Elemental Analysis of Local Chewing Sticks in Bangladesh

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Abstract

Proton Induced Gamma Emission Technique (PIGE) was employed for the determination of trace elemental concentrations in some local chewing stick plants of pharmacological importance. The familiar local name of chewing stick plants studied are Nishinda, Neem, Bely-asra, Bhat, Joytun, Kaminee, Akondo, Khejur, Bohera, Moth-bhringraj, Batul, Olut-kumbal, Sheora and Motkila. Proton beam from the 3 MeV Van de Graaff Accelerator at the AECD was used for irradiating the samples. Proton beam energy was 2.5 MeV and the beam current was 20 nA. As F has important dental carries inhibiting properties, the measurement of its concentration in the common chewing plants is of special interest. The PIGE reaction ^{19}F (p,p' γ)^{19}F was used for the determination of the concentrations. The certified values of concentrations in NBS SRM 1515 Apple leaves, NBS SRM 1573, Tomato leaves, NBS SRM 1573, Spinach and sediment standard AGV-1 were used as standard for the measurement of concentrations of ^{23}Na, ^{24}Mg, and ^{25}Mg. The natural abundances of Mg are: ^{24}Mg-78.99%, ^{25}Mg-10%, and ^{26}Mg-11.01%. Using the natural isotopic abundance, the concentrations of Mg isotopes have been calculated. The concentration of ^{19}F was calculated using the calibration curve of cellulose for 197 KeV energy peak. Both leaves and trunks of the chewing stick plants were analyzed. It has been observed that among all the leaf samples investigated, leaves of Neem, a common chewing stick plant, contain the highest amount of F, which is about 222 ppm. And among all the trunk samples, the highest amount of F has been observed in Bely-asra plant, which is about 122 ppm. Finally, the significance of the measured results are discussed in the light of their impact on the health care.

Key Words: PIGE, elemental analysis, chewing sticks.

Introduction

Bangladesh has varieties of plants, many of which are used for therapeutic purpose against different diseases and ailment. Most of the rural people and even some urban people in Bangladesh use varieties of chewing sticks for cleaning their teeth instead of toothpaste and brush. It is believed that these sticks are efficient, effective, and reliable for cleaning teeth and to maintain their health. It has been observed that, the teeth of the users are usually very strong, clean, fresh and devoid of germs and carries. Hardly any decayed, missed, filled or perforated teeth are found among the users of chewing sticks. Different parts such as leaves, roots, etc. of some familiar plants, in different forms such as paste, powder, etc. are used treating for teeth ailment as painkiller. An elemental analysis of these chewing sticks can provide us scientific information so that we can be certain of their efficacy.

It is known that Fluorides prevent the dental caries even though the mechanism in doing so is quite complicated and not well understood [1]. Fluorides are therefore usually introduced into modern toothpaste for therapeutic purposes. The objective of the present experiment is to estimate the concentrations of some specific trace elements ($Z<20$) especially fluorine, present in some local chewing sticks, using PIGE techniques. We are also interested in estimating the concentrations of these trace elements in those plants or parts of those plants that are commonly considered useful and efficient for various therapeutic (dental) purposes. Sometimes people use leaves or roots, in various forms (e.g. in the form of paste, powder, etc.) of some specific plants, as painkiller for tooth ailment. Therefore, investigation for fluorine in these chewing sticks is of special interest. However, excess intake of fluorine

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results in toxic symptoms described as fluorosis, which causes many diseases like arthritis, lameness, pain on movement, cancer etc[2-6].

Olabanji et al. have reported the PIGE measurements of elemental concentrations of the major, minor and trace elements in chewing sticks widely used in Nigeria. They have found in them light elements; F, Na, Mg, Al, P and Cl along with such heavy elements as Ca, Pb, transition elements and some rare earth elements. Of particular interest is the presence of F in all the samples they analyzed, and the presence of relatively high concentration of Ca, P, Na, Mg, Al and P in some. The therapeutic, preventive, and protective properties of the chewing sticks may be due to the combination of all these elements [7].

