# Fluoride Removal from Aqueous Solution Using Neem as Adsorbent

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***Abstract - Batch adsorption dynamics and equilibrium studies for the removal of fluoride ions from aqueous solution using neem leaf, has been carried out under various experimental condition. In the present study, neem leaf powder is used as an adsorbent for the removal of fluoride from aqueous solution. The process parameters are agitation time, adsorbent dose, pH of the solution, initial fluoride ion concentration and temperature. The result showed that maximum fluoride ion removal is possible at a dose of 12gm/L of neem leaf for a contact time of 90 minutes and temperature of $45^0$C.Charecterisation of neem leaf before and after fluoride adsorption was studied by SEM and FTIR study was done to get a better insight into the mechanism of adsorption. The rate of adsorption followed pseudo- second order kinetics. Freundlich isotherm fits well for the defluoridation of water using neem leaf.***

***Keywords: Fluoride ion, adsorption, low cost adsorption, SEM, isotherms, kinetic study.***

## I. INTRODUCTION

More than 95% of the rural and about 30-40% of urban population in India depends on ground water for its domestic requirement. High concentration of contaminants such as fluoride and nitrate in ground water render it unfit for drinking purpose. Fluoride is an essential element of drinking water but at higher concentration i.e.; more than 1.5 mg/L may cause harmful effects on human health. Fluoride contamination in ground water has been recognized as a global problem and its concentration in drinking water at many places of the world exceeds the permissible limits.[1]

An intake of more than 6 mg of fluorine per day results in fluorosis. Fluorine being cumulative bone-seeking mineral, the resultant skeletal changes is progressive. Fluoride increases the stability of crystal lattice in bone, but makes the bone more brittle. Drinking fluoridated water will double the number of hip fractures. The International Society for Fluoride Research (ISFR) has reported studies implicating fluoride in the rising rates of Down's syndrome, chronic fatigue syndrome and sleep disorder.

Defluoridation is the process of removal of fluoride ion in drinking water. The process may be classified broadly into two categories, namely, i) Additive methods, and ii) Adsorptive methods. The different methods so far tried for the removal of excess fluoride from water can be broadly classified into four categories: A) Adsorption methods, B) Ion exchange methods, C) Precipitation methods, and D) Miscellaneous methods. Some defluoridation techniques developed to control fluoride content in water are reverse osmosis, adsorption using sunflower plant dry powder, steam of phytomass, Holly Oke, neem bark powder, activated cotton jute carbon, bagasse ash, burnt bone powder, phosphate-treated saw dust, bone char, etc. as adsorbents, Nalgonda technique, activated alumina process and ion exchange process. However, due to high cost or lower efficiency or non-applicability on mass scale these techniques are not much in use.

The conventional method of fluoride removal includes: ion-exchange, reverse osmosis and adsorption [29, 30, and 31]. The ion-exchange, reverse osmosis is relatively expensive. Therefore, still adsorption is the viable method for the removal of fluoride. Adsorption is the process considered to be efficient to defluoridate the water. Adsorption involves the passage of contaminated water through an adsorbent bed, where fluorides removed by physical, ion-exchange or surface chemical reaction with adsorbent. Because of its ease of operation and cost-effectiveness, adsorption is still widely accepted pollution removal technique. Plant materials are reported to accumulate fluoride and hence application as defluoridating agents has been suggested. The use of medicinal plant materials for the fluoride removal was investigated.

Several publications are available on the effective removal of fluoride using low cost materials. The tested materials include activated alumina, amorphous alumina, activated carbon, calcite, zeolite clay, charcoal, bleaching earth, red mud etc [28, 32, 33]. The materials like kaolinite, bentonite, charfines, lignite and nirmali seeds were also investigated for the removal of fluoride [34].

