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Defluoridation of High Fluoride Waters from Natural Water Sources by Using Soils Rich in Bauxite and Kaolinite

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Abstract: A study was conducted to investigate performance of soils rich in bauxite and kaolinite in removing excessive fluoride from water samples collected from different natural water sources. Soil samples from the field were dried, crushed and sieved. Defluoridation processes were conducted in the laboratory using magnetic stirrers. Non-activated and activated soils rich in bauxite and kaolinite were used in this experiment. Results showed that soil rich in bauxite had higher capacity in defluoridation than that of kaolinite. Furthermore, activated soil had higher fluoride removal capacity than non-activated one for both two soils. It revealed thet defluoridation process is dependent on initial concentration, quantity of soil and contact time. The both activated soil have minimum turbidity of water and results into quick water settlement.

Key words: Defluoridation, time, concentration, activated soil, non-activated soil, bauxite, kaolinte

INTRODUCTION

Depending in the presence of some chemicals in the parent rocks, water may have excessive amount of fluoride. Moderate concentrations of fluoride act as skeletal stabilizer, however, high fluoride concentrations in drinking water cause health problems to human being (Wilkister *et al*., 2001). Fluoride, when taken in higher concentrations may result in endemic conditions known as fluorosis. Excessive fluoride in drinking water affects water quality in many parts of Tanzania. It has been observed that the northern part of Tanzania has dental fluorosis problem due to excessive fluoride in water (Kaseva, 2005).

Non skeletal fluorosis is whereby soft tissues are affected due to prolonged intake of fluoride in high concentrations. One way of avoid health problem associated with drinking high fluoride containing water is to avoid domestic use of such fluoride water. Therefore, where alternative sources of water are unavailable, defluoridation may offer practical solutions to the problem. Therefore, defluoridation is needed to reduce excessive fluoride from drinking water. A number of studies on defluoridation have been carried out including the use of bone char (Nahum *et al*., 2007), Manganese-oxide-coated alumina (Shihabudheen *et al*., 2006) and activated alumina (Vivek *et al*., 2007). All these methods have the disadvantage of cost, efficiency or require high technical competence. Priority therefore, should be given to development of cheap method using locally available material such as soils in defluoridation of high fluoride waters.

The objective of this study was to investigate the performance of soils rich in bauxite and kaolinite in removing excessive fluoride concentration from natural water.

MATERIALS AND METHODS

Collection of soil samples for defluoridataion: Soil samples rich in bauxite and kaolinite were collected from Usambara Mountains of North-East Tanzania and Pugu, Dar es Salaam, respectively (Fig. 1 and 2). Soil samples were collected in polyethylene bags and sent to laboratories for further analysis.

Drying, crushing and analysis of soil: Soil sample from the field were dried, crushed and sieved. Mineral content of the soils were analyzed by diffraction X-Ray Diffraction (X-RD), while their chemical composition were determined by fraction X-Ray Fraction (X-RF).

Activation of soil samples: The soil samples were divided into two parts. One part of the soil sample was used direct as a defluoridation media, while the other part was activated through heating. Samples were heated in the oven at 400°C for 30 min. Soils were heated in order to evaluate the effect of heating on defluoridating media.

Experimental set up: The experiments were conducted in the laboratory using magnetic stirrers. Non-activated and activated soils rich in bauxite and kaolinite were used in this experiment. Magnetic stirrer's experiments were conducted using natural water from rivers and boreholes.

The initial fluoride concentrations used were 5, 10.61, 20.22 and 25 mg L^{-1}. Different fluoride concentration levels were used in order to check the effect of initial fluoride concentration on the removal capacity of these soils. Furthermore, different quantities of soil, which include 25, 50 and 100 g were suspended in 100 mL of fluoride containing water, while stirring. Several soil dosages were used in order to evaluate the effect of different soil quantities on removal capacity. The contents were stirred at 100 revolutions per minutes for the maximum required contact time. The control for this experiment contained no soils. The aliquots were taken from each beaker at intervals of 2, 3, 5,10, 15, 30, 60, 90, 120, 150 and 180 min. Ten milliliters of treated water taken from each beaker after mentioned period and filtered through whatman filter paper. The filtrates were analyzed for fluoride concentration.