Theory

The concentration of an element in a sample can be obtained from the PIGE yield using the known yield from a standard. There are many multi-elemental standards available for the analysis of environmental, biological, medical and geological samples. Synthetic standard samples can also be used for materials' analysis. The concentration of an element or its isotope C_s can be calculated from its gamma ray yield Y_s at a particular energy using the yield Y_{st} of the standard from the formula

$$C_s = C_{st} \frac{S_s Y_s}{S_{st} Y_{st}}$$

The stopping powers S_s and S_{st} for the sample and the standard respectively can be obtained from the Stopping and Ranges software package SRIM: version 1997 developed by Ziegler and Biersack [8] or else can be obtained from the literature [9, 10]. The actual yields are obtained from the experimental yields using the relation:

$$Y = \frac{NQh^2\varepsilon}{2k2m_p E_p} g\Gamma_r$$

where ε is the efficiency of the detector at a particular energy, k is the loss of energy, m_P is the mass of the proton, and E_p is the proton energy in the lab system.

Methods

Sample preparation: The chewing stick samples were collected from different localities of Bangladesh and labeled. The samples were dried in an oven at a temperature of 130° C for about 10 hours. There after the samples were allowed to cool to room temperature in a dessicator and their weights were taken. The dried weighted samples were then grinded in a grinder and made into pellets with a pellet maker. These pellets were used as the targets for irradiation with proton beam.

Instrumentation: The experimental set up for the PIGE analytical technique comprises of a proton beam of 2.5 MeV energy from the horizontal type 3 MeV Van de Graaff accelerator, a target chamber with a sample holders and a gamma-ray detection system. The γ-ray detection and processing system consisted of a Princeton γ-tech HPGe detector having a resolution of 1.75 keV at 1332 keV energy, a preamplifier, a main amplifier, a Multi-channel Analyzer (MCA) in the pulse height analysis (PHA) mode and an IBM compatible 486 computer. A schematic diagram of the data acquisition setup used in the PIGE analysis is shown in Figure 1.

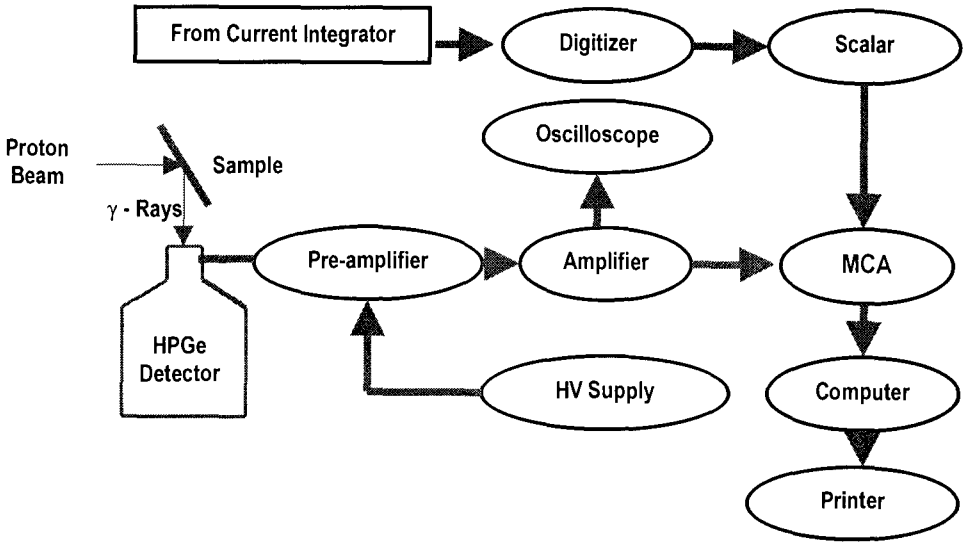

Figure 1 Schematic diagram of the data acquisition setup.

The proton beam current was maintained at 20 nA and each target was irradiated for a fixed charge of 80 μC. The count rate was kept below 2000 cps to avoid the occurrence of sum peaks in the spectrum.

