Defluoridation of Drinking Water Using Tamarind (Tamarindus Indica) Fruit Shell by Dip-In-Technique

KEYWORDS
Defluoridation, Dip in technique, Tamarind fruit shell

ABSTRACT
Aim:- The aim of the study is to examine and determine the potential application of tamarind fruit shell in its natural form using dip in technique. Methodology:- Drinking water from two different places (Faridpur and C. B. Ganj) was collected and their fluoride concentration was checked. The tamarind fruit shell is cleaned with distilled water and then sun dried and ground to a fine powder. From each site 50 samples each of 100ml water were tested. Then dip-in-bags were made using cloth containing 25mg of this powder. These bags were then dipped in 100ml of each sample and kept overnight. Then the bags were removed and the fluoride concentration of the water was checked using SPADNS technique. Results:- The baseline fluoride concentration of the two drinking water samples were found to be 1.53ppm and 1.83ppm respectively. The mean fluoride concentration of water treated by tamarind fruit shell powder were found to be 0.76ppm (SD 0.3) and 0.91ppm (SD 0.4) respectively. Wilcoxon signed rank test showed the difference in the fluoride concentration of samples between pretest and posttest to be statistically significant at a p value <0.001. Conclusion:- The present study showed that the tamarind fruit shell in its natural form using dip-in-technique could be used as a potential biosorbing agent for the removal of fluoride ions from drinking water.

Introduction:-
Water is one of the basic needs of life on the earth. The quality of drinking water may be affected by micro organisms or excessive amounts of inorganic components such as fluoride dissolved from rocks in the water. In India, there are around 19 states affected by excess fluoride in drinking water, in which people are consuming drinking water with fluoride concentration beyond permissible limit of 1.5ppm. The fluoride present in drinking water would lead to health problem, when concentration of fluoride is greater than 1.5 ppm like fluorosis (WHO technical report series 846, 1994).

Fluorosis is mainly of two types: skeletal fluorosis and dental fluorosis. Skeletal fluorosis is not easily recognizable until the disease has developed to an advanced stage. Maximum ill effects of fluorides are detected in the neck, spine, knee, pelvic and shoulders joints. It also affects the small joints of the hand and foot (WHO Report, 1984). Dental fluorosis in the initial stages results in the tooth becoming coloured from yellow to brown to black. Depending upon the severity, it may be only discolouration of the teeth or formation of pits in the teeth. The discolouration on the teeth may be in the form of spots or as streaks (Arlappa N, Qureshi I & Srinivas R, 2013).

Defluoridation of drinking water has to be practiced, if ground water sources have fluoride levels beyond the recommended limit of 1.5ppm. Methods practiced for removal of excess fluoride can be divided broadly into two categories, namely precipitation and adsorption. Precipitation methods depend on the addition of chemicals to the raw water, which leads to the formation of fluoride precipitates or adsorption of fluoride onto the formed precipitate (Arlappa N. et al, 2013). Lime and alum are used either individually or in combination. The Nalgonda Technique, as developed in India in 1974, involves the addition of alum and lime. Limitations of these methods are: the daily addition of chemicals; large volume of sludge production; and not effective with water sources having high total dissolved solids (TDS) and hardness (Piddennavar R. & Krishnappa P., 2013).

In adsorption method, fluoride of raw water is retained on the adsorbent due to physical, chemical or ion exchange interactions. Although wide varieties of adsorbents have been used for defluoridation, activated alumina (AA) technology has been the method of choice in developed countries. AA, Alcoa F-1 is used in most of these studies and many defluoridation plants based on this technology have been installed.

In recent years, the use of low cost adsorbents has been investigated by many to remove fluoride from water. Such materials include: hydroxylapatite, calcite, fluor spar, quartz, flyash, silica gel, bone char, spent catalyst, zeolites, red mud, bentonite, clay chips, lateritic soils, nano-alumina, aluminum hydroxide coated rice husk ash, waste mud, diatomaceous earth and other related materials. However, the applicability of these low cost methods is limited either due to their low efficiency or lack of public acceptance. Therefore, it is of paramount importance to identify materials with high rate of removal, economically, socially and technically feasible for applications in rural communities (Nedunuri K., Nadavala K. & Abburi K., 2012).