The same procedures were repeated at different fluoride concentrations, soil dosage and different time intervals.

Fluoride measurement: Preliminary analysis for fluoride was conducted by using an Orion ion specific electrode standard (Frant and Ross, 1966).

Fig. 1: A sample of soil rich in kaolinite from Pugu, Dar es Salaam-Tanzania

Fig. 2: Soil sample rich in bauxite from Usambara Mountains (North-East of Tanzania)

Solutions were made from approximate dilution of stock solution of 100 mgF L^{-1} and Total Ionic Strength Adjusted Buffer (TISAB) standards with fluoride concentrations of 0.1, 0.2, 0.5, 1, 2, 5, 10 and 20 mgF L^{-1} were prepared for plotting a calibration curve. Twenty mililiter of each standard were mixed with 20 mL of TISAB solution to provide a constant ionic strength background to minimize variation between samples and standards. The fluoride concentration was measured as Millivolt (Mv). The millivolt readings versus standards were plotted on graph paper. The milivolt reading decreased linearly as the concentration was raised.

RESULTS AND DISCUSSION

Table 1 presents, the percentage chemical composition of soils rich in bauxite and kaolinite. All soils had almost similar types of chemicals except for ZnO and Na_2O for bauxite and kaolinite, respectively. The dominant chemical in bauxite was Al_2O_3(65.70%); a similar chemical was found in kaolinite at 35.20%. SiO_2 constituted 50.60% of kaolinite whereas SiO_2 constituted 6.81% of bauxite. Other chemicals occurred in minor quantities in both soils except for Fe_2O_3 (9.60%) found in bauxite.

There experiment was carried out using initial fluoride of 5 mg L^{-1}. The maximum fluoride removed by using non-activated soil rich in bauxite was 58.80% after 120 min, while it was 41% for soil rich in kaolinite after 180 min (Table 2). In general, the maximum time required for defluoridation using non-activated soil ranged from 120-180 min. The observation made here is that, non-activated soil rich in bauxite are better sorbents than non-activated soil rich in kaolinite.

The experiment was also carried by using activated soil at initial fluoride concentration used during defluoridation was 5 mg L^{-1}. The maximum removal of fluoride using activated soil rich in bauxite at 5 mg L^{-1} was 72.60% after 180 min, while for activated soil rich in kaolinite was 53.6% observed after 180 min (Table 3).

Table 1: Chemical composition of soil (%) of soil rich in bauxite and kaolinite

Bauxite		Kaolinite	
Chemicals	%	Chemicals	%
ZrO_2	0.065	ZrO_2	0.05
Fe_2O_3	9.600	Fe_2O_3	1.85
Al_2O_3	65.700	Al_2O_3	35.20
SiO_2	6.810	SiO_2	50.60
Cr_2O	0.100	Cr_2O_3	0.04
ZnO	0.037	Na_2O	0.01
K_2O	0.620	K_2O	0.92
CaO	0.710	CaO	0.04
TiO_2	2.520	TiO_2	1.06
LON*	22.100	LON*	11.90

*LON = Loss on ignition

Table 2: Defluoridation using non-activated soils at initial fluoride concentration of 5 mg L^{-1} with pH of 8.3

	Non-activated soil rich in bauxite			Non-activated soil rich in kaolinite		
Time (min)	Residual F (mg L^{-1})	Removed F (mg L^{-1})	Removed F (%)	Residual F (mg L^{-1})	Removed F (mg L^{-1})	Remove F (%)
2	3.22	1.78	36.60	3.87	1.13	22.60
3	3.22	1.78	35.60	3.66	1.34	26.80
5	3.52	1.47	29.40	3.66	1.34	26.80
10	3.00	1.68	33.60	3.27	1.73	34.60
15	3.22	1.74	34.80	3.30	1.70	34.00
30	3.52	1.48	29.60	3.00	2.00	40.00
60	2.15	2.85	57.00	3.00	2.00	40.00
90	2.05	2.88	57.60	2.95	2.05	41.00
120	2.15	2.94	58.80	3.10	1.90	38.00
150	2.22	2.78	55.60	3.00	2.00	40.00
180	2.10	2.90	58.00	2.95	2.05	41.00

