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PREPARATION OF HERBAL FORMULATION FOR LOWERING FLUORIDE IONS IN DRINKING WATER

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ABSTRACT

The described are the findings of research project titled "Lowering of fluoride levels using herbal preparations in drinking water of Kachchh region" is funded by GUJCOST under minor research project scheme. The paper presents the results of defluoridation obtained by various herbals like amla, babool bark, chitosan, orange peel powder, coconut husk etc. The defluoridation was observed as a function of pH, dose of bio-adsorbent, time, concentration of fluoride after treatment and temperature. The herbals giving best results in combination were used to prepare a formulation which can be used for defluoridation purpose within safe drinking limits as prescribed by WHO and can be used by a layman household for daily use.

KEYWORDS: Defluoridation, fluoride ions, drinking water, Kachchh.

INTRODUCTION

It is a worldwide problem of getting pure and safe drinking water. Fluoride levels occurring in untreated drinking water are not same everywhere. The fluoride levels depend on the salt contents of soil, rocks, industries present in the vicinity etc. The WHO recommends safe fluoride levels in drinking water as NMT 1ppm. The fluoride levels in Mundra region of Kachchh is reported to be 2.4 – 3.5 mg/L and in Chhasara region it is reported to be in the range of 3.42 mg/L.[1] Some amount of fluoride is required to be present in drinking water as it is beneficial for health and growth but in exceeding concentrations above the recommended limits fluoride ion is found to have adverse effects on human health. It is reported that excess fluoride may cause Fluorosis, a disfigurement of teeth, causes and/or aggravates bone and joint diseases, and poses a higher the risks of fractures, aggravates diabetes and kidney

disease, damages thyroid gland by replacing necessary I_2, lowers IQ etc. The major problem is that with higher levels of fluoride present in drinking water everyone receives the same concentration regardless of age, size, activity level and state of health. A plethora of literature is available for the use of herbal base bioadsorbents like peepal, lime, amla, chitosan, coconut husk, sweetlime, babool bark and many more for the defluoridation of drinking water.[2-33]

EXPERIMENTAL SET UP

The standard fluoride solutions of strength 3ppm, 5ppm and 7ppm were prepared using NaF obtained from S D Fine Chemicals. Various herbals like coconut husk, babool bark, peepal leaves, neem leaves, amla powder were obtained from nearby areas. Chitosan was procured from S D Fine Chemicals. Reverse osmosis treated drinking water was used for the preparation of standard as well as working solutions. The defluoridation was observed as a function of time, pH, bio-adsorbent dose and temperature as reference to initial fluoride concentration.

The stock solution of fluoride was prepared by dissolving 0.221 g of sodium fluoride (NaF) in 1000 ml of double distilled water. The concentration of fluoride ions in the synthetic solutions, prepared from the standard solution, was kept between the ranges of 1.5 to 15 mg/L. Before mixing the adsorbent, the pH of each fluoride solution was adjusted to the required value by adding 0.1 M NaOH or 0.1 M HCl solution. All precautions were taken to minimize the loss due to evaporation during the preparation of solutions and subsequent measurements. The solutions were prepared fresh for each experiment as the concentration of the solution may change on long standing.

The bio-adsorbents used were in dried and powdered form. They were individually mixed with the standard solution of known fluoride concentration and at different periods of time were filtered through whatman filter paper no. 42 and analyzed for fluoride concentration present in solution after bio treatment. The analysis was done by using ISE (ion selective electrode method) method. After analysis, the percentage defluoridation was calculated by using the formula:

$$\% \text{ defluoridation} = \frac{F_s - F_t}{F_s} \times 100$$

Where, F_s is the fluoride concentrations of standard and treated water samples.

