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Fluorosis survey and comparision of fluoride removing capacity from aqueous solution using low cost adsorbents

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Abstract
The present study is investigated to conduct the fluorosis survey among school children and to determine the cheap and better adsorbent for defluoridation by adsorption process. Dharmapuri district is the most fluoride affected areas in Tamilnadu. Therefore, we selected the survey among school children, age group between five and fourteen conducted in the Sunnampatti village of Dharmapuri district. The defluoridation process is conducted by two different adsorbents such as natural clay and brick powder. These adsorbents are powdered and preserved. The defluoridation process was examined using standard fluoride solution under different conditions viz., pH, Adsorbent Dose, Interaction duration and temperature. Before and after fluoride removal process, the physical attributes of natural clay adsorbent were studied under XRD and FT-IR spectrum. The results from the fluorosis survey among school children indicate that the 8 to 9 age group is the maximum extent for dental fluorosis. In defluoridation process, at low pH (pH=2) the maximum amount of fluoride is adsorbed by the natural clay (41%). The quantity of fluoride adsorption is nearly constant, when the quantity of the adsorbent material is 9g and it has rapidly reached equilibrium at 3:30 hours and high portion of fluoride was adsorbed. Generally fluoride adsorption increased from 30% to 40% with the maximum temperature 70 °C. The results indicated that the natural clay can be effective adsorbent for fluoride removal and it can be prepared simply, low-cost, effective and environmentally fit adsorbent of fluoride removal since aqueous solutions. We decided that clay is a potential defluoridating adsorbent than brick powder which can be applied in high fluoride contaminating water.

Keywords: Fluorosis survey, defluoridation, natural clay, brick powder, adsorption, pH

1. Introduction
Water is the precious gift of nature to mankind. It is an elixir of life. About 97.2% of water on earth is salty and only 2.8% is present as fresh water from which about 20% constitutes groundwater. The Groundwater is contaminated mainly by the broad range of physical, chemical, biological, and radioactive parameters. In India, recent research on the assessment of water quality, especially with reference to fluoride has been carried out by various workers [1].

Fluoride is estimated to be the 13th most abundant ion in the earth's crust. The natural abundance of fluoride in the earth's crust is 0.06% to 0.09% and the average crustal abundance is 300mg/Kg. Fluorine is highly reactive and has a strong affinity and it to combine with other elements to produce compounds called fluorides. Fluoride originates from the weathering of fluoride-containing minerals enters to the surface waters with run–off and groundwater through direct contact [2].

Around one-third of the world's population drinks water from groundwater resources. Of this, about 10 percent, approximately 300 million people, obtains water from groundwater resources that are heavily contaminated with arsenic or fluoride. These trace elements derived mainly from weathering minerals [3, 4].

Table 1: Status of Fluoride in various districts of Tamil Nadu

S. No	Status	District
1	Severe	Dharmapuri, Salem
2	Moderate	Coimbatore, Madurai, Trichy, Dindigul, Chidambaram
3	Less	Tirunelveli, Pudukkottai, Ramnad

The presence of high fluoride in groundwater often recognized only when people exhibit symptoms of fluorosis and it is an endemic disease resulting from excess intake of fluoride through drinking water, food in a concentration of 1.5 mg/L or above [5-7].

The proper amount of fluoride helps prevent and control tooth decay in children and adults. Fluoride consumed during tooth development can also result in a range of visible changes to the enamel surface of the tooth. These changes have been broadly termed dental fluorosis [8]. Skeletal fluorosis is a bone disease caused by excessive intake of fluoride in the bones. In advanced cases, skeletal fluorosis causes pain and damage to bones and joints [9].

In this aspect, there is a need for the removal of fluoride in groundwater. Thus the general demineralizing methods like distillation, reverse osmosis, electrodialysis and resin de-anionisation, which are able to remove fluoride from the water [10]. Boiling, UV treatment, most methods of filtration, and most chemical treatment options do nothing to remove fluoride concentrations from water. Synthetic ion exchange and precipitation processes, activated alumina filters, and reverse osmosis are typically used to remove fluoride from water in the developed world [11].