Table 3: Fluoride removal using activated soils at initial fluoride concentration of 5 mg L^{-1} with pH of 8.3

	Activated soil rich in bauxite			Activated soil rich in kaolinite		
	Flouride content (mg L^{-1})		Removed flouride	Flouride content (mg L^{-1})		Removed flouride
Time (min)	Residual	Removed	(%)	Residual	Removed	(%)
2	2.65	2.35	47.00	2.93	2.07	41.40
3	2.24	2.76	55.20	2.88	2.12	42.49
5	2.15	2.85	57.00	2.62	2.38	47.60
10	2.24	2.76	55.20	2.56	2.44	48.80
15	2.25	2.75	55.00	2.60	2.40	48.00
30	2.10	2.90	58.00	2.55	2.45	49.00
60	1.40	3.60	72.00	2.44	2.56	51.20
90	1.38	3.62	72.40	2.37	2.63	52.60
120	1.44	3.56	71.00	2.40	2.60	52.00
150	1.38	3.62	72.40	2.32	2.65	53.00
180	1.37	3.63	72.60	2.35	2.68	53.60

Another experiment was carried out at initial fluoride concentration of 10.61 mg L^{-1} (Table 4). Non-activated soil rich in bauxite removed fluoride from water at initial fluoride concentration of 10.61 by 53.25% after 180 min, while non-activated soil rich in kaolinite removed by 37.13% within 150 min.

Experiment was also carried out using activated soils at 10.61 mg L^{-1}. The fluoride removed by activated soil rich in bauxite was 71.63%, while it was 43.35% for activated soil rich in kaolinite (Table 5). Another experiment was carried out at initial of fluoride concentration of 20.22 mg L^{-1}. At initial fluoride concentration of 20.22 mg L^{-1} by non-activated soil rich in bauxite was 46.74%, while for non-activated soil rich in kaolinite was 34.62% (Table 6).

Furthermore, initial fluoride concentration of 20.22 mg L^{-1} was used, using activated soils rich in bauxite and kaolinite. The maximum fluoride removal using activated bauxite was 54.6%, while for soil rich in Kaolinite removed by 39.31% both after 180 min (Table 7). The maximum time of fluoride removal by using non-activated soil at initial fluoride concentration of 25 mg L^{-1} the pH of 9.1 was 180 min, when non-activated soil rich in bauxite was used fluoride was removed by 43.48% (Table 8). When, non activated soil rich in kaolinite was used fluoride was removed by 28%, this was attained at a time range of between 90 and 180 min.

At initial fluoride concentration of 25 mg L^{-1} the maximum fluoride removed using activated soil rich bauxite was 53.40% after 150 min, while it was 31.76% after 180 min (Table 9). Results indicate that activated soil rich in bauxite (at initial fluoride concentration of 5 mg L^{-1}) removed the highest percentage of fluoride followed by non-activated soil rich in bauxite above 70% (Fig. 3). Non-activated kaolinite had the lowest percentage of fluoride removed at initial fluoride concentration of 25 mg L^{-1} (Fig. 4).

In general, the maximum contact time required for defluoridation using soil rich in kaolinite and bauxites ranged from 90-180 min at 100 revolutions per minute. However, soil rich in bauxite found to have high ability of adsorbing fluoride from water than soil rich in Kaolinite. Furthermore, the ability of soils to adsorb fluoride increased when the soils when heated (activated) as indicated in Table 3, 5, 7 and 9. This is probably due to some of cations become more active when soil is heated and is able to reactive quickly with negative ions of F^-, which found in the fluoride water.