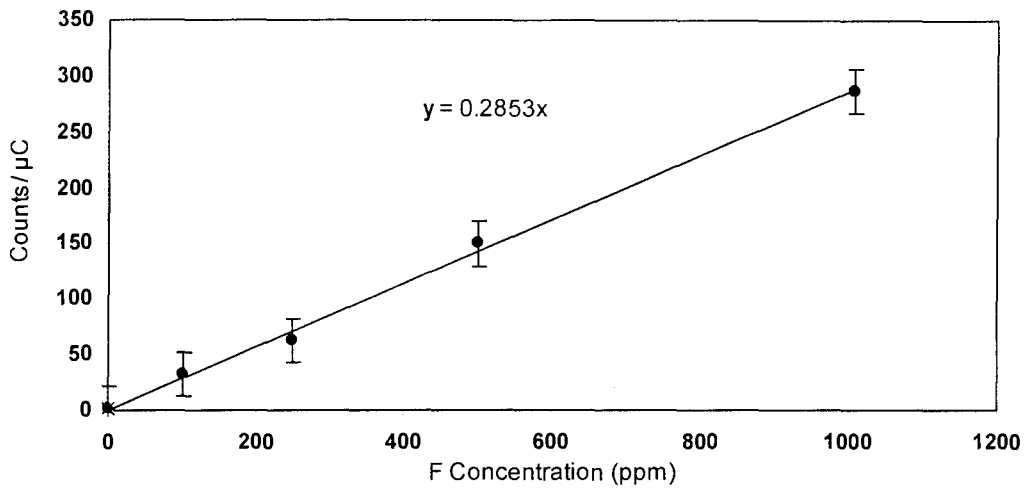

Figure 2 Calibration curve for F measurement.

Concentration calibration for F: In order to obtain the fluorine concentration calibration curve (Figure-2), Analar grade NaF in the concentration ranging from 10 to 1000 mg/L in a $CaCO_3$ matrix was used. The nuclear reaction $^{19}F(p,p'\gamma)^{19}F$ is used to construct the calibration curve.

NaF standards were homogeneously dispersed in 100 mg of $CaCO_3$ with methanol, and the resulting matrices were dried under an infrared lamp. Pellets were made with (taking) 50mg of dried samples. For testing homogeneity three 50mg pellets were irradiated and the results obtained were found to be reproducible within ±5% and the calibration curve is assumed to be correct within this limit[11].

Data analysis: The analytical information about the presence and the concentrations of the elements in a sample are contained in the peaks of their characteristic γ-ray spectra. A typical spectrum of the external PIGE from chewing stick target at the energy 2.9 MeV is shown in Figure 3. The contributions to the background came from the natural radiation, beam induced radiation such as the gamma-ray from the elements present in the experimental setup and the target chamber. The trapping of γ-rays in the detector and charge collection losses contributed to the low energy tailing of the peaks. A computer code obtained from APTEC was used for unfolding the γ-ray spectra.

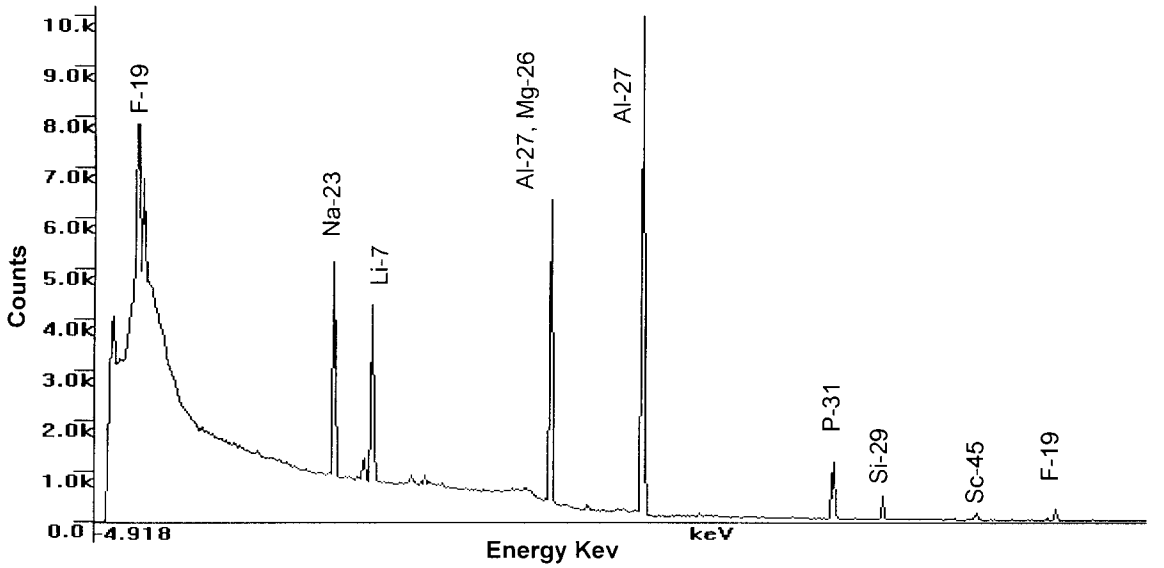

Figure 3 A typical spectrum of the external PIGE from chewing stick target at the energy 2.9 MeV.

