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RESEARCH ARTICLE

Removal of fluoride from aqueous solutions by precipitation

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ABSTRACT : Fluoride although beneficial for the mineralization of tissues in the human body, can be toxic to human being with chronic exposure to elevated concentration. Fluoride considered beneficial in drinking water at levels of 0.5 ppm but harmful once it exceed a level of 1.2 ppm. To date various methods to remove fluoride from water and wastewater have been proposed and applied. An attempt has been made in the present paper to summarise the outcome of study carried out to evaluate the feasibility of using alum to precipitate fluoride from synthetic solution under varied experimental conditions *viz.*, pH (6.5 to 9.5), alum dosage (200 to 500 mg/l), initial concentration of fluoride (Co : 3, 4 and 5 mg/l). Inverse relationship between removal efficiency and Co as well as pH were recorded. On the other hand higher dosages of alum was found to be effective for fluoride precipitation. From aqueous solutions maximum of 78.8 per cent removal of fluoride was recorded with alum dosage of 500 mg/l, pH of 6.5, Co being 3 mg/l. However, all the dosages of alum at all the pH values and Co had successfully reduced the fluoride content from the aqueous solution to the permissible limits.

KEY WORDS : Aqueous solution, Jar test, Fluoride precipitation, Initial concentration, Alum, pH

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INTRODUCTION

Besides the natural geological enrichment of fluoride in ground waters, there can also be formidable contribution from industries. Industries like electroplating, rubber and fertilizer manufacturing, glass and ceramic production, coal power plants, semi conductor manufacturing, generate wastewaters with high concentration of fluoride. Thus, with the increase in industrial activities, water bodies with excess levels of fluoride are becoming a matter of great concern. In lower concentration (0.5 mg/l) is an essential nutrient which prevent tooth decay, where as at higher concentration (^{3} 1.2 mg/l), it causes fluorosis, bristling of bones etc. It is estimated that about 300 million people are adversely affected in 30 countries of the world. Specially in India affected states include – Andhra Pradesh, Orissa, Gujarat and some parts of Karnataka.

Currently a wide range of defluoridation methods are in practice, namely: adsorption, chemical precipitation, removal by ion exchange, membrane filtration and biological. In this present paper an attempt has been made to document and discuss the results of lab scale studies carried out to evaluate the feasibility of fluoride precipitation from aqueous solution using alum under varied experimental conditions.

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EXPERIMENTAL PROCEDURE

Stock aqueous solution was prepared by dissolving predetermined, stiochometrically calculated quantity of sodium fluoride in doubly distilled water. This stock solution which is prepared based on stiochometrical calculation was further rechecked for its absolute concentration by employing process as given in APHA (2006).

Further samples of required concentration *viz.*, 3, 4, and 5 mg/l were prepared by suitably diluting the stock solution so prepared using doubly distilled water and were used for

experimentation. Alum (coagulant) and lime (for pH adjustment) were added in solution form. All the reagents used were of AR grade.

The effectiveness and required doses of coagulant under varied condition of experimentation were evaluated by using jar test apparatus. The apparatus is operated to simulate a mixing, flocculation and settling. Each set of experimentation was carried out by varying initial concentration of fluoride and keeping pH and alum dosage constant. Further experiments were repeated to evaluate the effect of pH and alum dosage. The mixing of contents of jars were carried out at high speed for a short period of time (around 2 – 3 minutes). This rapid mixing phase was followed by about 20 minutes of gentle mixing to promote formation of flocs. The suspension was allowed to settle for 2 hours and then supernatant was tested for its fluoride concentration. For analysis of fluoride ion analyser with fluoride ion selective electrode (Model ISE) was used and procedure as given in instrument suppliers manual was used for analysis.

The variables considered include, pH (6.5 to 9.5), alum dosage (200 to 500 mg/l) and Co (3 to 5 mg/l).

EXPERIMENTAL FINDINGS AND ANALYSIS

The findings of Jar test lab studies are summarized in Table 1 and Fig. 1 show effect of variables considered for study on removal efficiency of fluoride. Based on the analysis of results inferences were drawn and are documented.

Table 1 : Effect of experimental variables on the defluoridation of wastewater

Alum dosage mg/l	Initial F.conc. mg/l	Removal efficiency at stated pH values			
		pH 6.5	pH 7.5	pH 8.5	pH 9.5
200	3	65.4	61.5	52.4	47.7
	4	61.2	50.6	46.8	41.2
	5	54.8	42.7	39.5	33.9
300	3	72.6	66.1	61.8	55.4
	4	65.4	60.0	53.6	46.5
	5	56.4	52.8	43.5	40.1
400	3	78.0	75.1	68.4	62.6
	4	72.5	62.7	55.2	48.6
	5	61.7	55.7	50.3	42.3
500	3	78.8	75.4	68.3	62.5
	4	73.0	63.0	55.4	48.5
	5	61.5	55.8	50.8	42.2

Decrease in efficiency with increase in initial concentration of fluoride was noticed. In the ranges of pH values considered for experimentation, decrease in removal