A comprehensive review of the literature reveals that fluoride removal through adsorption on to various materials is the most promising method. In the past few years, surface adsorption was the most interesting area of research to remove fluoride from water. Several adsorbent materials have been tried in the past to identify an efficient and economical defluoridating agent. Activated alumina, activated carbon, activated alumina-coated silica gel, calcite, activated saw dust, activated coconut shell carbon-activated fly ash, groundnut shell, coffee husk, rice husk, magnesia, serpentine, tri-calcium phosphate, bone charcoal, activated soil Sorbent, carbon, etc. are different adsorbent materials reported. The fluoride-removing efficiency of activated alumina gets affected by hardness, pH, and surface loading. This adsorption process can remove fluoride up to 90%, and the treatment is very cost-effective [12].

In this aspect, the adsorbent material can be prepared easily, locally available, cheap, efficient and eco-friendly. This adsorption technique for defluoridation of water due to its availability, high selectivity, low cost, high efficiency and being a comparatively more environmental friendly technique. Natural clay and Brick powder is evaluated as an appropriate adsorbent due its low cost and high removal efficiency [13].

This study conducts fluorosis survey among children and goes for building up an ease novel adsorbent of eliminating fluoride in groundwater using natural clay and brick powder.

2. Materials and methods

2.1 Data collection of school survey

This was a cross-sectional observation study. It was conducted in the Sunnampatti village situated in Pennagaram Taluk, Dharmapuri district, Tamilnadu, India. This study comprised 101 school children aged between 5 to 14. This school children's residential area since birth was formerly confirmed. It consent and approval from concerned school authority were obtained in advance.

2.2 Readiness of adsorbent

The clay powder was obtained in a lake from Sogathur village situated in near Dharmapuri, Tamilnadu, India. The Brick powder collected in a brick furnace from the Sunnampatti village of Begarahalli situated in near to Palacode, Dharmapuri District, Tamilnadu, India. Before use, these adsorbents are washed with distilled water and dried in an oven at 100 °C for 24hrs, powdered and preserved. These adsorbents were carefully packed in an airtight container immediately. It is used for various batch adsorption and characterization studies.

Characterization studies, like XRD were taken from Alagappa University, Karaikudi. FT-IR was taken from the Instrumentation Laboratory, St. Joseph's College, Tiruchirappalli.

2.3 Standard Fluoride Solution

S. M. M. Nazeeb Khan et.al, suggests the Pallipatti village having a high fluoride affected area [14]. It is situated in Pennagaram Taluk of Dharmapuri district, Tamilnadu, India. So we collect the ground water sample from this area for two liters in a pre-cleaned polythene container. It is filtered and preserved. In this ground water sample is analyzed for fluoride concentration. These results show the concentration of fluoride in this ground water sample is 2.01mg/L or ppm. This water is used for a standard fluoride solution.

3. Results and discussion

3.1 Dental Fluorosis Survey

Table 4: Percentage of dental fluorosis among school children in various age groups

Age	5 – 7 Year		8 – 9 Year		10 – 11 Year		12 – 14 Year		Total	
	Boys	Girls	Boys	Girls	Boys	Girls	Boys	Girls	Boys	Girls
No. of children examined	10	9	16	12	11	13	14	16	51	50
No. of children affected	7	4	13	11	09	11	13	11	42	37
% of victims	70%	44%	81%	91%	81%	84%	92%	68%	82%	74%