This communication presents the findings of an investigation on the use of leaf powder from various trees for the defluoridation of water. [2]

## II. MATERIALS AND METHODS

All the reagents used were of AR grade. Fluoride stock solution was prepared by dissolving 221 mg anhydrous sodium fluoride in 1000 ml distilled water in volumetric flask. Fluoride standard solution was prepared by diluting 100 ml stock solution to 1000 ml distilled water in volumetric flask. This 1 ml solution has 0.1 mg of fluoride.

*Equipment*

The pH of the solution was measured with a EI (Electronics India) make digital pH meter (Model No. 111) calibrated with standard buffer solution and treated solution after adsorption experiments. Scanning electron microscope (SEM) study was carried out to observe the surface texture of the adsorbents. FTIR studies were carried out to determine the type of functional group responsible for fluoride adsorption.

*Synthesis of adsorbents*

There are a lot of bio-adsorbents present in the world that can be used to remove fluoride from water irrespectively their capacity. We used in this study neem (Azadira chtaindica) leaves as a bio –adsorbents. Neem leaves were obtained from neem tree. It was washed several times with distilled water and dried at $105^0$C for 6 hrs in hot air drier to remove adherent moisture. Then it was grinded in mixer grinder and sieved through standard sieve to obtain particle of mesh size 60. After that the sample was stored in an air tight container.

## III. BATCH EXPERIMENTS FOR FLUORIDE REMOVAL

For adsorption experiments, stock solution of fluoride was prepared by dissolving sodium fluoride salt in distilled water (100mg/L). We studied batch adsorption for analysis. Adsorption experiments were carried out for the determination of concentration, optimum pH value, contact time, dose of the adsorbent & temperature. The influence of pH (2.0-10.0), contact time (15,30,45,60,90 and 120 min), initial fluoride concentration (10.0,12, 14,16,18 and 20.0 mg/L), temperature (5, 10, 25, 45, 55, 60), weight of the adsorbent dose (3, 5, 7, 10, 12, 14 g/l) were evaluated during the study in a 50 ml flasks with fluoride solution of known concentration. The Powdered neem leaves as an adsorbent were added in each flask and the flasks were kept for constant shaking for 1hr and then the solids were separated through filtration. The solutions were collected and developed the colour and fluoride concentration in the solution was determined by spectrophotometer. Each experiment was conducted three times and average values were reported. The amount of fluoride adsorbed per unit adsorbent (mg fluoride per g adsorbent) was calculated according to a mass balance on the fluoride concentration using Eq:

$$q_e = \frac{(C_i - C_f)X}{w} \quad \text{.....................(1)}$$

Where, X = volume of the solution.

W = weight of adsorbent (g)

The percent removal (%) of Fluoride was calculated using the following equation:

$$\text{Removal(\%)} = \frac{(C_i - C_f) \times 100}{C_i} \quad \text{......(2)}$$

*Effect of equilibrium pH*

Experiments were carried out by varying initial solution pH values within the range of 2-10, adjusted with 0.1 M NaOH or .01 M HCL. Accordingly, adsorption was done by adding different amount of adsorbent.

## IV. RESULTS AND DISCUSSION

*Effect of initial concentration*

Effect of initial concentration on the removal of fluoride has been studied in the specified range of concentrations.6 sample of volume 50ml of different concentration in a 100ml flask. Adsorbent of dose 10.0 gm/l were added in each flask. Then the sample was kept for constant shaking at 150 rpm for 1hr. After adsorption sample were filtered out and take 20 ml for each and mixed with 5ml ZR-SPANDS solution and we got the final concentration of fluoride by using spectrophotometer.

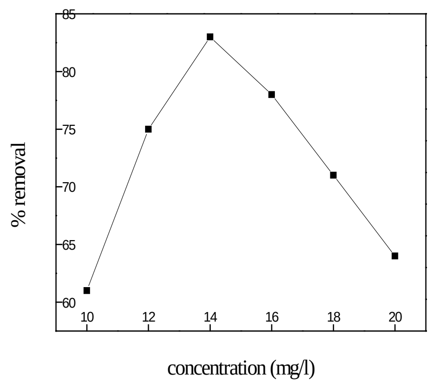


Fig 1. Effect of concentration on the adsorption of fluoride by neem leaves.