Tamarind fruit cover is brittle, easily cracked shell of ripe tamarind pod that separates out from the fruit. This is available in plenty as a waste byproduct of tamarind pulp industry (Nedunuri K. et al, 2012). V. Ramanjaneyulu et al (2013) and Nedunuri Phani Kumar et al (2012) have already used tamarind fruit shell as an adsorbent in their studies. But techniques used in these studies were difficult to practice by common people. In this study we used a new technique of fluoride removal which is apparently easier than...
the techniques used in previous studies. Thus the aim of the study is to examine and determine the potential application of Tamarind fruit cover in its virgin form in removing fluoride ion from aqueous medium by dip in technique.

Methodology:-
Adsorbate:-
Drinking water from two different places C. B. Ganj (site 1) and Faridpur (site 2) from Bareilly district was collected. 50 samples each of 100ml (n=100) for both sites were collected in plastic containers.

Biosorbent (adsorbent):-
The shell was removed from ripened tamarind fruit, washed thoroughly with distilled water and completely dried under sun. Then this substrate was powdered, sieved with a standard screen mesh no. 52/75 to get a uniform sized powder and preserved.

Defluoridation by dip in method:-
Dip in bags made up of cotton cloth containing 25 mg of absorbent were prepared and dipped in each sample of 100 ml drinking water for overnight and then the cloth bag was removed in the morning from the plastic container. The tamarind fruit shell gave slight brown colour to the water sample which was decolourised by adding hydrogen peroxide (H2O2) drop by drop (around 8 – 10). Then the concentration of fluoride in the solution was checked using colorimeter by SPADNS technique.

SPADNS technique:-
This technique relies on the fact that when fluoride reacts with zirconium dyes (Zr-SPADNS (sodium 2-(parasulphophenylazo)-1,8-dihydroxy-3,6-naphthalene disulphonate)) a colourless complex anion and a dye are formed. The complex, which is proportional to the fluoride concentration, tends to bleach the dye which therefore becomes progressively lighter as the fluoride concentration increases. The resulting coloured complex is measured in a spectrophotometer at 570 nm. The instrument used in this study (HI 729, Hanna Instruments, USA) works with the same technique.

Statistical analysis:-
Wilcoxon signed rank test was used to compare the fluoride reduction between pre-test and post-test readings. Statistical significance was set at p < 0.05. The pre-test and post-test data was entered into MS excel sheet and SPSS (version 21.0) was used for data analysis.

Results:-
The baseline fluoride concentrations of the two sites were found to be 1.53ppm and 1.83ppm respectively. The mean fluoride concentration after treating with tamarind fruit shell powder in water decreased from both the sites.

Table 1 and graph 1 shows that the mean fluoride concentration of the water samples post-treatment from site 1, it was 0.76 (SD 0.03) and for site 2, it was 0.91 (SD 0.04). Wilcoxon signed rank test showed the difference in the fluoride concentration of samples between pretest and posttest to be statistically significant at a p value <0.001. Approximately 50% reduction was seen in fluoride concentration from both the sites.

Discussion:-
The present study was an experimental trial to assess the efficacy of tamarind fruit shell in reducing fluoride concentration in drinking water by dip-in-technique. The result of the study showed that tamarind fruit shell can effectively remove fluoride from water. The study samples were collected from two different places of Bareilly district, U. P. The baseline fluoride concentration was 1.83ppm and 1.58ppm and post-treatment reduced to 0.7ppm and 0.76ppm respectively.

As India comes under fluoride belt with 19 states out of which 15 states are declared of having endemic fluorosis (Arappa N. et al, 2013), so it is of paramount importance to identify materials with high rate of removal of fluoride, economically, socially and technically feasible for applications in rural communities of India. Tamarind fruit cover is brittle, easily cracked shell of ripe tamarind pod that separates out from the fruit. This is available in plenty as a waste byproduct of tamarind pulp industry. Tamarindus indica fruit shells (TIFSs) are naturally calcium rich compounds. The fluoride scavenging ability of TIFS carbons is due to naturally dispersed calcium compounds. X-ray diffraction (XRD) shows that TIFS carbon contained a mixture of calcium oxalate and calcium carbonate both of which can remove fluoride from drinking water (Sivasankar V., Rajkumar S., Murugesh S & Darchen A., 2012).