It is interesting to note that soil rich in bauxite has higher capacity of removing fluoride from the water even if it was untreated than soils rich in kaolinite. Another notable advantage was the absence of turbidity in the case of activated soil rich in bauxite. Unlike in the case of

Table 4: Defluoridation using non-activated soils at initial fluoride concentration of 10.61 mg L^{-1} with pH of 8.7

	Non-activated soil rich in bauxite			Non-activated soil rich in kaolinite		
Time (min)	Residual F (mg L^{-1})	Removed F (mg L^{-1})	Removed F (%)	Residual F (mg L^{-1})	Removed F (mg L^{-1})	Remove F (%)
2	9.23	1.38	13.00	9.23	1.38	13.00
3	9.00	1.61	15.17	8.47	2.14	20.16
5	8.55	2.06	19.42	9.23	1.38	13.00
10	6.33	4.28	40.34	8.00	2.61	24.60
15	6.32	4.29	40.43	8.23	2.38	22.43
30	6.75	3.86	36.38	7.05	3.56	33.55
60	5.23	5.38	50.71	6.92	3.69	24.77
90	5.66	4.95	46.65	6.89	3.72	35.06
120	4.98	5.30	53.06	6.76	3.85	36.29
150	5.23	5.38	50.71	6.67	3.94	37.13
180	4.96	5.65	53.25	6.68	3.93	37.04

Table 5: Fluoride removal using activated soils at initial fluoride concentration of 10.61 mg L^{-1}, pH of water = 8.7

	Activated soil rich in bauxite			Activated soil rich in kaolinite		
	Flouride content (mg L^{-1})		Removed flouride	Flouride content (mg L^{-1})		Removed flouride
Time (min)	Residual	Removed	(%)	Residual	Removed	(%)
2	6.23	4.38	41.28	7.28	3.33	31.38
3	6.47	4.14	39.02	7.11	3.50	32.98
5	5.32	5.29	49.85	7.11	3.50	32.98
10	4.76	5.85	55.14	6.76	3.85	36.28
15	4.55	6.06	57.12	6.63	3.98	37.51
30	3.11	6.06	57.12	6.25	4.55	42.88
60	3.25	7.50	70.68	6.12	4.49	42.32
90	3.11	7.36	69.37	6.35	4.26	40.15
120	3.02	7.59	71.54	6.22	4.39	41.38
150	3.21	7.50	69.74	6.01	4.60	43.35
180	3.03	7.60	71.63	6.11	4.54	42.78

Table 6: Defluoridation by using non-activated at initial fluoride concentration of 20.22 mg L^{-1} with pH of 8.9

	Non activated soil rich in bauxite			Non activated soil rich in kaolinite		
Time (min)	Residual F (mg L^{-1})	Removed F (mg L^{-1})	Removed F (%)	Residual F (mg L^{-1})	Removed F (mg L^{-1})	Remove F (%)
2	18.00	2.22	10.97	18.55	1.65	8.16
3	18.52	1.70	8.40	18.10	2.12	10.48
5	17.20	3.02	14.93	17.45	2.77	13.70
10	16.00	4.22	20.87	18.00	2.22	10.98
15	16.20	4.02	19.88	15.00	5.22	25.82
30	14.00	6.00	29.67	14.24	5.98	29.57
60	11.27	8.95	44.26	14.55	5.67	28.04
90	11.22	9.00	44.51	13.25	6.97	34.47
120	10.77	9.45	46.74	13.47	6.75	33.38
150	11.01	9.21	45.55	13.47	6.75	33.38
180	10.77	9.45	46.74	13.22	7.00	34.62

non-activated soils rich in bauxite and kaolinite, produced turbid treated water, while treated soils produce clear water without turbidity and faster settling.

Effects of soil quantity/soil on fluoride removal: It was found that the removal capacity of soil material increases with increased dosage. Three different soil dosages were used in the process of defluoridation as shown in Table 10. Table gives a summary of results obtained by adding several dosages of soil. The fluoride removal capacity of the soil material increases with increasing soil dosage as shown in Fig. 5.

