A Comparative Study Of Defluoridation Of Water Using Bioadsorbents- Tea Waste And Rice Husk.

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Abstract- The water available for daily consumption may be contaminated by natural sources or by industrial effluents. One such a contaminant is fluoride. In drinking water fluoride has a number of adverse effects on human health. There are many naturally available alternatives for defluoridation of water like tea waste, seed powder of Moringa Oleifera, powder of bark of Pipal tree, Rice husk etc. Here the comparison is done between adsorption efficiency of Tea waste and Rice Husk powder. Using colorimetric method tests were conducted to get optimal values of pH of water in acid & alkali treated powder of both. Optimal contact time for 212µ and 600µ were determined. For higher removal percentage efficiency of fluoride, optimal dosage of adsorbent was also determined.

Indexed Terms- Fluoride, adsorption, Tea Waste, Rice Husk,, contact time, adsorbent dose, P^H, particle Size.

I. INTRODUCTION

Pure water is scarce and is not easily available to all. Deprived sections of the society consume contaminated water and take ill periodically, often resulting in epidemics. The water may be contaminated by natural sources or by industrial effluents. One such a contaminant is fluoride. Geological formation is the main source of fluoride in the groundwater. The other sources of fluoride occurrence in water are industrial discharge from aluminum industries, phosphate industries, coal plants as well as due to water, food, air, medicines and cosmetics. Removal of fluoride from water is important because of its ill-effects for human. Defluoridation is removal of fluoride from water. Although there are several sources of fluoride intake, it is roughly estimated that about 60% of the total intake is through drinking water. There are various methods adopted for fluoride removal but they are costlier and some are time consuming. Hence, there is an urgent need of developing low cost method of defluoridation.

Fluoride is one such compound that is widely present in groundwater worldwide. Exposure to fluoride in drinking water has a number of adverse effects on human health including crippling skeletal fluorosis that is a significant cause of morbidity in a number of regions of the world. Fluoride is more toxic than lead even in minute dose as it accumulates in brain damaging it, also affects on mind development of children. Ground water is polluted due to industrial effluents and municipal waste in water bodies. In rural and undeveloped countries people living in extreme poverty are presently drinking highly turbid and microbiologically contaminated water. They lack knowledge of proper drinking water treatment and they do not afford costly chemical coagulants. To overcome chemical coagulant problems it is necessary to increase the use of natural coagulants for drinking water treatment. Naturally occurring coagulants are usually presumed safe for human health. Some studies on natural coagulants have been carried out and various natural coagulants were produced or extracted from microorganisms, animals or plants.

The use of Tea ash as well as Rice Husk as an added advantage over the chemical treatment of water because it is biological and has been reported as edible. Hardness removal efficiency of both was found to increase with increasing dosage .Tea Ash acts as a natural adsorbent and antimicrobial agent as well. Tea ash has been shown to be one of the most effective as a primary coagulant for water treatment and can be compared to that of alum a conventional chemical coagulant. The leaves and powder can be stored but the paste needs to be fresh for purifying the water.

Tea Plant Fig. I

Rice Husk Fig .II

Rice Husks are the hard protecting covering of grains of rice. Rice Husks are the waste materials after the rice grains have been removed. Some industries or rice mills uses these material as fuel& feed for generation of electricity Earlier studies have found as Rice Husk to be non-toxic and recommended it for use as a coagulant. In India approximately 20 million tons of Rice Husk is produced. The use of Rice Husk has an added advantage over the chemical treatment of water because it is biological. Among all the plant materials that have been tested over the years, powder processed from the Tea Ash & Rice Husk has been show effective bioadsorbents. From the waste materials we can develop very effective, cheap & useful method of defluoridation of water.