Calibration of the HPGe Detector: For energy calibration sources such as ^{60}Co, ^{137}Cs, ^{22}Na were used and the energy resolution (FWHM) of the detector was found to be 1.75 keV at 1332 KeV.

Measurement of Efficiency of the HPGe detector: The efficiency of a detector is a function of the active volume and shape (geometry) of the detector crystal, the source-detector geometry, and the interactions of the γ-ray with the materials of the detector[12]. The relative efficiencies curve for the detector in the energy range of 186 - 2448 keV was obtained with a ^{226}Ra source. The intensities were normalized at the 609.23 keV line, which is the most intense line in the ^{226}Ra spectrum. The intensities of the gamma lines were taken from literature [13] and the relative efficiencies were obtained by dividing the normalized quantities by these intensities. The minimum detection limit, which is a measure of the sensitivity, was also measured.

Results & Discussion

The dental caries-inhibiting properties of some commonly used chewing sticks in Bangladesh have prompted us to undertake the present research in order to find their constituents using PIGE technique. Dental caries is a pathogenic process of microbial etiology that results into localized destruction of tooth tissues. It is a multifactorial disease. Though the mechanism of the clinical effect of fluoride is still debated, the fluoride has been used widely in dentistry for

many years for the caries prevention. Fluoride therapy is used to increase the resistance of teeth. High concentration (3761 ppm to 6903 ppm) of Mg was detected in all the chewing stick samples. The specific functions of Na with regards to improved qualities of human teeth are not yet known well. It has been found that among all the leaf samples, leaves of Neem contain the highest amount of ^{19}F about 222 ppm. Among all the trunk samples, the highest amount of ^{19}F has been observed in Bely-asra plant which is 122 ppm. Since 1,000 ppm. of fluoride is introduced to the most of the available toothpastes[14], it is therefore possible that the naturally fluoridated chewing stick plants may play the same role as the expensive modern toothpastes.

Local and scientific names of the chewing stick samples along with the locations of the samples collected are given in Table 1. The results of PIGE measurements for the concentrations of three trace elements ^{23}Na, Mg, and ^{19}F in local chewing sticks are presented in Table 2. The PIGE technique is a multi-elemental analytical technique, and therefore, a number of strong gamma ray lines at different energies in the PIGE spectrum of each sample were observed. We were however interested in 110, 197, 440, 585 and 1369 keV lines only.

Table 1
Local and scientific names and the parts of the plants used for the present study

Local Name	Scientific Name	Parts of the plant analyzed	Places of Collection
Nishinda	Vitex negundo L.	Leaves, Trunk	Savar
Neem	Atzadiracha indica A. Juss	Leaves, Trunk	Savar
Bely-asra	Achyranths aspera	Leaves, Trunk	J.U. campus
Bhat	Clerodendrum viscosum	Leaves, Trunk	J.U. campus
Joytun	Sesbania sesban (L.) Merr.	Trunk	Market, Dhaka
Kaminee	Murraya paniculata (L.) Jacq.	Leaves, Trunk	Savar
Akondo	Calotropis procera	Leaves, Trunk	Savar
Khejur	Phoenix sylvestris	Trunk	J.U.campus
Bohera	Terminalia belerica Roxb.	Leaves, Trunk	Shirajgonj
Moth-bhringraj	Wedelia chinensis (osb.) Merr.	Leaves, Trunk	Shirajgonj
Batul	Sapium indicum Willd.	Trunk	Shirajgonj
Olut-kumbal	Abroma angusta L.	Root	Savar
Sheora	Streblus asper Lour.	Trunk	Shirajgonj
Motkila	Glycosmis arborea Dc.	Leaves	Savar

From the Table 2 it can be seen that magnesium is present in all the samples investigated in the present experiments and its concentration is found to be relatively high. The concentration of sodium is lower than that of the magnesium. And the concentration of F, on the average, has the lowest value among the three elements detected and in some samples it has not been observed at all. In most cases, both leaves and sticks were analyzed experimentally as we were interested to find which of the two contains more F. It has been found that in some plants either leaves or trunks contain the desired element fluorine, and in some cases both the trunks and leaves found to contain F.