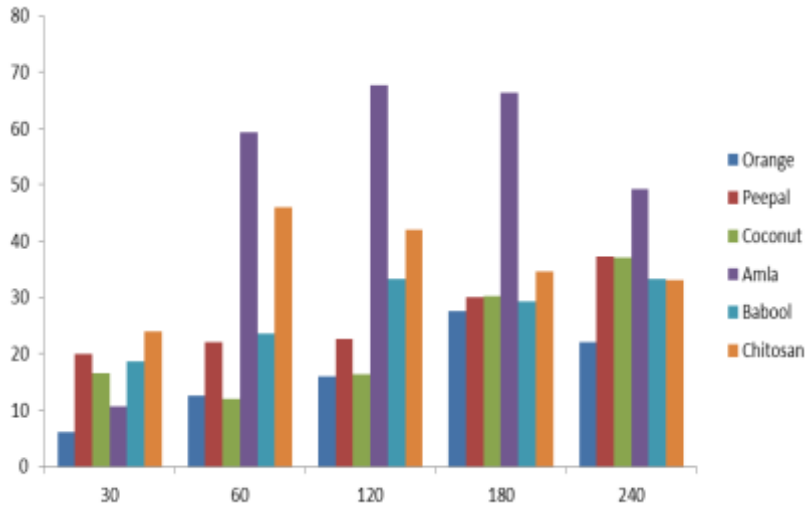
After evaluating the individual herbals for their fluoride adsorption capacity, best three defluoriding herbals were identified and combined with activated charcoal and silica and

hand moulded tablets were prepared using starch paste as a binder. The moulded tablets were prepared using 20% amla powder, 25% chitosan, 25% dried and powdered coconut husk, 20% activated charcoal and 10% silica. The dried tablets were tested for their fluoride adsorption capacity using 3ppm, 5ppm and 7ppm standard fluoride solutions and analyzed using ISE method. All the readings were taken in triplicate and performed on two different sets of solutions. Each time the changes in water characteristics post defluoridation with respect to total hardness, calcium hardness and acidity. Fluoride electrode and pH electrode are plugged in with instrument. Temperature rod was also connected with the instrument. The instrument was standardized with the freshly prepared 3 ppm, 5 ppm and 7 ppm standard solutions of fluoride. After standardization readings were taken for fluoride concentration using ion selective electrode and measure pH using pH electrode.

RESULTS AND DISCUSSION

Table 1: Gives the results obtained for different bio-adsorbents using 3ppm standard fluoride solution and 5g per l of bio-adsorbent concentration in water.

Bio-adsorbent	Avg. % Fluoride adsorption in time in minutes					Avg. pH	Avg. Temp. (°C)
	30 min	60 min	120 min	180 min	240 min		
Orange peel powder	6	12.6	16	27.6	22	5.3	20.2
Peepal leaves	20	22	22.6	30	37.3	7.4	20.2
Coconut husk	16.6	12	16.3	30.3	37	5.8	20.2
Amla powder	10.6	59.3	67.6	66.3	49.3	3.3	16.6
Babool bark	18.6	23.6	33.3	29.3	33.3	5.4	16.7
Chitosan	24	46	42	34.6	33	6.6	16.7



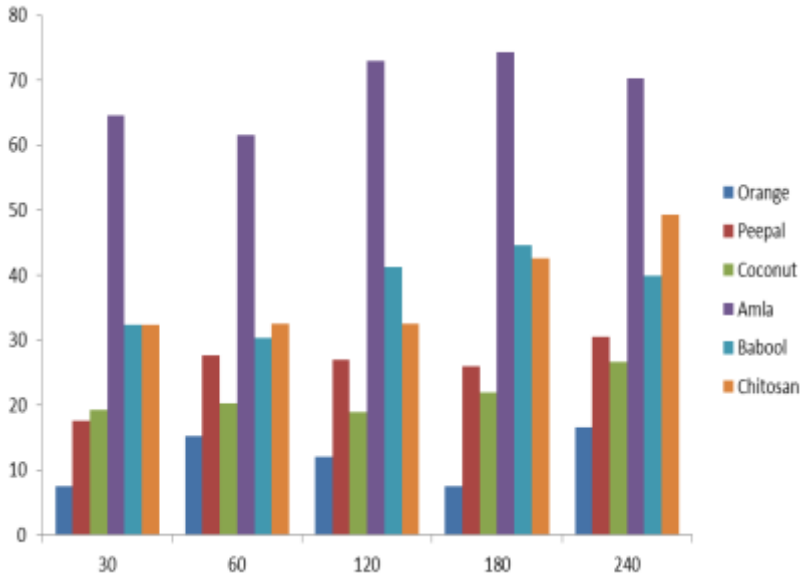
Graph 1: Gives the results obtained for different bio-adsorbents using 3ppm standard fluoride solution and 5g per l of bio-adsorbent concentration in water with time in minutes on x axis and % fluoride adsorption on y axis.