II. LITERATURE REVIEW

In Nalgonda technique[16]a sequential addition of sodium aluminates or lime, bleaching powder and filter alum to the fluoride water is followed by flocculation, sedimentation and filtration. The disinfection is ensured by sodium aluminates settlement of precipitate and bleaching powder. The poly aluminum hydroxylsulphate (PAHS) was used for defluoridation studies[10] and it was concluded that, flocks formation and settling are quick and volume of resulting sludge is less. The use of Aloe Vera (Indian aloe), a medicinal plant was done[14]concluding maximum defluoridation at neutral pH .A study of defluoridation methods was done[20] by using indigenous chemicals and minerals. Moringa Oliefera [2] is one of the effective natural adsorbent for defluoridation of water. Tea ash [21] & Tamrind seeds[25] are also studied. For optimum alum dose, alkalinity of water, pH and colloidal concentration, effective defluoridationcan be ensured. The fluoride removal technique by using poly aluminum chloride (PAC) wasstudied[14] and compared with Nalgonda technique. It was observed that PAC can be an effective coagulant for fluoride removal with higher removal efficiency of about 65% -75% with less detention time. Also it was observed that fluoride removal was dependent on initial fluoride ion concentration and dose of coagulant. Defluoridation techniques by using lignite rice husk and rice husk powder as adsorbent was studied[18] by varying pH, concentration of fluoride, weight of adsorbent and contact time. A better result was obtained after 2 hrs at pH 6 by using lignite. Tea waste [21] is also one of the economical bioadsorbent. Fishbone charcoal prepared from fishbone was used for defluoridation studies. The fluoride removal was found to be a function of pH, contact time, initial fluoride ion concentration and adsorbent (fishbone charcoal) dose. The boiler bottom ash was successfully used[4]as an adsorbent material for separating fluoride from water for the pH between 6to 7.

- Effects of Fluoride on Human Health

The effects of fluoride on human health were dependent on the concentration of fluoride in Water.

Permissible Limits for Fluoride Concentration in Drinking Water by various agencies**:**

Table 1: Biological Effects on Human Health

Fluoride conc. (mg/lit	Source	Effects

0.002	Air	Destructive effect on plants
1.00	Water	Prevention of dental caries
2	Water	Effect dental enamel
3 to 6	Water	Osteoporosis
8	Water	10 % Osteoporosis
20 to 80	Air & Water	Crippling skeletal fluorosis
50	Food & water	Changes in thyroid
100	Food & water	Defective development
>125	Food & water	Changes in Kidney
2500	Food & water	Changes in Kidney

1. Indian Council of Medical Research (ICMR-1975) - 1 mg/lit
2. Bureau of Indian Standards (BIS)-0.6 to 1.2 mg/lit
3. World health Organization (WHO-1984) for drinking water-1 to 1.5 mg/lit
4. World Health Organization (WHO) European Standards -0.7 to 1.7 mg/lit related to temperature.

III. MATERIALS AND METHODS

In the present study, it is proposed to use the adsorption method by using natural adsorbent. Adsorption is defined as the change in concentration at the interfacial layer between the two phases of a system due to surface forces. Adsorption is mass transfer operation in that a constituent in the liquid phase is transferred to the solid phase. The adsorbent is the solid, liquid, or gas phase onto which the adsorbate accumulates. Factors affecting adsorption methods are surface area, nature of adsorbate, pH, temperature, presence of mixed solutes and nature of adsorbent.

The Camellia Sinesis (Tea plant) is evergreen plant that mainly grow in tropical and subtropical climate around the world and they have been used in drinking water treatment in small scale in Sudan and India for generations. The coagulant in the powder is believed to be one or several proteins that act as a cationic polyelectrolyte. The soluble particles in the water attached to the active agent, that binds them together creating large flocs in water. Due to the small size of the Tea ash &Rice Husk coagulant protein, a bridging effect may not be considered as the likely coagulation mechanism .The use of Camellia Sinesis (Tea Ash) has an advantage over the chemical treatment of water because it is biological and has been reported as edible. It is believed that the seed is an organic natural polymer. The active ingredients are dimeric proteins. The protein powder is stable and totally soluble in water.

India is the first country in production of rice among top 10 countries in the world. India produces approximately 43 millions of hectors rice per year. So rice husk is easily available, waste material & biological .We can solve, Rice Husk disposal problem also. In the present study synthetic sample is prepared and used for experimental purpose of defluoridation of water.

Tea Ash Powder Fig. III

Rice Husk Powder Fig. IV

- Materials used

For Tea Ash- Regular tea ash powder is used .For rice husks, husks were collected after removing the rice. INDRAAYANEE rice sample is used as adsorbent for tests.

For preparing synthetic fluoride water sample anhydrous sodium fluoride (NaF) and distilled water was used. The nitric acid (1 N HNO3) was used for acid washing of adsorbent. The sodium hydroxide (0.5N NaOH) was used for alkali washing of adsorbent. The adsorbent powder acid washed or alkali washed was further used. For fluoride detection studies with spectrophotometer, various solutions were prepared. Reference solution was prepared by using conc. hydrochloric acid (HCL) and SPADNS reagent (zirconyl chloride octahydrate).

