, Effect of stirring rate and temperature on fluoride removal by fishbone charcoal, Indian J. Environ. Health 35 (1993) 81–87.
- [10] P.K. Shrivastava, A. Deshmukh, Defluoridation of water with natural zeolite, J. Inst. Public Health Eng. (India) 14 (1994) 11–14.
- [11] Roholm, K., "Fluoride Intoxication. A Clinical-Hygiene Study," H.K. Lewis and Co., Ltd., London (1937)
- [12] S.K. Gupta, A process for defluoridation of water by a filter bed using indigenous material, Indian J. Environ. Sci. 1 (1997) 149–156.
- [13] S. Kumar, Studies on desorption of fluoride from activated alumina, Indian J. Environ. Protect. 16 (1995) 50–53.
- [14] Standard methods for the examination of water and wastewater. APHA, AWWA, WEF, APHA, AWWA, WEF, Washington, DC, 1995.