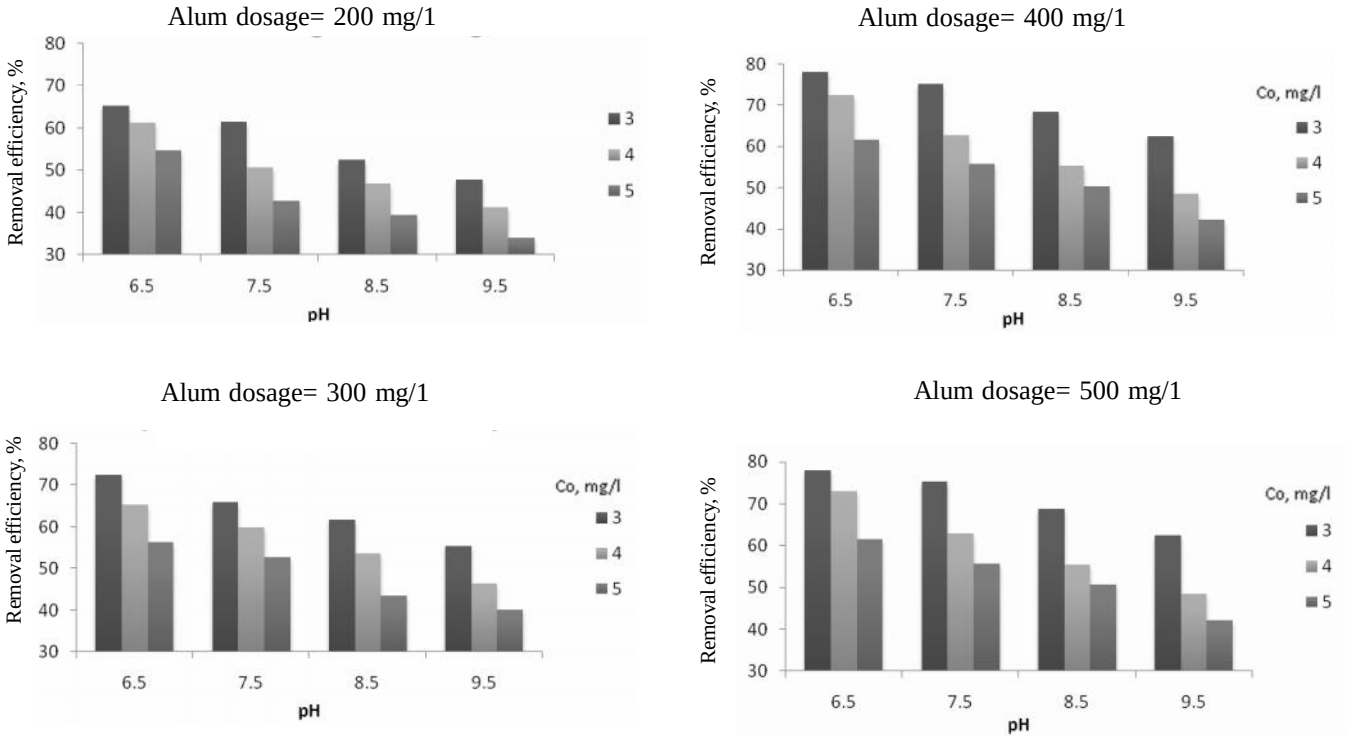

Fig. 1: Effect of pH on removal efficiency

efficiency with increase in pH of synthetic samples was noticed. Best removal efficiency was observed at pH of 6.5. Linear relationship between alum dosage and removal efficiency was recorded. Increase in removal efficiency with increase in alum dosage from 200 to 500 mg/l was noticed. However, within the limitation of experimental and statistical validity no significant changes in removal efficiency for alum dosage of 400 and 500 mg/l was recorded.

Maximum removal efficiency of 78.8 per cent was recorded for alum dosage of 500 mg/l, Co being 3 mg/l and pH being 6.5. Under these conditions of experimentation but for pH values of 7.5, 8.5 and 9.5, removal efficiency of 75.1, 70.2 and 62.6 per cent, respectively were recorded. Also for Co of 3 mg/l removal efficiency recorded ranged from 65.4 to 47.7 per cent, alum dosage being 200 mg/l. These values accordingly for alum dosage of 300 mg/l ranged between 72.6 to 55.4 per cent. Further with alum dosage of 400 mg/l, Co of 4 mg/l removal efficiency noticed was 72.5 per cent (pH – 6.5).

For highest concentration of fluoride (Co–5 mg/l) considered for study in present work, maximum removal efficiency of 61.7 per cent (Alum dosage – 400 mg/l, pH – 6.5) was recorded.

Based on the analysis it was inferred that the optimum design parameter could be as documented below; Co – 3 mg/l, alum dosage – 500 mg/l, settling time – 2 hours, pH – 6.5.

Eventhough the best removal efficiency observed corresponds to pH of 6.5, settling time 2 hours, alum dosage of 500 mg/l and for initial fluoride concentration of 3 mg/l, the following optimum alum dosages have been recommended to achieve the disposal standards of 2 mg/l (Table 2). Similarly Neelo Razabe *et al.*, (2013) also studied removal of fluoride ion from aqueous solution.

Table 2 : Optimum alum dosage recommended (pH – 6.5)

Co	Alum dosages, mg/l		
	200	300	400
3	✓	-	-
4	✓	-	-
5	-	-	✓

Conclusion :

On optimizing all the experimental variables studied in the present work, it was concluded that fluoride from aqueous solution can be best treated by coagulation using alum to meet disposal standard of 2 mg/l. Solution pH of 6.5 was found to be more effective in precipitation of fluoride than higher pH values studied. It was observed that effective alum dosage to achieve the disposal standards depends on the initial concentration of fluoride in the aqueous solution. The results revealed that the disposal standards can be achieved with alum dosage of 200 mg/l for initial fluoride concentrations of 3 and 4 mg/l. But the initial concentration of 5 mg/l can be brought down to less than 2 mg/l only by using alum dosage of 400 mg/l.

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