Synthetic fluoride bearing water sample having initial fluoride ion concentration of 10mg/lit was used. The sample was filtered by using Whatmann's filter paper no. 41 for further uses. In this filtrate, SPADNS and zirconyl acid solution of 5 ml each was used. The sample was checked for fluoride detection in spectrophotometer at wavelength 570nm. Absorbance readings were compared with standard curve and the removal efficiency was found.

- For preparing synthetic fluoride water sample anhydrous sodium fluoride (NaF) and distilled water were used.

40 gm of powder sample was added to 400 ml of 1N HNO_3 for acid treatment and 0.5N NaOH for alkali treatment. The mixture was boiled for about 20 minutes. Washing of the powder sample was carried out by using distilled water until maximum color was removed and clear water was obtained. Finally, it was dried again in an oven at 50℃ for 6 hrs.

- The nitric acid (1N HNO_3) was used for acid washing of adsorbent.
- The sodium hydroxide (0.5 N NaOH) was used for alkali washing of adsorbent.
- For fluoride detection studies with spectrophotometer, various solutions were prepared.
- Reference solution was prepared by using conc. hydrochloric acid (HCl), SPADNS reagent zirconyl chloride, octahydrate reagent were used.

Synthetic fluoride bearing water sample having initial fluoride ion concentration of 10 mg/lit used. The sample was filtered by using Whatmann's filter paper no.41 for further uses. In this filtrate, SPADNS and zirconyl acid solution of 5ml each was used. The sample was checked for fluoride detection in spectrophotometer at wavelength 570nm. Absorbance readings were compared with standard curve and the removal efficiency was found.

- Development of standard curve

The fluoride standard sample in the range of 1 mg/lit to 11 mg/lit was prepared by taking appropriate quantities of standard fluoride solution with distilled water. Then pipette 5 ml each of SPADNS solution and zirconyl acid solution to each standard and mix it well or 10 ml SPADNS and zirconyl acid mix solution. Avoid contamination. The spectrophotometer was set to zero absorbance with reference solution and absorbance readings of standard curve were obtained. Reference solution was used as a blank solution. Spectrophotometer used at 480 nm wavelength; filter number 4, sensitivity medium was taken as per standard procedure. The procedure is same for both the bioadsorbents.

Table II: Standard curve

Sr. No.	Initial Fluoride ion Concentration (mg/lit)	Absorbance Reading (for Tea Ash)	Absorbance Reading (for Rice Husk)
1	1	0.341	0.275
2	2	0.388	0.370
3	4	0.419	0.505
4	6	0.469	0.695
5	8	0.502	0.890
6	10	0.539	1.060