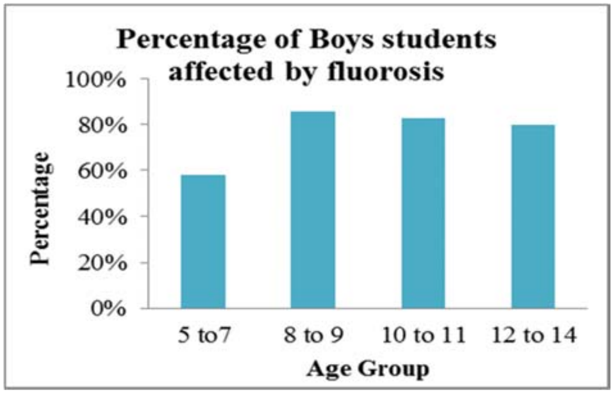

Fig 1: Percentage of dental fluorosis among boy children in various age groups

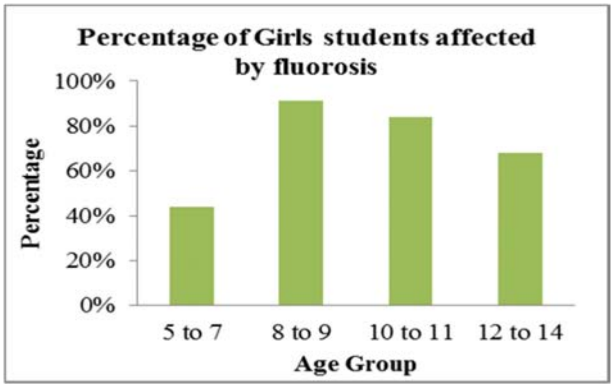

Fig 2: Percentage of dental fluorosis among girl children in various age groups

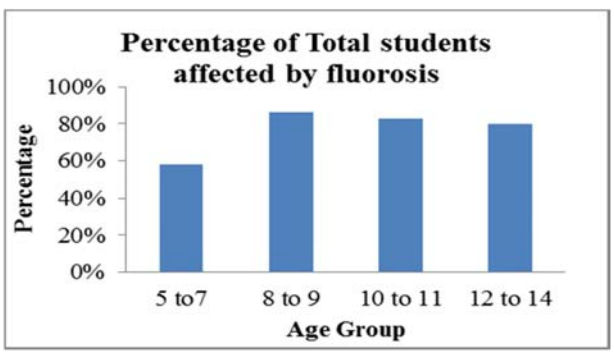

Fig 3: Percentage of dental fluorosis among school children in various age groups

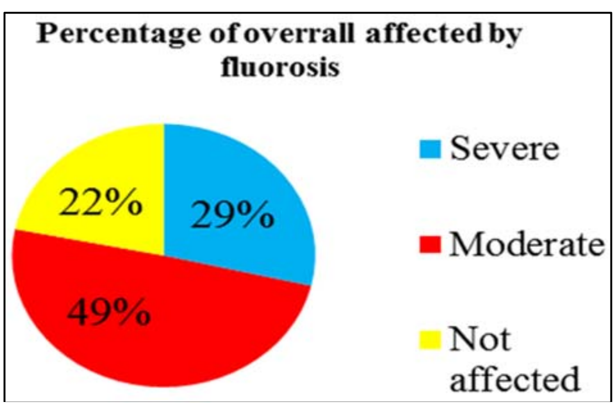

Fig 4: Percentage of dental fluorosis among school children for different categories

The results shown that 8 to 9 age groups of children are affected maximum extent. Out of 101 children examined 79 children are affected by the fluorosis disease. The percentages of victims are 78%. Around 101 students are examined, nobody affected by skeletal fluorosis.

3.2 Mitigation of fluoride
3.2.1 Effect of pH
Generally, the pH is an important variable affecting adsorption of the solute/ solid by the interface. The effect of pH on fluoride removal by brick powder and clay powder material was studied at five altered pH such as, 2,4,6,8, and 10 by keeping all factors constant (Adsorbent material dose 5g, temperature 29 °C, shaking speed 100 rpm and constant time four hours, initial Fluoride concentration 2.01mg/L), and the results are indicated in Fig. 5. The pH of the medium was attained using necessary quantities of 0.1N solution hydrochloric acid and sodium hydroxide solution.

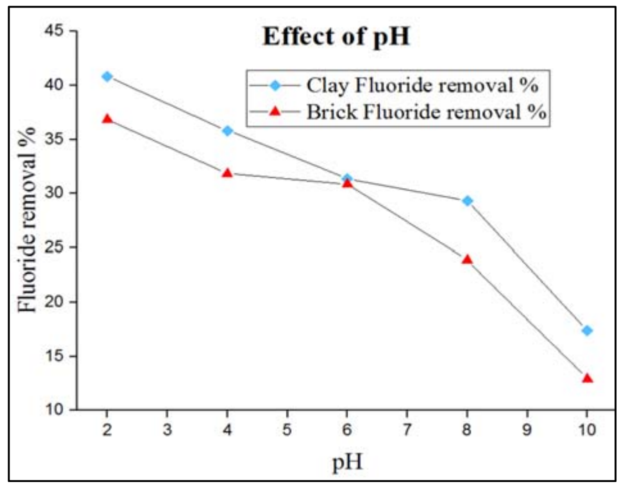

Fig 5: Shows the effect of pH on fluoride adsorption for clay and brick powder

For both adsorbents, fluoride adsorption was found to increase at lower pH, because the surface is highly protonated in an acidic medium maximum fluoride is removal attributed. The trend for the fluoride elimination in the alkaline pH range was low; that was due to competition of hydroxyl ions with fluoride for adsorption sites owing to resemble in fluoride and hydroxyl ions responsible and ionic radius.