Effects of contact time on fluoride removal: The study showed that 2 types of soils (soil rich in bauxite and kaolinite) used in the experment were also affected by contact time. Removal capacity of soil materials increased with time and in later stage fluoride removal progressively decreased. All experiment revealed that after 120-180 min removal of fluoride was reduced, perhaps due to saturation of the anion exchange sites (Basulu and Newlakhe, 1998) (Fig. 3 and 4).

Generally, the maximum time required for defluoridation using non-activated soils ranged between 90-180 min. Activated soil rich in bauxite were better

Table 7: Fluoride removal by using activated soils at initial fluoride concentration of 20.22 mg L^{-1} with pH of 8.9

	Activated soil rich in bauxite			Activated soil rich in kaolinite		
	Flouride content (mg L^{-1})		Removed flouride	Flouride content (mg L^{-1})		Removed flouride
Time (min)	Residual	Removed	(%)	Residual	Removed	(%)
2	14.33	5.89	29.13	16.32	3.90	19.29
3	14.33	5.89	29.13	16.3	3.92	19.39
5	14.25	5.97	29.52	16.45	3.77	18.65
10	13.45	6.77	33.48	14.22	6.00	29.67
15	13.23	6.99	34.57	13.05	7.17	35.42
30	10.26	9.96	49..26	12.72	7.50	37.09
60	9.95	10.27	50.79	12.48	7.74	38.27
90	9.92	10.3o	50.94	12.85	7.37	36.45
120	9.20	11.02	54.50	12.96	7.26	35.91
150	9.21	11.01	54.45	12.28	7.94	39.26
180	9.80	11.04	54.60	12.27	7.95	39.31

Table 8: Defluoridation by using non-activated soil at initial fluoride concentration of 25 mg L^{-1} with pH of 9.1

	Non activated soil rich in bauxite			Non activated soil rich in kaolinite		
Time (min)	Residual F (mg L^{-1})	Removed F (mg L^{-1})	Removed F (%)	Residual F (mg L^{-1})	Removed F (mg L^{-1})	Remove F (%)
2	23.00	2.00	8.00	23.00	2.0	8.00
3	22.50	2.50	10.00	22.70	2.3	9.20
5	20.39	4.61	18.44	21.70	3.3	13.50
10	17.40	7.60	30.40	21.50	3.5	14.00
15	16.33	8.67	34.68	21.20	3.8	15.20
30	15.80	9.20	36.78	20.00	5.0	20.00
60	15.12	9.88	39.52	18.21	6.8	27.16
90	15.10	9.90	39.90	18.00	7.0	28.00
120	15.30	11.00	42.00	18.19	6.8	27.16
150	15.20	9.80	39.20	18.00	7.0	28.00
180	14.13	10.87	43.48	18.00	7.0	28.00

Table 9: Defluoridation by using activated soils at initial fluoride concentration of 25 mg L^{-1} with pH of 9.10

	Activated soil rich in bauxite			Activated soil rich in kaolinite		
	Flouride content (mg L^{-1})		Removed flouride	Flouride content (mg L^{-1})		Removed flouride
Time (min)	Residual	Removed	(%)	Residual	Removed	(%)
2	18.55	6.45	25.80	19.22	5.78	23.12
3	18.26	6.74	26.96	19.26	5.74	22.96
5	17.80	8.00	32.00	19.00	6.00	24.00
10	16.33	8.67	34.68	18.36	6.64	26.56
15	15.13	9.87	39.48	18.00	7.00	28.00
30	12.65	12.35	49.40	17.18	7.82	31.28
60	12.23	12.77	51.08	17.96	7.04	28.16
90	11.93	13.07	52.28	17.13	7.87	31.48
120	12.05	12.95	51.80	17.25	7.75	31.00
150	11.65	13.35	53.40	17.00	8.00	32.00
180	12.05	13.35	53.40	17.06	7.94	31.76

sorbents than non-activated soil rich in kaolinite. Also, for activated soils, bauxite rich soil removed more fluoride than the soil rich in kaolinite.