The influence of sorption rate is shown in Figure.1. Initially it was observed that the percent fluoride removal increased as the concentration increased from 10.0 to 14.0 mg/l but above the concentration 14.0 mg/l the fluoride removal decreases. Therefore, the solution concentration was maintained at 14.0 mg/l.

*Effect of pH*

Effect of pH on removal of fluoride was studied at 14 ppm of initial concentration and 10gm/l of adsorption dose and 60 min of contact time with a constant shaking rate 150 rpm. The values of pH were varied from 2 to 10. Initially it was observed that the percent fluoride removal increased as the pH increased from 2.0 to 4.0 but above pH 4 the removal becomes constant. Therefore, the pH of the solution maintained at4.0. The influence of the pH on sorption rate is

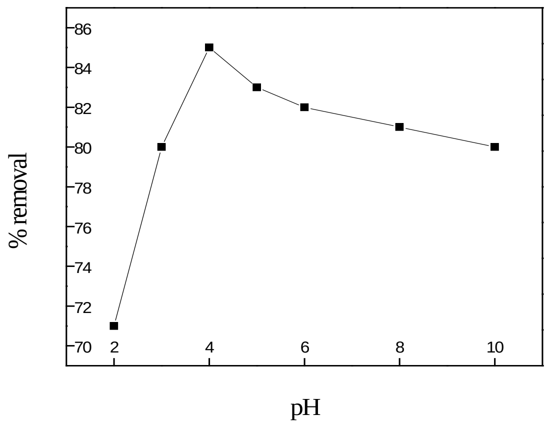


Fig 2: Effect of pH on removal of fluoride by Neem leaves at initial concentration of 14ppm, contact time of 60min, adsorption dose of 10gm/l.

shown in Figure.2.It is evident that fluoride removal reached maximum 85% at pH 4.

*Effect of Contact Time*

The studies were conducted by varying contact time from 15 to 120 minutes. The result shown that as increased the time the percentage of removal was also increased and after some time decreased. In the present case, the optimum contact time was obtained at 90 min and the influence of the time on sorption is shown in Figure. 3.

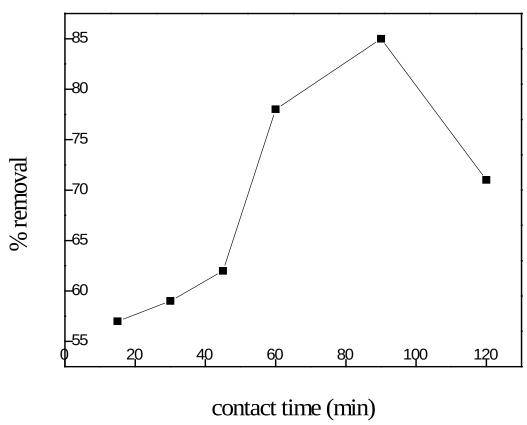


Fig 3: Effect of contact time on removal of fluoride by Neem leaves at initial concentration of 14 ppm, pH of 4, adsorption dose of 10gm/l

There was slow removal (%) of fluoride after 90 minutes so this time period was taken as optimum time for this process. Vacant adsorption sites, high solute concentration gradient, electrostatic affinity and ion – exchange may attribute to the higher removal in the initial stages (Mariappan et al., 2002).

*Effect of Temperature*

It is found that the removal of fluoride ions increases with increase in contact time to some extent. Further increase in contact time does not increase the uptake due to deposition of fluoride ions on the available adsorption sites on adsorbent material. The influence of the temperature on sorption rate is shown in Figure.4. From temperature study the optimum temperature was found 45$^{o}$C.