The reduction in the percentage of adsorption of fluoride at higher pH levels is due to increasing the electrostatic repulsion between the negatively charged surface sites of the adsorbent and fluoride ions [15]. In this series clay material was highly protonated. Therefore, clay material was highly activated for fluoride adsorption than brick powder.

3.2.2 Effect of adsorbent dose
The influence of adsorbent dosage on fluoride removal was studied at an initial fluoride ion concentration of 2.01 mg/L by allowing a contact time of 4 hours. The amount of clay and brick powder significantly influences the amount of fluoride adsorptions. As the quantity of these adsorbents is increased from 3 to 15 g, there is a specious rise as the percentage of fluoride removal (Fig. 6). It was witnessed

that percent of fluoride removal increased from 40 to 50% with an increase in clay adsorbent dose.

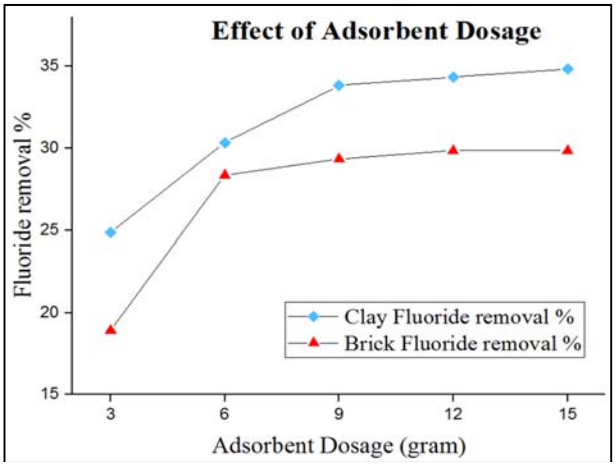

Fig 6: Shows the effect of Adsorbent dose on fluoride adsorption for clay & brick powder

This specifies that the number of active adsorption spot at a higher dose is more than sufficient to hold fluoride ions. Nevertheless, there is almost constant in the removal of fluoride, when the dosage of the adsorbent is above 9 g. This implies that adsorption reaction is feasible to attain to a dynamic equilibrium.

3.2.3 Effect of equilibrium time

The effect of contact time between the adsorbent and fluoride ions was studied at the one hour time intervals up to 5 hours. The effect of contact time between the adsorbent and fluoride ions can be observed in (Fig 7). Apparently, the equilibrium time is the time at which the curves appear almost asymptotic to the time axis.

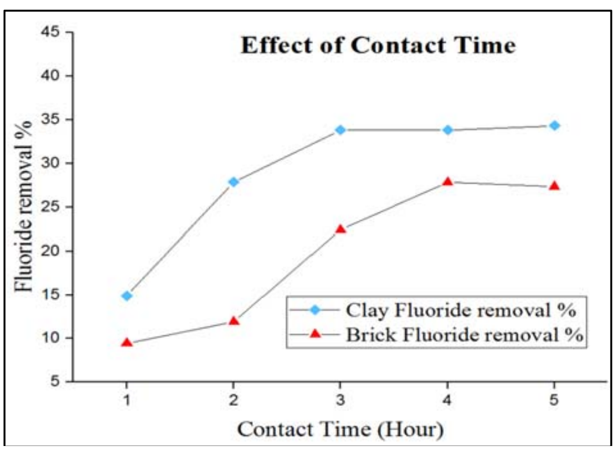

Fig 7: Shows the effect of contact time on fluoride adsorption for clay and brick powder

In clay adsorbent, the equilibrium was attained at three hours; the sorption percentage altered from 14% at one hour to 33.83% at three hours, at which point it raised ground. Similarly, brick powder attained equilibrium at 4 hours; the sorption percentage altered from 9.45% at one hour to 27.86% at four hours. This perhaps owing to the fact that once a certain amount of fluoride ions gets doped onto these composites within a given time, no more doping occurs afterwards, i.e. the doping level might have attained [16].

Further, the F- rapidly reached equilibrium at three hours, and 33.83% of F- was adsorbed, after that there is no more fluoride uptake takes place on clay material. But, the brick powder adsorbs Fluoride rapidly reached equilibrium at four hours, and 27.86% of fluoride was adsorbed, after that there is no more fluoride excess adsorption takes place. Hence, three-hour contact time was chosen as the optimum time for the clay adsorbent and four-hour contact time was chosen as the optimum time for the brick adsorbent for further inspection.