Fig. V- Spectrophotometer

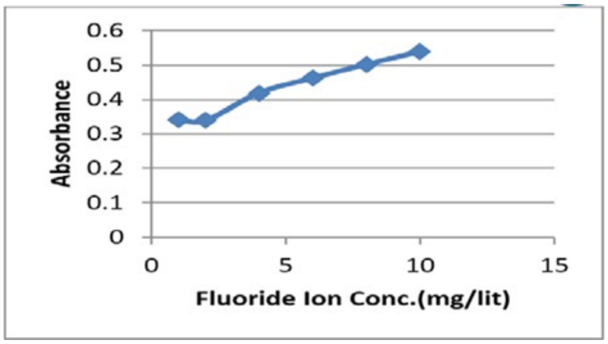

Fig.VI- Standard Curve –Tea Ash

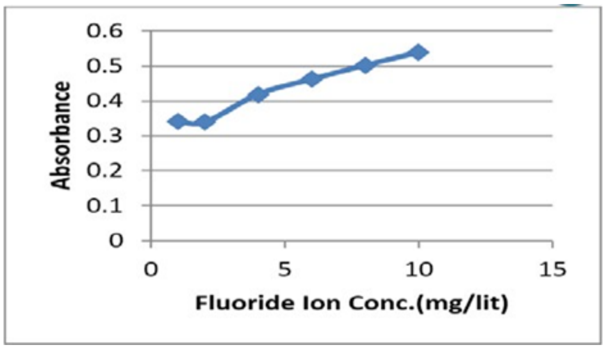

Fig.VII- Standard Curve -Rice Husk

IV. EXPERIMENTAL SETUP

The fluoride removal studies by adsorption were conducted in 250 ml conical flask using 100 ml of synthetic water sample containing different pH and initial concentrations of fluoride ion. In these conical flasks adsorbent with varied dosage was added. Then the contact period was given for different particle sizes. After giving the required contact time, the contents of the flasks were filtered using Whatmann's filter paper number 41. The filtrate was used for fluoride ion estimation using SPADNS method. The above procedure was repeated for different pH, contact times, adsorbent doses, particle sizes and different initial fluoride ion concentrations. The pH was varied from 1 to 10. The contact time was varied from 0.5 hrs to 3.5 hrs for various adsorbent sizes. The adsorbent dosages used were 0.5 gm/lit to 4 gm/lit in multiples of 0.5 gm/lit. The initial fluoride ion concentration was varied from 2 mg/lit to 11 mg/lit for the Camellia Sinesispowder (tea ash powder) as well as for Rice Husk Powder. The parameters were varied to find the maximum fluoride removal efficiency. Synthetic fluoride bearing water sample having initial fluoride ion concentration of 10 mg/lit was used. Under same experimental conditions percentage removal efficiency of fluoride content for two different bioadsorbents are calculated.

V. RESULTS AND DISCUSSIONS

A. Optimal pH

a) For Tea Ash-

The adsorbent having 600 μ size, acid washed as well as alkali washed, was used to determine optimal pH at which the adsorption was maximum. For these experiments initial fluoride ion concentration was 10 mg/lit, with adsorbent dose of 2.5 gm/lit and contact time of 1 hr.In case of acid washed adsorbent the maximum removal efficiency was 72 % at pH10. Whereas in case of alkali washed adsorbent the maximum removal efficiency was 62.85 % at pH 1. But the extreme pH values will give rise to higher costs for post treatment. It is generally recommended to maintain near neutral pH for the solutions. From figure it is seen that the percentage removal was 67 % and 47.35 % for acid washed and alkali washed adsorbents respectively.

Table III: Optimal pH -Tea Ash

Sr .No.	pH	Acid Washed Powder (600μ)		Alkali Washed Powder (600μ)	
		Absorbance	% Removal Efficiency	Absorbance	% Removal Efficiency
1	1	0.340	47	0.242	62.85
2	2	0.325	49	0.255	60.07
3	4	0.250	61	0.267	58.41
4	6	0.211	67	0.388	47.35
5	8	0.199	70	0.350	45.48
6	10	0.185	72	0.376	41.43

b) For Rice Husk-

The experiments were carried out for acid treated and alkali treated Rice Husk powder for determining optimum p^H. The procedure was similar for acid and alkali treated powder. The pH was varied from 1 to 10 for acid treated Rice Husk powder and 2 to 10 for alkali treated Rice Husk powder. The adsorbent having 600 μ size, acid washed as well as alkali washed, was used to determine optimal pH at which the adsorption was maximum. For these experiments initial fluoride ion concentration was 10 mg/lit, with adsorbent dose of 2.5 gm/lit and contact time of 1 hr. In case of acid washed adsorbent the maximum removal efficiency was 41 % at pH 1. Whereas in case of alkali washed adsorbent the maximum removal efficiency was 51 % at pH 10.

Table IV - Optimal pH- Rice Husk

Sr.No.	pH	Acid Washed Powder (600μ)		Alkali Washed Powder (212μ)	
		Absorbance	% Removal Efficiency	Absorbance	% Removal Efficiency
1	1	0.680	41	0.763	32
2	2	0.765	32	0.760	33
3	4	0.815	26	0.730	38
4	6	0.850	21	0.665	43
5	8	0.909	14	0.595	50
6	10	0.920	13	0.580	51

The extreme pH values will give rise to higher costs for post treatment. Therefore it is not advisable to adopt extreme pH values. It is generally recommended to maintain near neutral pH for the solution. Therefore at pH of 8, the percentage removal was 14 % and 50 % for acid washed and alkali washed adsorbents respectively.

In case of acid treated, the results were due to neutralization of the negative charge at treated Rice Husk bioadsorbent surface by greater hydrogen ion concentration at lower pH values, thus reducing hindrance to diffusion of the negatively charged fluoride ions on to the increased active surface of acid treated Rice Husk bioadsorbent.

In case of alkali treated Rice Husk bioadsorbent, the maximum removal observed at high p^H. It was due to increase of hydroxyl ion concentration in the solution, hence the rate of fluoride ion adsorption was maximum on the active surface, due to cation exchange phenomenon of alkali treated Moringa Oleifera bioadsorbents at high pH value. Therefore, it was decided to use alkali washed adsorbent and to maintain pH 8.