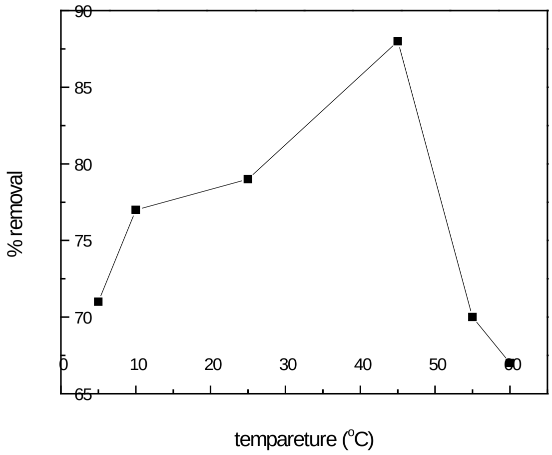


Fig 4: Effect of temperature on removal of fluoride by Neem leaves at initial concentration of 14 ppm, pH of 4, and adsorption dose of 10 gm/l.

*Effect of Adsorbent Dose*

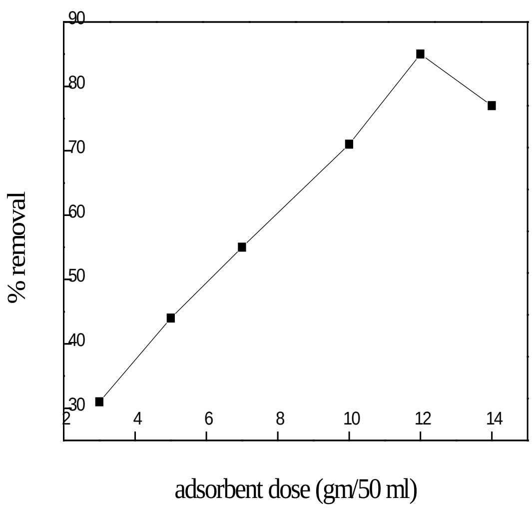


Fig 5: Effect of adsorbent dose on removal of fluoride by Neem leaves at initial concentration of 14 ppm, pH of 4, and temperature of 45$^{o}$C

From Figure. 5, it is observed that the removal of fluoride ions increases with an increase in the amount of adsorbent. For all these runs, initial fluoride ion concentration was fixed at 14 mg/l. The amount of adsorbent dose was varied between 3.0 and 14 g/l in aqueous solution at their optimal pH values. Results showed that maximum removal of 85% was observed at 12 g/l at temperature of 45$^{o}$C.

## V. ADSORPTION ISOTHERMS

*Adsorption isotherm*

The equilibrium relation between the amount of adsorbed q (mass of adsorbed per unit mass of the adsorbent) and the concentration (C) of the adsorbate or solute at a constant temperature, is called an adsorption isotherm. A few mathematical forms of isotherms, which can be fitted with experiment data, have been given here.

*Freundlich Isotherm*

The empirical Freundlich isotherm represents adsorption on a surface that is energetically non-uniform. As a result, the heat of adsorption at different sites is not the same. This isotherm is practically useful and assumes

$$\mathbf{q = K'C^{\frac{1}{n}}} \quad \text{................ (3)}$$

There are two adjustable parameters. One is n (value of n varies between 1 and 5) and other one is K. where q is the amount of solute adsorbed per unit weight of material (mg/g), n is adsorption intensity.

*Langmuir Isotherm*

The Langmuir isotherm is based on the simplest model of physical adsorption which assumes that (i) the molecules are adsorbed at discrete active sites on the surface (ii) each active site adsorbed one molecule only(iii) the adsorbing surface is energetically uniform(iv) there is no interaction among the adsorbed molecules. Langmuir equation can be rearranged in the form

$$\mathbf{\frac{C}{q} = \frac{1}{q_m K} + \frac{C}{q_m}} \quad \text{............. (4)}$$

$q_m$ is the maximum adsorption capacity (mg/g),

The two adjustable parameters are $q_m$ and k.