3.2.4 Effect of temperature

The effect of temperature on fluoride elimination of clay material as well as brick powder was examined at four different temperatures, viz., 28 °C, 40 °C, 55 °C and 70 °C. The result of such tests is shown in fig. 8. It is more experienced than the adsorption potentials of clay material for fluoride increases from increasing the temperature as an adsorbent dosage of 5g, initial fluoride concentration of 2.01 mg/L respectively.

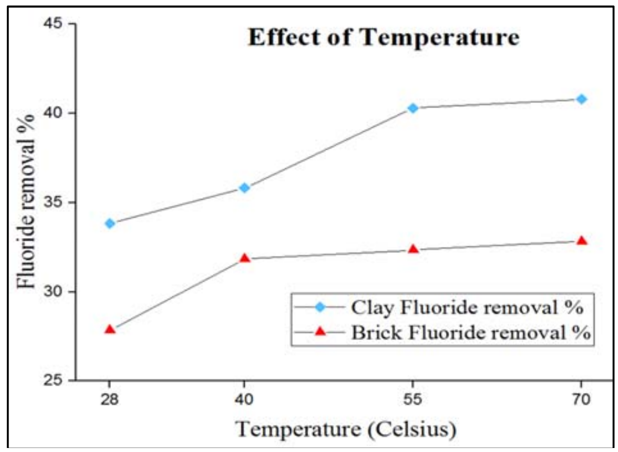

Fig 8: Shows the effect of temperature on fluoride adsorption for clay and brick powder

The contact time during the experiment was four hours. The maximum amount adsorbed increased from 1.0 to 1.6 mg/L (at 2 mg/L, initial fluoride concentration, and Contact time 3 hours). The observation shows that the interaction between adsorbate and adsorbent is endothermic in nature, and the temperature seems to be a significant parameter**.**

3.2.5 XRD Spectrum for fluoride adsorption

The XRD patterns of untreated and fluoride treated clay materials are given in Fig. 9 and 10. The XRD patterns of adsorbent after fluoride adsorption showed significant changes. It indicates the fluoride ion involved in the internal arrangement of Clay material. XRD model of the adsorbent material before the fluoride treatment clay material shows the intensity of peak subsequent to the 2ϴ values at 20, 26, 27, 35, 49, 61, and 67. The XRD model of the after fluoride treatment clay material showed considerable changes in the intensity of peak following to the 2ϴ values at 19, 26, 34 and 61.

The X-ray Diffraction pattern of the treated clay material gives an indication of small alteration over the crystal cleavages. This proves the strong adsorption of fluoride lying on the surface of the adsorbent. Furthermore, after fluoride adsorption, the appearance of some peaks confirms that the adsorption process is purely chemical adsorption.

This suggested that the uptake of fluoride ions through the adsorbent is by chemisorption, which accordingly modifies the structure of the adsorbent.