Fluorine in dentifrices

An optimum level of fluoride in the water supply provides significant protection against dental caries. The daily use of mouthwashes containing fluoride is helpful in the control of caries. It is noticed that 0.5 % sodium fluoride, 0.1 % stannous fluoride and 0.4 % stannous

Table-2 : Concentrations of various elements found in the chewing stick samples studied.

Concentration of various elements found in the chewing stick samples expressed in ppm

Name of the element	Nishinda				Bely-asra				Neem			
	Trunk		Leaves		Trunk		Leaves		Trunk		Leaves	
	Conc.	% error	Conc.	% error	Conc.	% error	Conc.	% error	Conc.	% error	Conc.	% error
Na	208	2	31	9	298	1	23	12	17	12	23	14
^{24}Mg	6332	3	3135	3	3834	3	3499	2	3496	3	3454	3
^{25}Mg	272	26	298	32	581	27	483	27	339	23	368	21
^{26}Mg	299	---	328	---	639	---	531	---	373	---	405	---
Mg (total)	6903	---	3761	---	5054	---	4513	---	4208	---	4227	---
F	---	---	147	15	123	20	---	---	---	---	223	12

Name of the element	Bhat				Kaminee				Akondo			
	Trunk		Leaves		Trunk		Leaves		Trunk			
	Conc.	% error	Conc.	% error	Conc.	% error	Conc.	% error	Conc.	% error	Conc.	% error
Na	54	5	31	10	34	11	10	21	82	3	856	1
^{24}Mg	4593	2	4189	2	5285	3	3314	2	4426	3	5755	3
^{25}Mg	588	28	667	23	405	54	552	30	383	32	365	37
^{26}Mg	647	---	744	---	446	---	608	---	422	---	401	---
Mg (total)	5828	---	5610	---	6136	---	4474	---	5231	---	6521	---
F	78	49	---	---	---	---	---	---	---	---	---	---

Table 2 continuation.

Name of the element	Bohera				Moth-bhringraj				Motkila			
	Trunk		Leaves		Trunk		Leaves		Trunk		Leaves	
	Conc.	% error	Conc.	% error	Conc.	% error	Conc.	% error	Conc.	% error	Conc.	% error
Na	34	7	33	9	124	2	---	---	36	7	16	15
^{24}Mg	2909	1	6165	---	2995	3	2638	1	4905	3	3345	3
^{25}Mg	317	14	602	---	119	13	844	15	513	34	480	14
^{26}Mg	348	---	663	---	130	---	928	---	563	---	527	---
Mg (total)	3574	---	7430	---	3244	---	4410	---	5981	---	4352	----
F	---	---	109	31	78	29	50	50	---	---	110	25

Name of the element	Khejur		Batul		Sheora		Joytun		Olut-kumbal	
	Trunk		Trunk		Trunk		Trunk		Root	
	Conc.	% error	Conc.	% error	Conc.	% error	Conc.	% error	Conc.	% error
Na	347	1	291	7	46	5	46	---	291	1
^{24}Mg	3145	2	3500	2	2995	---	2995	0.5	3500	2
^{25}Mg	572	24	381	18	118	---	118	---	381	14
^{26}Mg	629	---	410	---	130	---	130	---	410	---
Mg (total)	4346	---	4301	---	3243	---	3243	---	4301	---
F	58	65	---	---	---	---	---	---	---	---

fluoride gels are internationally recommended for use [15]. It is thought that enhanced protection occurs if the teeth are exposed to fluoride shortly after eruption of milk teeth because of the enamel surface remains much reactive in this period and absorbs additional fluoride rapidly. Now a days most of the available dentifrices are fluoridated. 0.76 % of Mono-Fluoro-Phosphate (MFP) or 1,000 ppm of fluoride is the most commonly used therapeutic ingredient in commercial toothpastes [16]. Besides, stannous fluoride, sodium fluoride, amine fluoride etc. are also clinically effective fluoride abrasives system in dentifrices. It has been observed that only about 40% of the chewing stick samples contain ^{19}F. The samples containing fluorine along with their concentration are presented in Table 3. It is found that the most commonly used chewing stick Bely-asra contains the highest amount of F in its trunk, whereas its leaves contain no F at all. The other popular chewing stick plant Neem contains highest amount of F in its leaves, but its trunk contains no F.