V. Ramanjaneyulu et al (2013) conducted a study to develop adsorbents from leaves of Ficus religiosa (Pipal) and Tamarindus Indica (Tamarind) fruit shell to remove fluoride from drinking water. The results showed that tamarind fruit shell exhibited highest fluoride removal efficiency about 85% at pH 2.7 Another study was conducted by Ne- dunuri Phani Kumar et al (2012) for defluoridation of water using Tamarind fruit cover (TNFC) as a natural adsorbent in virgin and acid treated forms. The results showed that the tamarind fruit cover in its natural and acid treat-

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**TABLE 1: Pre-test and post-test comparisons of F levels in water samples**

<table>
<thead>
<tr>
<th>Variable</th>
<th>N</th>
<th>Mean</th>
<th>Std. Deviation</th>
<th>z value</th>
<th>p value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Site 1 – Baseline</td>
<td>50</td>
<td>1.53</td>
<td>0.00</td>
<td>6.165</td>
<td>&lt;0.001(s)</td>
</tr>
<tr>
<td>Site 1 – Post F level</td>
<td>50</td>
<td>0.76</td>
<td>0.03</td>
<td>6.165</td>
<td>&lt;0.001(s)</td>
</tr>
<tr>
<td>Site 2 – Baseline</td>
<td>50</td>
<td>1.83</td>
<td>0.00</td>
<td>6.160</td>
<td>&lt;0.001(s)</td>
</tr>
<tr>
<td>Site 2 – Post F level</td>
<td>50</td>
<td>0.91</td>
<td>0.04</td>
<td>6.160</td>
<td>&lt;0.001(s)</td>
</tr>
</tbody>
</table>

Wilcoxon signed rank test. P < 0.05(s)

**GRAPH 1: Pre-test and post-test comparisons of F levels in water samples**

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ed forms could be used as a potential biosorbing agent for the removal of fluoride ions from an aqueous media. Upto 80% uptake of fluoride ions occurred at pH 6.0. Our study showed only 50% reduction of fluoride content in the treated water which is much lesser than the studies reported. This might be because in our study we did not do any pre-treatment to the adsorbent with acid or alkali. But the technique employed in our study is not only innovative than these two previous studies, it can be used in day-to-day life and benefit a large section of the society especially those who are poor and under privileged.

E. Subramanian & R. Dhana Ramalakshmi (2010) and D. Santhi (2007) have also used tamarind seed and tamarind bark and leaves respectively for defluoridation of drinking water. These parts of the tamarind tree have also shown significant reduction in the amount of fluoride in drinking water. These studies prove that the other parts of the tamarind tree can also be used as defluoridating agents.

The tamarind shell powder imparted slight brown colour to the drinking water that had to be decolourized by addition of H2O2 drop by drop. But by adding tamarind fruit shell powder and H2O2, the taste of the treated water was not affected. The present study was not without limitation as the contact time of the tamarind fruit shell was too long. Further research is needed to find out the efficacy of tamarind fruit shell with respect to contact time with affected fluoridated water and with high fluoride concentration in water.

Conclusion:-

The study results showed that the tamarind fruit shell powder can effectively remove fluoride from drinking water. The tamarind fruit cover shell is also easily available naturally and is a waste product of many fruit pulp industries so it is quite cheap also. This can be the most efficient and cheap material to use as defluoridating agent. The technique developed and used in the present study is quite simple, cheap and can be used in remote villages having deprived people who have no access to pre-treatment of adsorbent.

**REFERENCE**

1. Arlappa N, Qureshi A. & Srinivas R (2013). Fluorosis in India: An overview. International Journal of Research and Development of Health, 1(2), 1-6. Received from http://www.ijrdh.com/files/11.Fluorosis.pdf | 2. D. Santhi (2007). Defluoridation of drinking water by tamarind bark and leaves respectively for defluoridation of drinking water. These parts of the tamarind tree have also shown significant reduction in the amount of fluoride in drinking water. These studies prove that the other parts of the tamarind tree can also be used as defluoridating agents.

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