Table 2: Analysis of water parameter for different bio-adsorbents using 3ppm standard fluoride solution and 5g per l of bio-adsorbent concentration in water.

Sr. No.	Adsorbent Name	Total Hardness (mg/l)	Calcium Hardness (mg/l)	Acidity
1	Drinking water	12.5	15	275
2	Orange peel powder	26.5	22.5	350
3	Peepal leaf powder	126	47.5	725
4	Coconut husk	22.5	25	612.5
5	Amla powder	20	10	350
6	Babool bark powder	20	10	70
7	Chitosan	22.5	20	50

Table 3: gives the results obtained for different bio-adsorbents using 3ppm standard fluoride solution and 10g per l of bio-adsorbent concentration in water.

Bio-adsorbent	% Fluoride adsorption in time in minutes					Avg. pH	Avg. Temp. (°C)
	30 min	60 min	120 min	180 min	240 min		
Orange peel powder	7.6	15.3	12	7.6	16.6	5.1	20.3
Peepal leaves	17.6	27.6	27.0	26.6	30.6	8.0	2.3
Coconut husk	19.3	20.3	19.0	22.0	26.6	6.0	20.3
Amla powder	64.6	61.6	73.0	74.3	70.3	3.3	17.3
Babool bark	32.3	30.0	41.3	44.6	40.0	4.9	17.4
Chitosan	32.3	32.6	32.6	42.6	49.3	6.8	17.6



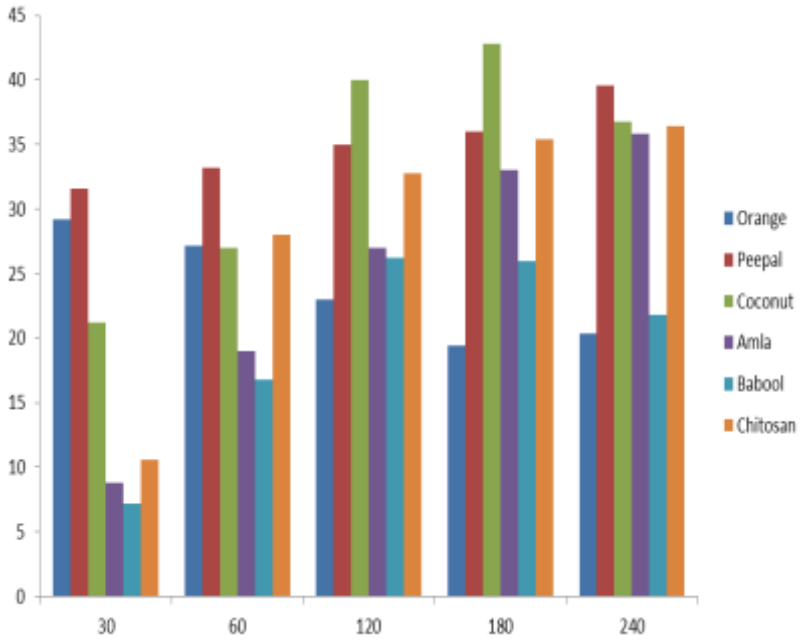
Graph 2: Gives the results obtained for different bio-adsorbents using 3ppm standard fluoride solution and 10g per l of bio-adsorbent concentration in water with time in minutes on x axis and % fluoride adsorption on y axis.