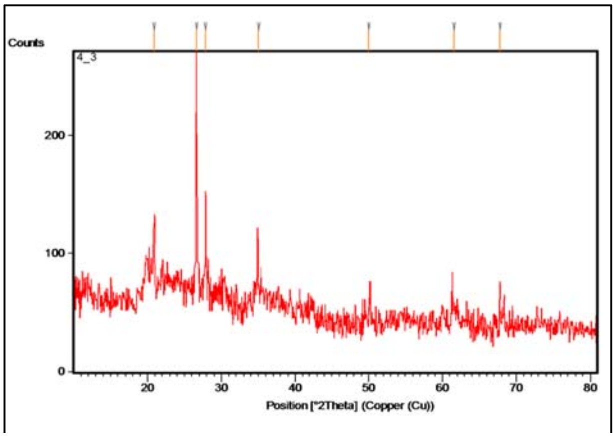

Fig 9: X-ray diffraction pattern of clay material Before Fluoride adsorption

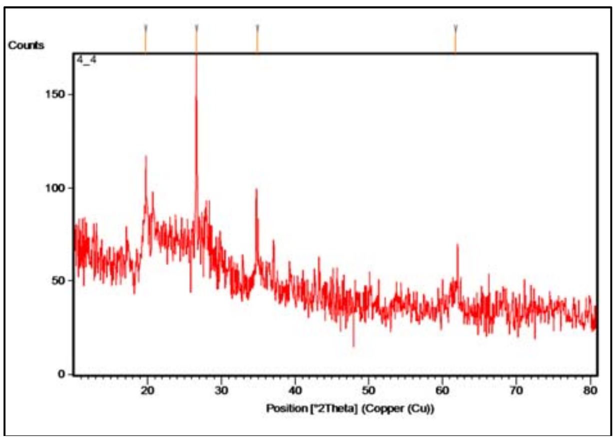

Fig 10: X-ray diffraction pattern of clay material After Fluoride adsorption

3.2.6 FT-IR Spectrum for Fluoride adsorption

The FT-IR spectrum of untreated and fluoride treated natural clay material was characterized and illustrated in Fig. 11 and 12. The shift in stretching frequency from 3623 to 3422 cm-1 is assigned to the FT-IR spectrum involvement of hydroxyl groups. The Stretching frequency of the Hydroxyl group shows the spectrum image at 2361 cm-1. The adsorption bands in between 3300 to 3600 cm-1 show the characteristics of -OH group [17].

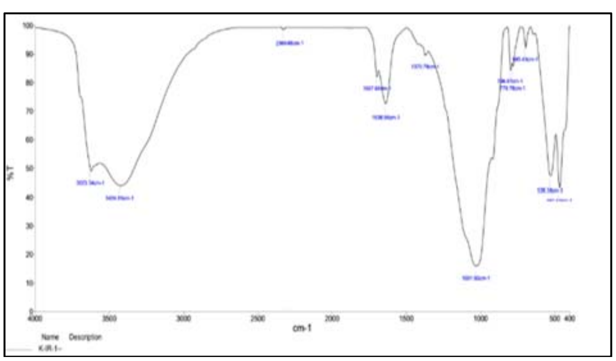

Fig 11: FT-IR spectrum of Clay material Before Fluoride adsorption

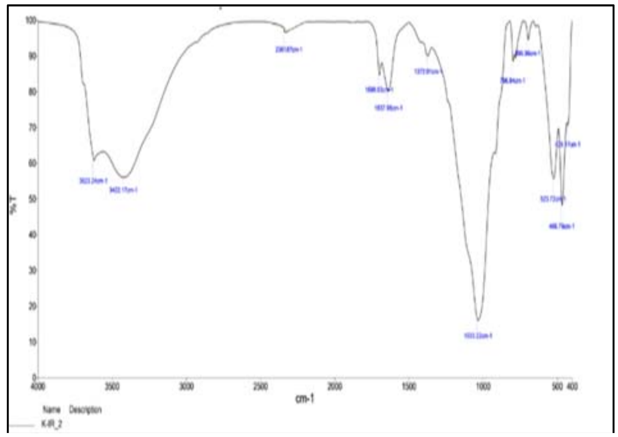

Fig 12: FT-IR spectrum of Clay material After Fluoride adsorption

FT-IR spectrum revealed that the hydroxyl groups on the adsorbent surface were involved in the sorption of fluoride. Anion exchange and electrostatic interaction were suggested as the main mechanisms involved in the sorption of fluoride on the adsorbent. The changes of stretching frequency of Fluoride treated clay material compared to natural clay material confirm the chemical modification.

4. Conclusion

From the survey, it practices an increasing trend in dental fluorosis among school children predominantly recorded in 8 to 9 age group are affected by the maximum level of fluorosis. This study indicates a serious need to recover the quality of water and educate defluoridation of drinking water in affected areas to decrease the problem of dental fluorosis. From the defluoridation technique, the clay powder fundamentally improved the adsorption ability of fluoride removal than compared brick powder. At low pH (pH=2) the maximum amount of fluoride is adsorbed by the clay (41%). The amount of fluoride adsorption is almost constant, when the quantity of the adsorbent material is 9g and it has rapidly reached equilibrium at three and half hours and high percentage of fluoride was adsorbed. Generally fluoride adsorption increased from 30% to 40% with the maximum temperature 70 °C. The results indicated that the clay can be effectively utilized as a compelling adsorbent for fluoride removal. It can be prepared simply, low-cost, effective and environmentally fit adsorbent of fluoride removal since aqueous solutions. We decided that clay is a potential defluoridating adsorbent than brick powder.

5. Acknowledgement

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