*Adsorption Isotherm*

The sorption isotherm studies were conducted by varying the adsorbent dosage of 3.0 g/l to 14 g/l and maintaining initial concentration of fluoride from 14 mg/l. Adsorption isotherm parameters of neem leaves are given in Table 1

TABLE 1: CALCULATION OF LANGMUIR AND FREUNDLICH ISOTHERM:

| Dose (g/l) | $C_i$ (mg/l) | $C_e$ (mg/l) | $C_i$-$C_e$ | $Q_e$(mg/g) |
|---|---|---|---|---|
| 3 | 14 | 9.6 | 4.4 | 0.073333 |
| 5 | 14 | 7.8 | 6.2 | 0.062 |
| 7 | 14 | 6.2 | 7.8 | 0.055714 |
| 10 | 14 | 3.95 | 10.05 | 0.05025 |
| 12 | 14 | 2 | 12 | 0.05 |
| 14 | 14 | 3.16 | 10.84 | 0.038714 |

| $1/C_e$ | $1/Q_e$ | $lnC_e$ | $lnQ_e$ |
|---|---|---|---|
| 0.104167 | 13.63636 | 2.261763 | -2.61274 |
| 0.128205 | 16.12903 | 2.054124 | -2.78062 |
| 0.16129 | 17.94872 | 1.824549 | -2.88752 |
| 0.253165 | 19.9005 | 1.373716 | -2.99074 |
| 0.5 | 20 | 0.693147 | -2.99573 |
| 0.316456 | 25.83026 | 1.150572 | -3.25155 |

*Freundlich Isotherm*

Freundlich equation has the general form of $q_e = K_f C^{1/n}$.

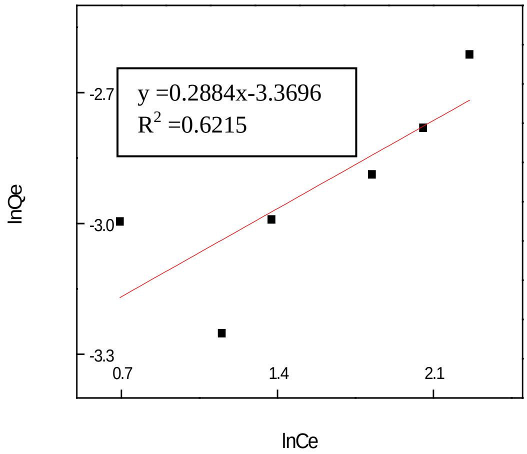


Fig 6. Linear model of Freundlich isotherm

From above Figure 6, the observation from the study are given below.The value of k and n are 29.06 mg/g and 3.46 respectively.

Langmuir Isotherm-

The observation from Figure. 7 are given below

The values of Langmuir parameters, $q_m$ and k are 0.068 and 0.847 respectively.

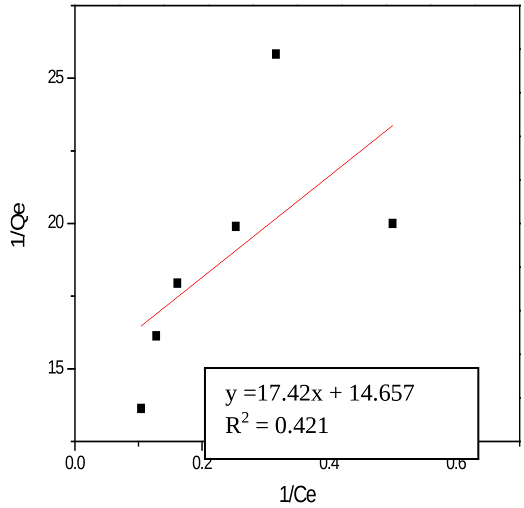


Fig 7. Linear model of Langmuir isotherm

TABLE 2: COEFFICIENT OF CORRELATION OF NEEM LEAVES FOR DIFFERENT ISOTHERMAL EQUILIBRIUM MODELS

| Langmuir Isotherm | | Freundlich isotherm | | |
|---|---|---|---|---|
| $q_m$ (mg/g) | $R^2$ | K | n | $R^2$ |
| 0.068 | 0.421 | 29.06 | 3.46 | 0.6215 |