B. Optimal Contact Time

a) For Tea Ash-

The experimental study was carried out to determine optimal contact time using adsorbents with different particle sizes. The pH was 8 and dose was 2.5 gm/lit for the study. From figure, it is seen that the contact time reduces with decrease in particle size. For the given particle size, after a particular contact time, the removal efficiency remains almost constant. Therefore contact time of 2 hrs to 2.5 hrs were optimal for adsorbents with particle size of 212 µ and 600 µ. A 480 nm wavelength, providing a light path of at least 1 cm. is used.

Table V- Optimal contact time

Sr.No.	Contact Time (min)	Acid Washed Powder (600µ)		Alkali Washed Powder (212µ)	
		Absorbance	% Removal Efficiency	Absorbance	% Removal Efficiency
1	30	0.392	39	0.372	42
2	60	0.338	47	0.319	50
3	90	0.294	54	0.264	59
4	120	0.243	62	0.224	65
5	150	0.212	67	0.206	68

b)For Rice Husk-

The adsorbent dose of 2.5 gm/lit was taken and kept constant throughout the experimental work. The contact time was varied from 0.5 to 2.5 hrs for alkali treated Rice Husk powder of 600µ and 212µ respectively. The experimental study was carried out to determine optimal contact time using adsorbents with different particle sizes. The pH was 8 and dose was 2.5 gm/lit for the study. It is seen that the contact time reduces with decrease in particle size. For the given particle size, after a particular contact time, the removal efficiency remains almost constant. Contact time of 2 hrs to 2.5 hrs were optimal for adsorbents having particle size of 212 µ and 600 µ.

Table V- Optimal contact time

Sr.No.	Contact Time (min)	Acid Washed Powder (600µ)		Alkali Washed Powder (212µ)	
		Absorbance	% Removal Efficiency	Absorbance	% Removal Efficiency
1	30	0.680	40	0.565	53
2	60	0.615	49	0.480	62
3	90	0.545	57	0.520	68
4	120	0.495	62	0.415	70
5	150	0.485	63	0.415	70

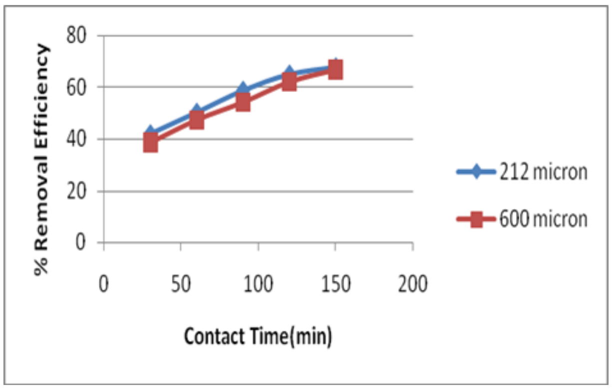

Fig.VIII- Optimal contact time (Tea Ash)

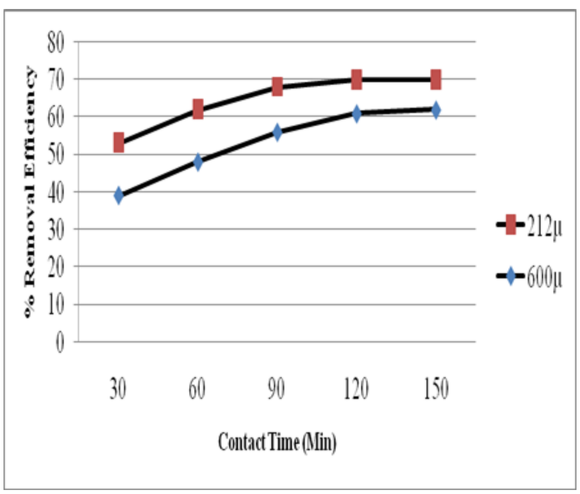

Fig.IX- Optimal contact time (Rice Husk)

C.OPTIMAL ADSORBENT DOSE-

a) For Tea Ash-

It was seen that the removal of fluoride increases with an increase in the amount of adsorbent. For all the experiments, initial fluoride ion concentration was fixed at 10 mg/lit, pH 8, and optimum contact time 2 hrs. to 2.5 hrs for 600 µ and 212 µ. The amount of adsorbent dose was varied from 0.5 gm/lit to 400 mg/lit in aqueous solutions. Results show that for 212µ alkali treated tea ash bioadsorbent, the maximum removal efficiency of fluoride was 74 % at 400 mg/lit. Similarly, for 600µ alkali treated tea ash bioadsorbent, the maximum removal efficiency of fluoride was 72 % at 400 mg/lit.