Table 4 Analysis of water parameter for different bio-adsorbents using 3ppm standard fluoride solution and 10g per l of bio-adsorbent concentration in water.

Sr. No.	Adsorbent Name	Total Hardness (mg/l)	Calcium Hardness (mg/l)	Acidity
1	Drinking water	15	12.5	75
2	Orange peel powder	45	70	650
3	Peepal leaf powder	180	85.5	450
4	Coconut husk	20	82.5	475
5	Amla powder	35	15	400
6	Babool bark powder	22.5	17.5	75
7	Chitosan	17.5	10	50

Table 5: Gives the results obtained for different bio-adsorbents using 5ppm standard fluoride solution and 5 g per l of bio-adsorbent concentration in water.

Bio-adsorbent	% Fluoride adsorption in time in minutes					Avg. pH	Avg. Temp. (°C)
	30 min	60 min	120 min	180 min	240 min		
Orange peel powder	29.2	27.2	23.0	19.4	20.4	5.5	20.5
Peepal leaves	31.6	33.2	35.0	36	39.6	7.6	20.4
Coconut husk	21.2	27.0	40.0	42.8	36.8	6.6	20.4
Amla powder	8.8	19.0	27.0	33.0	35.8	3.2	20.0
Babool bark	7.2	16.8	26.2	26.0	21.8	5.3	19.9
Chitosan	10.6	28.0	32.8	35.4	36.4	6.7	20.0



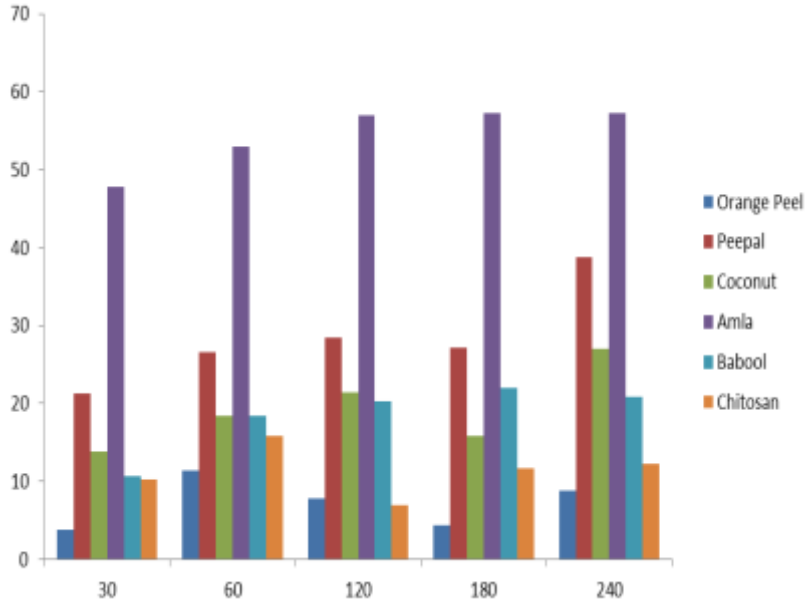
Graph 3: Gives the results obtained for different bio-adsorbents using 5ppm standard fluoride solution and 5 g per l of bio-adsorbent concentration in water with time in minutes on x axis and % fluoride adsorption on y axis.

Table 6: Analysis of water parameter for different bio-adsorbents using 5ppm standard fluoride solution and 5 g per l of bio-adsorbent concentration in water.

Sr. No.	Adsorbent Name	Total Hardness (mg/l)	Calcium Hardness (mg/l)	Acidity
1	Drinking water	17.5	47.5	350
2	Orange peel powder	20	25	250
3	Peepal leaf powder	115	57.5	625
4	Coconut husk	25	40	450
5	Amla powder	15	5	375
6	Babool bark powder	10	5	50
7	Chitosan	17.5	5	50

Table 7: Gives the results obtained for different bio-adsorbents using 5ppm standard fluoride solution and 10 g per l of bio-adsorbent concentration in water.