Table 3

Local Name	Scientific Name	F Contained Parts of the sample	Conc. of F in ppm.	Error %
Nishinda	*Vitex negundo L*	Leaves	147	15
Neem	*Atzadirachta indica A. Juss*	Leaves	222	12
Bely-asra	*Achyranthes aspera*	Trunk	122	20
Bhat	*Clerodendrum viscosum*	Trunk	78	49
Khejur	*Phoenix sylvestris*	Trunk	57	65
Bohera	*Terminalia belerica Roxb*	Leaves	109	31
Moth-bhringraj	*Wedelia chinensis (osb.) Merr*	Leaves	50	55
		Trunk	78	29
Motkila	*Glycosmis arborea Dc.*	Leaves	110	24

Mg in dentifrices

Mg is used in dentifrices as an abrasive. Abrasives give the mechanical aids to the paste and help to remove the stains from the tooth surface. MgO and $MgCO_3$ are the important abrasives used in dentifrices. These compounds of Mg are also incorporated in some dentifrices to neutralize the acid product on the surface of the tooth, thus inhibiting the decay. Magnesium is found in all the samples investigated with relatively high concentration in the range of 3243 ppm to 7430 ppm.

Na in dentifrices

The main use of Na in dentifrices is a foaming agent, which flushes and cleanses the cavity by removing the food debris and bacterial growth. The compounds of Na used in this purpose are: Sodium-lauryl-sulphate, Sodium N-lauryl sarcocinate. Na-benzoid is a nontoxic antiseptic and is used to inhibit bacterial proliferation in the preparation. Sodium-Mono-Fluoro-Phosphate (SMFP), another compound of Na which inhibits caries possibility by 17 % to 34 %. Joytun is the most popular chewing stick, especially to the Muslim devotees, because it's use is mentioned in the Quran. It contains no F, however it contains as much as 50 times larger Na than found in other samples.

Sources of uncertainties:

In table 2 the errors involved in the measurements are shown. Large fluctuations in errors are observed. There may several reasons for the fluctuation in the errors.

a) All experiments were done at a fixed proton energy of 2.5 MeV, which probably was not high enough for exciting all elements in a thick target sample. Proton beam of

higher energy (>2.5 MeV) might be required to excite all elements and isotopes present in a sample, and also when the intensity of the emitted gamma lime is weak then the experiment have to be run for a longer time to obtain statistically significant number of counts in the gamma peaks.

b) Proper selection of peak areas can also reduce the errors because the sharper is the peaks the better is the selection. The errors are relatively high in the case of fluorine measurements, because in most cases the γ- ray peaks of energies 110 & 197 keV lied near the Compton edges and were not sufficiently sharp. Proton beam of higher energy or longer time for an experiment might have been able to overcome this difficulty.

c) Errors may also arise due to fluctuations in high voltage power supply, inability of the preamplifier to extract properly the weak current signal from the detector, inadequate analyzing power of MCA for which an energy peak may be shifted from its proper position and become broaden. [17-19]

Conclusion

This study provides some authentic information about the data of chewing sticks in dental care. Previously there were no scientific explanations about their usefulness, nonetheless people use these sticks, partly for the sake of tradition and partly as they can't afford to buy the expensive modern toothpaste. The results of the present work may influence people including the urban people to use these chewing sticks for dental care. If it can be shown that the commonly used popular chewing sticks contain standard amount of desired trace elements, that are efficient and effective for dental health and care, it would then be possible to raise funds for large scale cultivation of these trees, and produce and market chewing sticks commercially. For these to happen further studies are needed. The results of the present experiment can still be used to satisfy the baseline data for dental health care.

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