Soil rich in bauxite had higher capacity of removing fluoride from water even if it was not activated than with those rich in kaolinite. The differing defluoridation capacities two soils types is probably due to their variation in the mineral and chemical contents.

Chemical analyses of these soils showed that soil rich in bauxite contain large amount of iron oxide and aluminium oxide than soil rich in kaolinite. Coetzee *et al.* (2003) and Msonda *et al.* (2007) had similar observations. The high defluoridation capacity of the bauxite was attributed to gibbsite and kaolinite minerals (Msonda *et al.*, 2007).

Aluminium oxide is found to be a good fluoride remover because of the reaction between Al and F molecules. Several fluoride removal methods were carried out using the aluminium based compounds (Busulu and Nawlakhe, 1998). These showed high fluoride removal capacity. Kaolinite contains high percentages of SiO_3 than soil rich in bauxite. The presence of SiO_3 tends to reduce the capacity of soil to adsorb fluoride. This factor might have resulted into poor adsorption by soil rich in kaolinite in the present study.

Table 10: Effect of dosage/quantity of activated soils rich in bauxite on fluoride removal at initial fluoride concentration of 25 mg L^{-1}

	Soil dosages								
	25 g/100 mL			50 g/100 mL			100 g/100 mL		
Time (min)	Residual F (mg L^{-1})	Removal F (mg L^{-1})	Removed F (%)	Residual F (mg L^{-1})	Removal F (mg L^{-1})	Removed F (%)	Residual F (mg L^{-1})	Removal F (mg L^{-1})	Removed F (%)
2	20.11	4.89	16.00	18.55	6.45	25.80	15.33	9.77	39.08
3	20.67	4.33	17.32	18.26	6.74	26.96	15.51	9.49	37.96
5	19.54	5.46	21.84	17.80	8.00	32.00	15.33	9.67	38.68
10	19.68	5.32	21.28	16.33	8.67	34.68	14.78	10.22	40.88
15	17.04	7.96	31.84	15.13	9.87	39.48	13.85	11.15	44.60
30	16.57	8.43	33.72	12.65	12.35	49.40	12.54	12.46	49.84
60	15.95	9.05	36.20	12.23	12.77	51.08	11.46	13.54	54.16
90	14.60	10.40	41.60	11.93	13.07	52.28	10.25	14.75	59.00
120	14.55	10.45	41.80	12.05	12.95	51.80	8.35	16.65	66.60
150	14.45	10.55	42.20	11.65	13.35	53.40	8.57	16.43	65.75
180	13.83	11.15	44.60	12.05	12.95	51.80	8.50	16.50	66.00
210	14.26	10.74	42.96	11.87	13.13	52.52	8.60	16.40	65.60
240	13.97	11.30	44.12	11.75	13.25	53.00	8.41	16.59	66.36

Dosage of material used: 25, 50 and 100 g/100 mL, Solid/solution ration; 1:4, 1:2 and 1:1

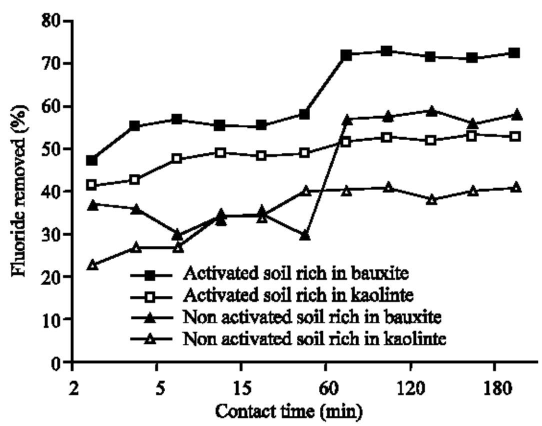

Fig. 3: Comparison between activated and non-activated soil rich in kaolinite and bauxite in fluoride removal at initial fluoride of 5 mg L^{-1}