*Adsorption kinetics*

The pseudo-first-order, pseudo-second-order and intraparticle diffusion kinetic models were used to investigate the adsorption kinetics of fluoride and to quantify the extent of uptake in the adsorption process. A simple pseudo first-order kinetic model, pseudo second-order model, intra-particle diffusion model are represented by the as given below:

Pseudo first-order-

$$\mathrm{Log}(q_e - q_t) = \log(q_e) - \frac{K_1 t}{2}.303 \quad \ldots\ldots (5)$$

*Pseudo second-order*

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \left(\frac{1}{q_t}\right)t \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (6)$$

*Kinetics of adsorption*

Kinetics of adsorption is one of the most important characteristics to be responsible for the efficiency of adsorption. To investigate the mechanism of adsorption Lagergren, Morris-Weber pseudo-first order and Ho and McKay pseudo-second order kinetic models have been applied for the experimental data to predict the adsorption kinetics. The pseudo second-order rate model given by Ho and McKay Equation was also applied to investigate the data for kinetics of sorption.

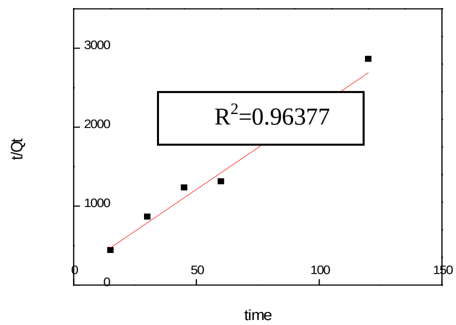


Fig 8. Ho and McKay model of fluoride onto neem leaves adsorbent.

Figure 8 shows a plot of t/ct versus t, which exhibits a straight line with $R^2$ 0.96377 values for fluoride ion. It is evident that the kinetic model follows the Ho and McKay model.

*Instrumental analysis of adsorbent*

*Scanning electron microscope (SEM) analysis*

SEM analysis is another useful tool for the analysis of the surface morphology of an adsorbent. The porous and irregular surface structure of the adsorbent can be clearly observed in the SEM image shown in Figure10. The heterogeneous pores and cavities provided a large exposed surface area for the adsorption of fluoride. The size of pores is clear indication of the expected adsorption of fluoride onto the surface of the adsorbent.