Bio-adsorbent	% Fluoride adsorption in time in minutes					Avg. pH	Avg. Temp. (°C)
	30 min	60 min	120 min	180 min	240 min		
Orange peel powder	3.8	11.4	7.8	4.4	8.8	5.0	19.9
Peepal leaves	21.2	26.6	28.4	27.2	38.8	7.8	20.0
Coconut husk	13.8	18.4	21.8	15.8	27.0	6.0	20.1
Amla powder	47.8	53.0	57.0	57.2	57.2	3.4	21.6
Babool bark	10.6	18.4	20.2	22.0	20.8	5.0	21.5
Chitosan	10.2	15.8	7.0	11.6	12.2	7.0	21.5



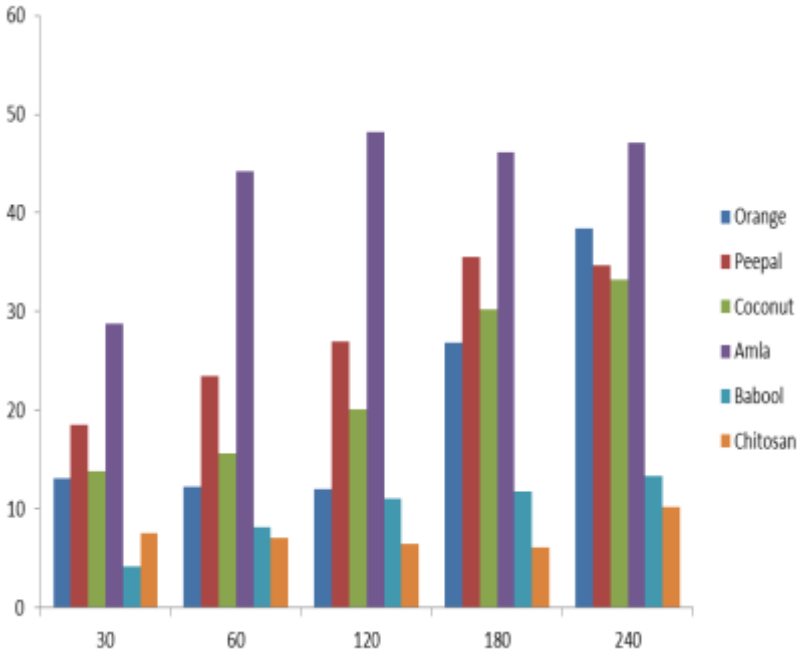
Graph 4: Gives the results obtained for different bio-adsorbents using 5ppm standard fluoride solution and 10 g per l of bio-adsorbent concentration in water with time in minutes on x axis and % fluoride adsorption on y axis.

Table 8: Analysis of water parameter for different bio-adsorbents using 5ppm standard fluoride solution and 10 g per l of bio-adsorbent concentration in water.

Sr. No.	Adsorbent Name	Total Hardness (mg/l)	Calcium Hardness (mg/l)	Acidity
1	Drinking water	30	15	75
2	Orange peel powder	45	30	850
3	Peepal leaf powder	105	70	400
4	Coconut husk	20	60	550
5	Amla powder	25	17.5	450
6	Babool bark powder	15	15	50
7	Chitosan	17.5	7.5	50

Table 9: Gives the results obtained for different bio-adsorbents using 7ppm standard fluoride solution and 5 g per l of bio-adsorbent concentration in water.