Table VII- Optimal adsorbent dose

Sr.No.	Adsorbent dose (mg)	(600µ)		(212µ)	
		Absorbance	% Removal Efficiency	Absorbance	% Removal Efficiency
1	100	0.367	43	0.343	47
2	200	0.298	54	0.282	56
3	300	0.245	62	0.230	64
4	400	0.179	72	0.167	74

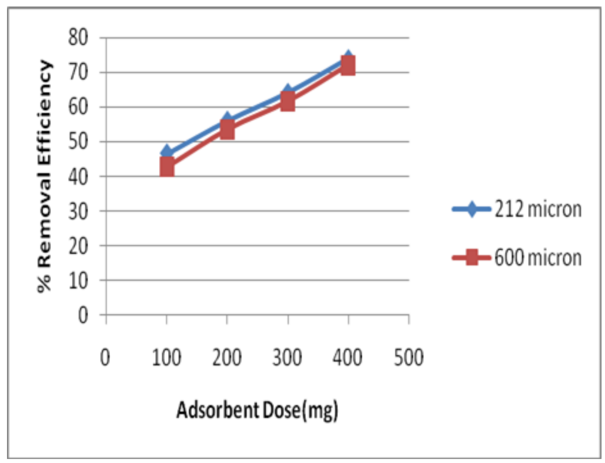

Fig.X- Optimal adsorbent dose (Tea Ash)

b) For Rice Husk-

It was seen that the removal of fluoride increases with an increase in the amount of adsorbent. For all the experiments, initial fluoride ion concentration was fixed at 10 mg/lit, pH was 8, and optimum contact time was 2.5 hrs and 2 hrs for 600 µ and 212 µ. The amount of adsorbent dose was varied from 0.5 gm/lit to 4 gm/lit in aqueous solutions. Results show that for 212µ alkali treated Rice Husk bioadsorbent, the maximum removal efficiency of fluoride was 76 % at 400 mg/lit. Similarly, for 600µ alkali treated Rice

Husk bioadsorbent, the maximum removal efficiency of fluoride was 68 % at 400 mg/lit.

Table VIII Optimal adsorbent dose

Sr .No.	Adsorbent dose (mg)	(600µ)		(212µ)	
		Absorbance	% Removal Efficiency	Absorbance	% Removal Efficiency
1	100	0.620	47	0.615	48
2	200	0.525	58	0.485	62
3	300	0.490	61	0.525	67
4	400	0.520	68	0.365	76

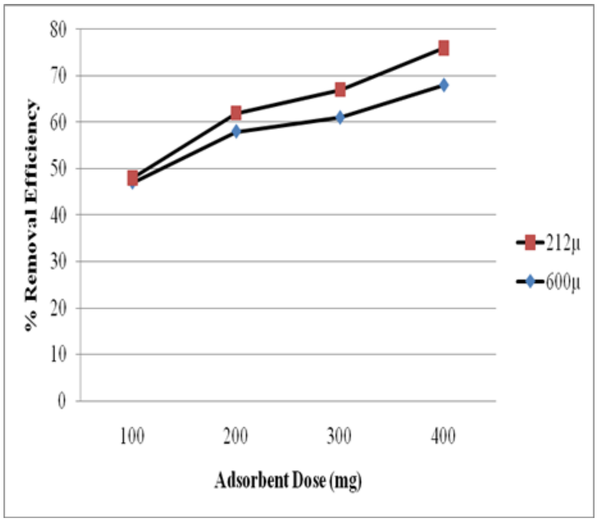

Fig.X- Optimal adsorbent dose (Rice Husk)

VI. IMPORTANCE OF WORK

Based on the present study following conclusions are drawn-

1. Use of Tea Ash Powder & Rice Husk powder as bioadsorbents for removal of fluoride is feasible.
2. Acid treated Tea Ash powder gives good results for Fluoride removal from neutral to higher P^H scale, while Alkali treated Tea ash powder is found better from acidic P^H to neutral P^H scale.
3. Alkali treated Rice Husk powder is better than acid treated Rice Husk powder for all P^H scales ,for fluoride ion removal.
4. The removal of Fluoride ion by adsorption increases as the p^H value increases for both bioadsorbents. But optimal PH is neutral.
5. The optimum contact time was 2.5hrs for 212 µ and 600 µ both for Tea Ash & Rice Husk.
6. Better removal efficiency of Fluoride ion is observed for 212 µ particle size than 600 µ due to increase in surface area for less particle size.
7. The removal by adsorption is found to be optimum at adsorbent dose of 400 mg/lit. for both bioadsorbents.

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