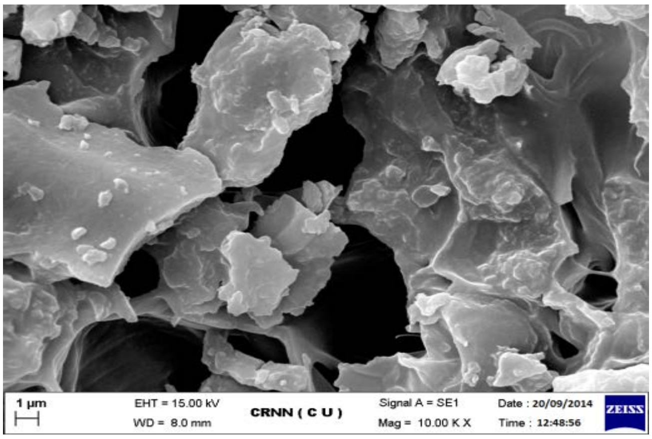


Fig 9: Scanning Electron micrograph of pure neem leaves before adsorption

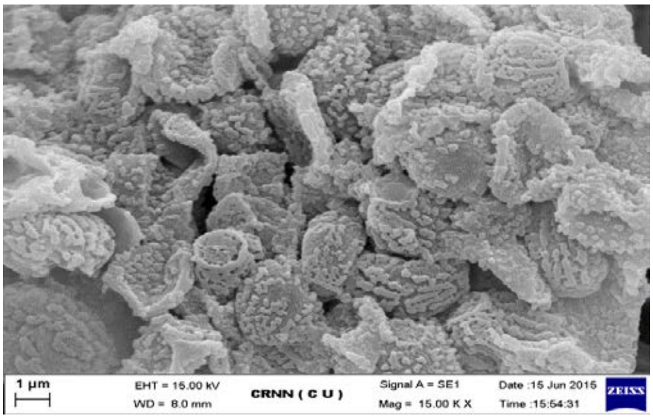


Fig 10: Scanning Electron micrograph of pure neem leaves after adsorption

Fourier transforms infrared spectra (FTIR) analysis- The FTIR spectrum of adsorbent was recorded to obtain the information regarding the stretching and bending vibrations of the functional groups which are involved in the adsorption of the adsorbate molecules. The FTIR analysis result of the adsorbent before and after the adsorption was given in figure (11,12).The FTIR spectral analysis of adsorbent shows the band at 3257cm-$^{1}$, 2916cm$^{-1}$ which are indicate the presence of N-H stretch, C-H stretch. The band at 1717cm-$^{1}$ indicates the presence of C=O stretch. The band at 1603cm$^{-1}$, 1540cm$^{-1}$, 1465cm$^{-1}$ indicates the presence of C-C stretch, asymmetric stretch, C-H bend.C-O stretch and the C-N stretch present at the band 1099cm$^{-1}$ and 1227cm$^{-1.}$Absorption at 526cm$^{-1}$ is due to carbon-halide stretching vibration. After adsorption of fluoride spectral shift was observed and an additional peak appears at 668cm$^{-1}$.

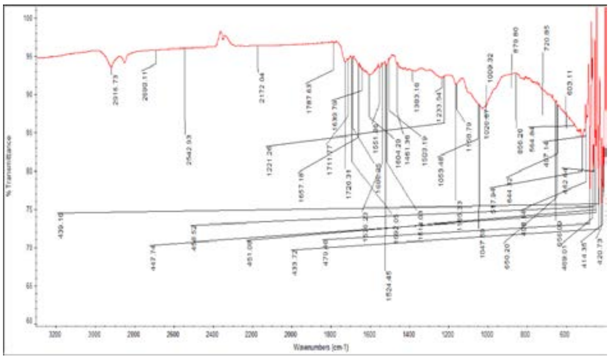

Fig11: -FTIR result of pure adsorbent before adsorption.

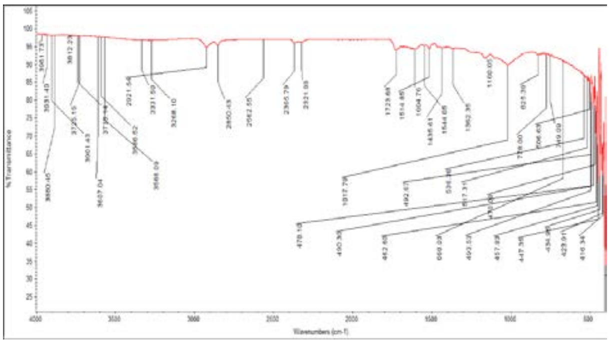

Fig 12:-FTIR result of pure adsorbent after adsorption.

## V. CONCLUSION

The experiment was aimed to remove the fluoride ion concentration from aqueous solution by using natural adsorbents (neem leaves) using adsorption technique. The adsorption of fluoride depends on pH, initial concentration, contact time, temperature as well as adsorbent dose which were optimized. The equilibrium agitation time for the adsorption of fluoride is 90 minutes. Percentage removal is increased up to pH of 4.The removal rate is increased with increasing the adsorbent dose and at 12 gm/l adsorbent dose the removal was found to be maximum. In the following conditions maximum removal was achieved 85%. According to Freundlich isotherm the value of $R^2$ is 0.6215 therefore the adsorption by adsorbent followed strictly the Freundlich isotherm and is well described by pseudo second order kinetics. The present study concluded that the neem leaf material as a natural adsorbent can be utilized effectively for fluoride removal.

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