Bio-adsorbent	% Fluoride adsorption in time in minutes					Avg. pH	Avg. Temp. (°C)
	30 min	60 min	120 min	180 min	240 min		
Orange peel powder	13.1	12.2	12.0	26.8	38.4	5.5	21.1
Peepal leaves	18.5	23.5	27.0	35.5	34.7	7.7	21.3
Coconut husk	13.80	15.6	20.1	30.2	33.2	6.5	21.3
Amla powder	28.8	44.2	48.2	46.2	47.1	3.1	21.0
Babool bark	4.14	8.2	11.1	11.8	13.4	5.2	21.0
Chitosan	7.5	7.1	6.5	6.1	10.2	6.7	21.0



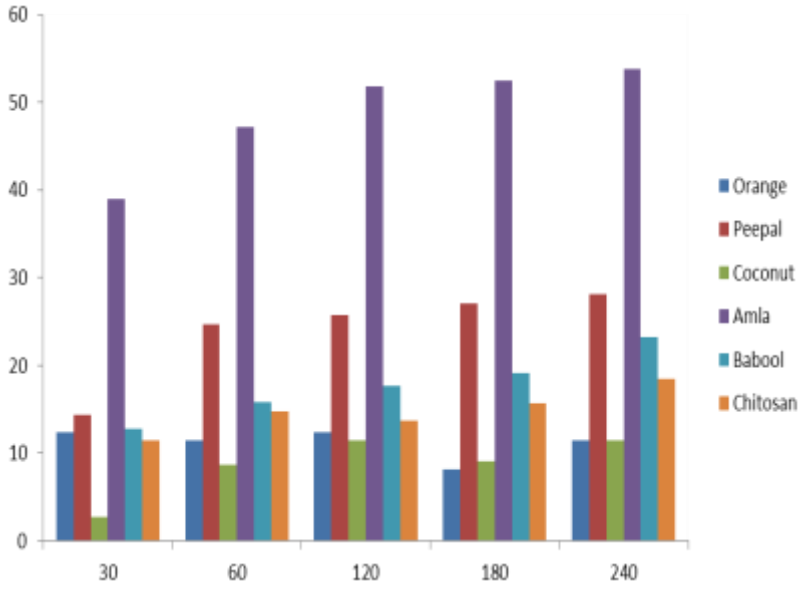
Graph 5: Gives the results obtained for different bio-adsorbents using 7ppm standard fluoride solution and 5 g per l of bio-adsorbent concentration in water with time in minutes on x axis and % fluoride adsorption on y axis.

Table 10: Analysis of water parameter for different bio-adsorbents using 7ppm standard fluoride solution and 5 g per l of bio-adsorbent concentration in water.

Sr. No.	Adsorbent Name	Total Hardness (mg/l)	Calcium Hardness (mg/l)	Acidity
1	Drinking water	30	15	250
2	Orange peel powder	45	30	850
3	Peepal leaf powder	105	70	400
4	Coconut husk	20	60	550
5	Amla powder	25	17.5	450
6	Babool bark powder	15	15	50
7	Chitosan	17.5	7.5	50

Table 11: Gives the results obtained for different bio-adsorbents using 7ppm standard fluoride solution and 10 g per l of bio-adsorbent concentration in water.

Bio-adsorbent	% Fluoride adsorption in time in minutes					Avg. pH	Avg. Temp. (°C)
	30 min	60 min	120 min	180 min	240 min		
Orange peel powder	12.4	11.5	12.4	8.1	11.4	5.1	18.7
Peepal leaves	14.4	24.7	25.7	27.1	28.1	7.5	18.7
Coconut husk	2.7	8.7	11.5	9.1	11.4	6.2	18.6
Amla powder	39.0	47.2	51.8	52.5	53.8	3.2	21.5
Babool bark	12.8	15.8	17.7	19.1	23.2	5.1	21.6
Chitosan	11.4	14.7	13.7	15.7	18.4	6.8	21.5



Graph 6: Gives the results obtained for different bio-adsorbents using 7ppm standard fluoride solution and 10 g per l of bio-adsorbent concentration in water with time in minutes on x axis and % fluoride adsorption on y axis.

Table 12: Analysis of water parameter for different bio-adsorbents using 7ppm standard fluoride solution and 10 g per l of bio-adsorbent concentration in water.

