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STUDIES ON DEFLUORIDATION OF WATER

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SUMMARY

Several materials including aluminium salts, calcined alumina, magnesia, lime, activated carbon, sulphonated carbonaceous materials and ion-exchange resins have been screened for their utility in defluoridation of water. Aluminium impregnated cation exchange resins and sulphonated carbonaceous materials, prepared from coconut shell, paddy husk have been found to have a significant defluoridation capacity. Filter alum (aluminium sulphate) solution is also found efficacious. On the basis of results of extensive investigations, a simple and economical domestic defluoridation process has been evolved. The process consists of mixing raw water with required quantity of saturated filter alum solution. Addition of some coagulant aids is found to hasten sedimentation of floccules.

INTRODUCTION

Water-born fluorosis is prevalent in severe intensity in Andhra Pradesh (India). In some parts of the State, there is no possibility of providing alternative low-fluoride drinking water. It has become essential to evolve simple and economical method for removing the excess fluoride from drinking water. Ever since the discovery that fluoride is the cause of mottled enamel (dental fluorosis) and skeletal fluorosis, several scientists in different countries tested numerous materials for defluoridating capacity [1-5]. Most of the methods suffer from many draw

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backs. Important defects are : high capital and operating costs, lack of selectivity for fluoride, poor attribution properties of the material and complexity of operating procedure. In the Institute of Preventive Medicine and State Public Health Laboratories, Hyderabad, many materials such as calcined alumina, magnesia, lime, activated carbon, sulphonated carbonaceous materials and ion-exchange resins were tested for defluoridating capacity. Experiments were also conducted with filter alum (aluminium sulphate). Salient features of the studies are discussed in this paper.

MATERIALS & METHODS

Sulphonated carbonaceous materials were prepared from coconut shell by treatment with sulphuric and fuming sulphuric acids. Commercially available material, carbion, prepared from lignite and ion-exchange resins, Tulsion and Zeocarb 225 were also tested. The tested coagulant aids were put in glass columns and pre-treated with a filter alum solution until a trace of aluminium was found in the filtrate. The materials were washed with distilled water till free of aluminium ion (tested with haematoxylin solution). A saturated solution of filter alum $Al_2(SO_4)_3.18\text{-}24\ H_2O$, Al_2O_3 was prepared for studies. Preparation of the solution using different samples of filter alum indicated that usually 65-70 g of solid is dissolved in 100 ml water. The defluoridation experiments were conducted with tap water after addition of sodium fluoride to ensure sufficient fluoride concentration.

RESULTS & DISCUSSIONS

Using sulphonated carbonaceous material and ion-exchange resins, fluoride bearing water (F = 5 mg/l) was percolated through the material bed and the filtrate was collected in 1 litre lots. The rate of filtration was controlled by attaching a tube fitted with pinch clamp at the bottom narrow end of the glass column. The filtrate was tested for pH, conductivity, hardness, chloride and fluoride following standard procedures [6].

Fluoride was determined with F Ion selective electrode (Orion) and Ion meter. The quality of filtered water showed that alkalinity and hardness were initially very low but gradually increased as filtration progressed. The fluoride concentration also increased from the initial trace amount to 1.5 mg/l. The filtration of raw water was discontinued as the fluoride leakage exceeded 1.5 mg/l. The pH of water increased from 4.0 to 7.0 while there was no change in the concentration of chloride and sulphate. The values of pH and residual fluoride of filtered water are recorded in Table I.

TABLE I

Salient features of treated water (Initial F 5.0 mg/l)

pH	Residual F content (mg/l)
4.0	0.1
5.0	0.3
6.0	0.5
7.0	1.0
8.0	2.0

It is seen from the data that there is a positive correlation between the two parameters.

The defluoridating capacities of the four materials are noted in Table II.

TABLE II

Defluoridation capacity of materials

Material	Capacity (mg/kg)
Sulphonated material from coconut shell	780
Carbion	820
Tulsion	960
Zeocarb 225	1650

It is evident from the results that Zeocarb 225 has the highest defluoridating capacity (1650 mg/l) and sulphonated material of coconut shell has the lowest. Comparison of cation exchange capacities of the materials showed that the defluoridating efficiency is proportional to the cation exchange capacity. The acidity of treated water can be neutralized by passing through deacidite resin or by mixing with raw water.

The ion exchange material could be regenerated by treatment with aluminium sulphate solution (2-4%) and re-used. In the present study, the material was regenerated 20 times without loss of defluoridation efficiency. An ordinary exchange softner can be used for defluoridation on a large scale after necessary pretreatment with alum. The cost of regeneration using filter alum varies from Rs. 1.50 - 2.00 per 1000 litres depending on fluoride concentration (3-5 mg/l). The process can be advantageous for water having excessive hardness or alkalinity.

STUDIES WITH FILTER ALUM SOLUTION

A series of jar tests were conducted by mixing aliquots of 10 litres of raw water with different quantities of alum solution. The raw water was first mixed with a small amount of coagulant aids such as calcite, lime, alumina, sodium silicate, followed by a known amount of a saturated solution of filter alum ($Al_2(SO_4)_3.24H_2O$). After mixing at the rate of 100 r.p.m. for one minute and at 50 r.p.m. for 4 more minutes, the floccules were allowed to settle for 30 minutes. The clear supernatant water was analysed for fluoride and other chemical substituents.

