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Investigations on activated alumina based domestic defluoridation units

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Abstract

Investigations were carried out on the defluoridation of fluoride-spiked ground water in domestic defluoridation units (DDU) with activated alumina (AA). Specific safe water yield (SSY) was determined as a function of AA amount and adsorbent depth. Reuse potential of exhausted AA was assessed by regenerating and reusing AA in multiple defluoridation cycles. High fluoride uptake capacity (FUC) from ground water matrix as well as retaining ≈95% FUC up to five cycles showed the suitability of AA for defluoridation in DDU. SSY, liters of safe water/kg AA, was dependent on the AA amount and its depth. There was a significant decrease in SSY with the decrease in AA depth in different DDUs, even though the amount was maintained constant. The derived data from four DDUs, with 3–5 kg AA and depth ranging from 5 to 13 cm, showed that DDU design is one of the most important parameter to be considered for optimizing SSY.

Keywords: Activated alumina; Defluoridation; Ground water; Domestic defluoridation units; Water treatment

1. Introduction

Fluorosis is a chronic disease resulting from the consumption of excess fluoride, mainly through drinking water. India is among 23 nations in the world, where fluorosis is prevalent. 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 [1]. Excess fluoride is generally encountered in ground water and more than 80% of rural population in India depends on ground water as their drinking water source. Taking health effects into consideration, the World Health Organization has set a guideline value of 1.5 mg/L as the maximum permissible level of fluoride for drinking water [2]. BIS standards, for fluoride in drinking water, are 1.0 mg/L as permissible and 1.5 mg/L as maximum permissible level, respectively [3].

Remedial measures have to be considered for the prevention of fluorosis, if fluoride concentration in a water source exceeds the permissible level. A wide range of treatment procedures has been reported for the removal of excess fluoride from water. These can be broadly divided into three categories: precipitation, adsorption and membrane based. Precipitation methods involve the addition of soluble chemicals to water. Fluoride is removed either by precipitation, co-precipitation or adsorption onto the formed precipitate [4–6]. Adsorption processes involve the passage of raw water through an adsorbent bed, where fluoride is removed by physical, ion exchange or surface chemical reactions with the solid matrix. A wide range of adsorbents, such as activated alumina, bone char, clay, zeolites, flyash, brick and specific ion exchange resins, have been reported for fluoride removal [7–14]. Other defluoridation methods include membrane based reverse osmosis and nanofiltration as well as electrodialysis and electrocoagulation [15–19].

Among these methods, activated alumina (AA) seems to be better suited for large-scale defluoridation due to its specificity and affinity towards fluoride, chemical/physical properties and bulk availability. There have been many reports on the use of AA for fluoride removal [20–26]. Among different modes of application of AA technology in the field, "point of use" domestic defluoridation units (DDU) may be more appropriate in the rural areas of developing countries, where the settlements are scattered.

The objective of the present study was to investigate the performance of domestic defluoridation units, with varying amounts of adsorbent and the bed depth, with an aim to arrive at design specifications. Reuse potential of AA was assessed by regenerating exhausted AA and reusing in subsequent defluoridation cycles.

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Table 1
Properties of activated alumina

Serial no.	Characteristics	Value
1	Particle form	Spheres
2	Particle size (mm)	0.4–1.2
3	Surface area (minimum) (m^2/g)	310
4	Pore volume (cm^3/g)	0.41
5	Bulk density (g/cm^3)	0.86
6	Loss on attrition (maximum) (wt.%)	0.1
7	Loss on ignition (250–1000 °C)	7

2. Materials and methods

2.1. Sorbent

An indigenous manufactured AA grade was used in this study. It was supplied in the particle size range of 0.4–1.2 mm. Properties of AA as provided by the manufacturers, are given in Table 1.

2.2. Test water

Test water for this study was prepared by spiking sodium fluoride to IIT tap water (borewell water) so as to maintain fluoride concentration of 10.5 ± 0.5 mg/L. Characteristics of IIT tap water, used for preparing test water, was periodically monitored and average values are presented as follows: pH 7.9–8.0; alkalinity, hardness, fluoride and sulphate of raw water were 325 mg/L as $CaCO_3$, 360 mg/L as $CaCO_3$, 0.8 and 55 mg/L, respectively.

2.3. Domestic defluoridation units (DDU)

Four domestic defluoridation units were used in this study. Amount of AA was varied from 3 to 5 kg and AA depth ranged from 5.0 to 13.0 cm (Table 2). Plastic microfilter assembly (slot opening <0.2 mm), with a 1.5 mm diameter hole in the threaded plug, was used to control raw water flow rate. With this device, around 9–10 L water could flow through the AA bed in an hour. These units were operated intermittently (so as to simulate field conditions) and around 40 L of raw water was passed through the units per day. Treated water samples were periodically collected for fluoride analysis.

2.4. Regeneration of exhausted AA

Exhausted AA was transferred from DDU to a fine mesh nylon bag. The bag was placed in a plastic bucket with flow control device (4 mm aperture). Ten liters of 1% NaOH was passed through AA bed. After washing once with raw water to remove excess alkali, 1% H_2SO_4 was passed through the alkali treated AA. After washing with water, AA was dried in the oven at 120 °C overnight and was reused for subsequent defluoridation cycle.

2.5. Fluoride analysis

Fluoride was analysed using combination fluoride electrode (Orion 96-09), which was connected to Orion meter (290A). The meter was calibrated with multiple standards in the range of 0.5–10 mg/L. Two milliliters TISAB III (Orion cat. no. 940911), recommended for samples having aluminium, was added to 20 mL sample. Concentration of fluoride was measured after 10 min.

3. Results and discussion

3.1. Performance of DDU

DDU S_1, with 3 kg AA, was used for this study. Fluoride concentration was determined in treated water periodically. Fig. 1 presents the plot of treated water volume versus F^- concentration. Fluoride level in treated water was more or less constant at around 0.4 mg/L up to 350 L. This was equivalent to ≈96.2% fluoride removal. Thereafter, fluoride concentration increased rapidly and around 550 L of test water (F^-, ≤1.5 mg/L) could be treated with 3 kg AA. This was equivalent to around 167 bed volumes and fluoride uptake capacity (FUC) of 1788 mg F^- removed/kg AA. AA surface is amphoteric in nature and can exist as $AlOH^+$, AlOH, and AlO^-. Fluoride binding to AA is proposed to be due to exchange of surface hydroxyl groups, which can be represented by the following reactions.

$$\overline{AlOH_2^+} + F^- = \overline{AlF} + H_2O, \qquad \overline{AlOH} + F^- = \overline{AlF} + OH^-$$

Observations of a recent study by Valdivieso et al. [27], on temperature and pH effects on zeta potential and fluoride adsorption at the α-Al_2O_3/aqueous solution interface, are also in agreement with the above proposed mechanism. Further it was shown that

Table 2
Specifications of domestic defluoridation units

Filters	S_1	S_2	S_3	S_4[a]
Diameter (cm) (top/bottom)	22/22	24/24	25.5/25.5	36/30
Height (cm)	23	26	24	36
AA bed depth				
3 kg	9.0 cm (0.023)	6.5 cm (0.014)	6.0 cm (0.011)	5.0 cm (0.005)
4 kg	11.5 cm (0.030)	9.0 cm (0.019)	7.5 cm (0.014)	6.5 cm (0.007)
5 kg	13.0 cm (0.034)	11.0 cm (0.024)	9.5 cm (0.018)	8.5 cm (0.009)

Values in the bracket represent AA depth/DDU cross-sectional area.
[a] Bucket type unit.