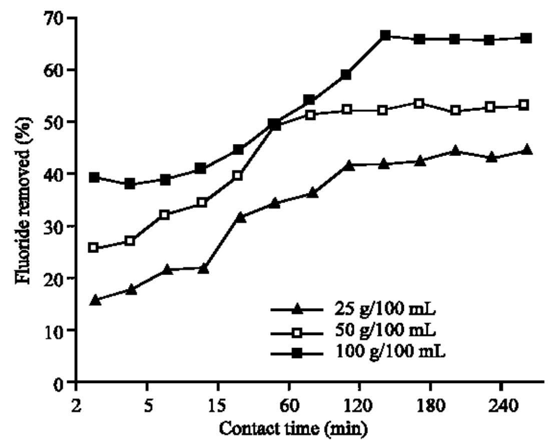

Fig. 5: Effect of dosage on fluoride removal at initial fluoride of 25 mg L^{-1} using activated soil rich in bauxite

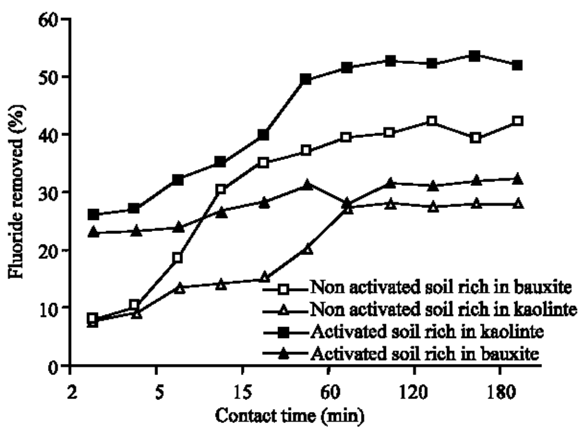

Fig. 4: Comparison between activated and non-activated soil rich in bauxite and kaolinite at initial fluoride of 25 mg L^{-1}

The length of contact time was found to have an influence on the efficiency, by which fluoride was removed from water. Removal capacity of soil materials increased with time to a maximum after which, the removal progressively decreased. The decrease may be attributed to saturation of the anion exchange sites and thereby causing the concentration of fluoride to shoot up (Busulu and Nawlakhe, 1998).

It was observed during the study that sorption or uptake of fluoride is dependent of the initial concentration of fluoride in raw water. Relatively more percentage of fluoride was removed when the concentration of fluoride in water was low. This is probably due to the fact that the increase of fluoride concentration leads into ions increase thereby, increasing the diffusivity and activity of F^-. This is attributable by the utilization of less accessible or energetically less active sites. These energetically less active sites become more fully occupied as the activity of F increases (Srimurali *et al.*, 1998).

Observations from Fig. 3 also shows the effect of dosage quantity of activated soil rich in bauxite on fluoride removal at initial fluoride concentration of 25 mg L^{-1}. Results show that the fluoride removal capacity of the soil material increased with increasing soil dosage. After 240 min at a soil dosage of 25 g/100 mL the maximum removed fluoride percentage was approximately, 43%; a dosage of 50 g/100 mL the removal was approximately, 51% and at 100 g/100 mL the removal was 66%. It revealed that the rate of defluoridation increased with an increase in soil quantity. It suggests that the more the quantity of soil used the more number of active sites for reaction with fluoride ions present in water. This means large amounts of fluoride will be removed from water.

CONCLUSION

Findings from this study show positive results on fluoride reduction by using activated soil rich in bauxite at initial fluoride level of 5 mg L^{-1}. Activated bauxite and kaolinite have an added advantage in that it has minimum water turbidity and had quick water settling.

The study gives hope for the use of low-cost, locally available natural material for removing excess fluoride from domestic water especially for rural areas.

RECOMMENDATIONS

It is recommended that further research on regeneration of materials be conducted as large amount of soil was used to remove fluoride from water.

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