Sr. No.	Adsorbent Name	Total Hardness (mg/l)	Calcium Hardness (mg/l)	Acidity
1	Drinking water	10	5	50
2	Orange peel powder	17.5	15	150
3	Peepal leaf powder	65	35	75
4	Coconut husk	15	10	50
5	Amla powder	42.5	35	750
6	Babool bark powder	20	15	100
7	Chitosan	15	10	50

Table 13: Gives the results obtained for the moulded tablet using 3, 5 and 7 ppm standard fluoride solution and 25 g per l of bio-adsorbent concentration in water as a moulded tablet.

Bio-adsorbent	Conc. Of standard F^- solution	% Fluoride adsorbtion	Avg. pH	Avg. Temp. (°C)
Moulded tablet	3 ppm	64.5	6.8	23.5
	5 ppm	72.3	6.7	23.7
	7 ppm	69.7	6.9	23.7

Table 14: Gives the results obtained for the moulded tablet using 3, 5 and 7 ppm standard fluoride solution and 25 g per l of bio-adsorbent concentration in water as a moulded tablet.

Sr. No.	Adsorbent Name	Total Hardness (mg/l)			Calcium Hardness (mg/l)			Acidity		
		3ppm	5ppm	7ppm	3ppm	5ppm	7ppm	3ppm	5ppm	7ppm
1	Drinking water	10	30	10	12	15	5	75	75	50
2	Moulded tablet	32	22	25	17	25	10	89	85	72

CONCLUSION

This research work investigated on the removal of fluoride from drinking water through batch process by using different low cost materials. These were Orange peel powder, Peepal leaf powder, Coconut powder, Amla powder, Babool bark powder, Chitosan on individual basis as well as by using a moulded tablet prepared by selected bio adsorbents. All experiments were made as a function of different adsorption parameters (pH, initial fluoride concentration, adsorbent dose, contact time and temperature).

Physico-chemical parameter of this solution was also measured after end of every experiment and minimum changes in water property was observed in babool bark powder and in chitosan and maximum in peepal leaf powder and orange peel powder. Orange powder changes pH of

water from neutral to acidic, colour from colourless to orange. It also increases the total hardness, calcium hardness and acidity. Maximum removal of fluoride was observed within first 30 min. Peepal leaves powder increase TDS more than three times compare to normal drinking water but in pH there is no significant changes was observed. Colour is change from colourless to green and it also creates minor change in odour. Total hardness is increase ten times and acidity is increase two times than normal water. Concentration of fluoride is continuously decreased with time. Coconut husk powder changes the colour from colourless to light red. It showed changes in pH and TDS, and increase acidity of water. Concentration of fluoride is continuously decreased with time. Amla powder changes colour from colourless to light green. It also changes the pH from neutral to acidic and increases the acidity of water seven times than normal drinking water. Amla powder has one of the highest efficiency of removing of fluoride from water. Babool bark powder changes the colour of water from colourless to brown and does not change pH and TDS of water. It increases hardness two times more than normal water. Concentration of fluoride continuously decreased with time. Chitosan does not change any water properties like, colour, odour and pH, hardness, and acidity. Concentration of fluoride is continuously decreased with time.

In the experiments of fluoride removal by the moulded tablets satisfactory results were obtained. However there is a scope for further investigation with respect to different formulation concentration of these bio adsorbents and requirement of pre-treatment like heating etc for better results.

As, from this study conclude that Amla powder and Chitosan are very good fluoride adsorbents but on the basis of water sample study we observe that amla powder changes the physico-chemical properties of water, which is again a major problem, as chitosan does not changes any of the water properties it would be the best option for fluoride removal. A good scope is projected for use of moulded tablets which after optimization and further development can be used for defluoridation of drinking water on daily basis in a cost effective and easy manner.

Thus, the use of these adsorbents is a thoughtful and economic attempt for its valuable, necessitous and needy utilization for drinking treatment process in affected areas of Kachchh district in Gujarat.

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