Raw water samples with different concentrations of fluoride but constant alkalinity value of 400 mg/l were studied. The dosage of alum solution and the residual values of fluoride were analysed in Table III.

TABLE III

Quantities of reagent required (10 lit. water alk. 400 mg/l)

Saturated Alum solution (ml)	Original F mg/l: 4	6	8	10
	Residual F after treatment mg/l			
4	2.7	3.8	6.0	8.2
8	1.7	1.9	2.8	4.2
10	1.4	1.5	1.9	3.0
14	0.8	1.0	1.2	1.5

It is seen that high dosages of alum are needed to obtain the desired reduction of fluoride. The experiments were repeated with water samples having increasing alkalinity values but constant fluoride concentration of 5.0 mg/l. The results are shown in Table IV.

TABLE IV

Defluoridation of water (for 10 litres; original F 5.0 mg/l)

Saturated Alum solution (ml)	Alkalinity of raw water 400 mg/l	800 mg/l	1000 mg/l
	Residual F after treatment mg/l		
6	1.5	3.4	3.8
8	1.3	2.8	3.2
10	1.0	2.0	2.2
12	0.8	1.5	1.7
14	0.7	1.2	1.5

It is observed that the reagent dosage steeply increases with alkalinity. It is interesting to note that fluoride removal is associated with reduction in pH of water (Table V) as observed with aluminium incorporated ion-exchange resins.

TABLE V

Relation between pH and F content of water

Amount of reagent	pH initial	pH final	Fluoride (mg/l) initial	Fluoride (mg/l) final
4	8.2	6.8	5.0	1.0
6	8.2	6.2	5.0	0.8
8	8.2	5.6	5.0	0.6
10	8.2	5.0	5.0	0.3
12	8.2	4.5	5.0	0.2

Critical review of all the data showed that the mechanism of fluoride removal is a function of formation of different complexes of aluminium hydroxide and fluoride, depending on pH of the water. The efficiency of the method has been tested with various natural water samples with varying concentration of fluoride and alkalinity. On the basis of results of numerous experiments, a reagent dosage chart has been prepared in Table VI.

TABLE VI

Domestic defluoridation of drinking water

F concentration (mg/l)	Alkalinity	ml reagent required for 10 litres water
2	300	4
4	450	8
6	550	10
8	600	14
10	700	16
12	800	18

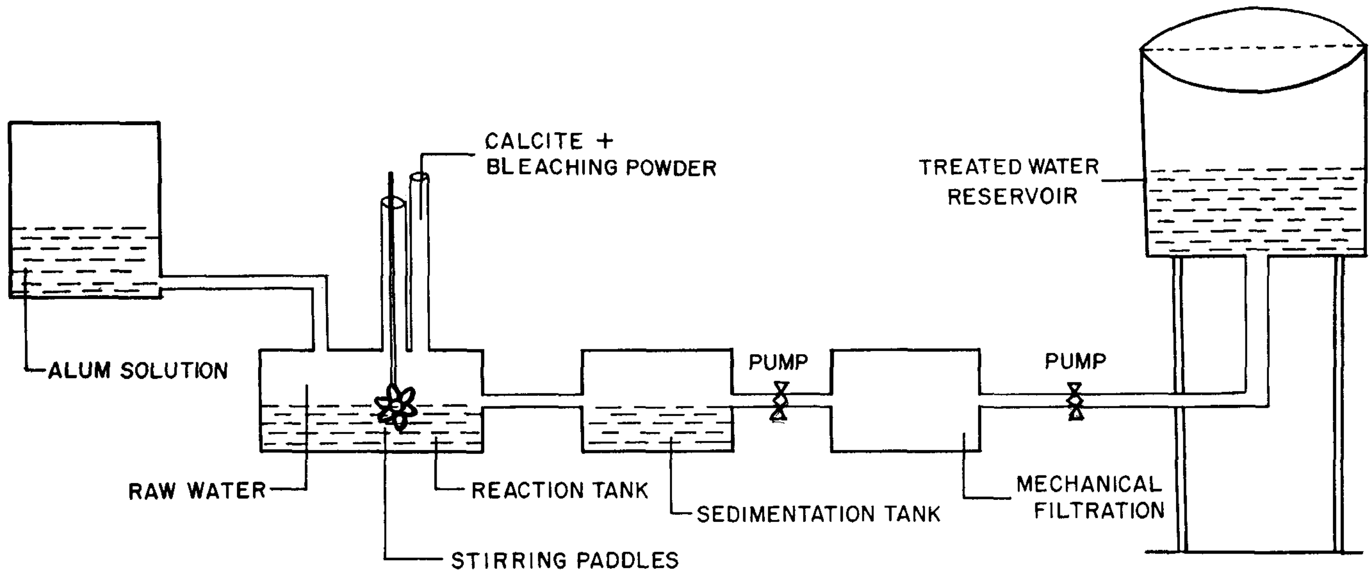

Fig. 1. Defluoridation of water.

After analysing a water sample for fluoride and alkalinity, the dosage of the reagent can be easily fixed by referring to the chart. The dosage should be noted with reference to alkalinity and fluoride values separately, and the average value should be fixed.

The domestic defluoridation process has been successfully demonstrated and popularized in several fluorosis-affected villages in the State. The process is highly economical, the cost being Rs. 1/- for family per month. A schematic diagram of a community defluoridation plant is shown in figure 1. The cost of treatment has been worked out to vary from Rs. 200-400 for treating 100.000 litres depending on alkalinity and fluoride content of raw water.

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