DETERMINATION OF FLUORINE IN TEA, BONES AND ROCKS THROUGH THE $^{19}F(p,\alpha\gamma)^{16}O$ REACTION USING A CYCLOTRON

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Abstract

The amount of fluorine present in various types of tea leaves, basaltic volcanic rocks and rabbit bones has been determined by observing the 6.13 MeV gammas from the reaction $^{19}F(p,\alpha\gamma)^{16}O$ using thick targets and 5 MeV protons from the Melbourne University Cyclotron. The fluorine contents in tea leaves from Darjeeling, Ceylon and Queensland have been determined to be 135 ± 5 p.p.m., 75 ± 5 p.p.m. and 60 ± 7 p.p.m. respectively. The volcanic rock sample TB4 is found to contain 295 ± 20 p.p.m. of fluorine. The bones of the fully grown up rabbits who were fed with fluoridated water (120 p.p.m.) for a period of 3 months show as much as 950 ± 50 p.p.m. of fluorine.

1. Introduction

Various chemical methods for fluorine determination in different samples are not only destructive but may also suffer from unreliability of the results due to a variety of reasons like the loss or gain of fluorine in the sample during chemical processing, incomplete extraction of fluorine from solid samples into solution, etc. The x-ray fluorescence method lacks sensitivity (limit of detection 0.2%)[1]) and is also difficult due to rather low energies of fluorescent x-rays from fluorine. Activation analysis with fast neutrons and bremsstrahlung may lead to the tedious task of having to separate exponentials when other positron emitting isotopes are also produced along with ^{18}F. On the other hand, fluorine determination through the $^{19}F(p,\alpha\gamma)^{16}O$ reaction is not only convenient and simple but very sensitive too owing to the large reaction cross section and clearly identifiable prompt gammas (mainly 6.13 MeV). This technique can be non-destructive and is applicable to bulk samples as well for fluorine concentration and depth profile measurements, even using low energy protons from electrostatic accelerators[2,3]). This paper describes the application of cyclotron beams for studying fluorine content in tea leaves, rocks and bones using this reaction.

A quantitative estimation of fluorine in different varieties of tea leaves may provide significant information which may be a help in the water fluoridation controversy. It has been believed for a long time that tea leaves contain some fluorine but the quantitative data is missing.

An accurate determination of fluorine content in the bones of animals who have been given fluoridated water is needed in order to study the best possible way of administering fluoride to human beings as well as to look into the effects of water fluoridation on bone pathology.

The development of an accurate and reliable method for fluorine determination in rocks has a number of geological applications like the search for economic nickel sulphide ores, etc.

2. Principle of the Method

The fluorine determination is based on the principle that when thick targets of an unknown sample and a known standard are bombarded with identical beams the fluorine concentration in the two is related by the following equation[4])

$$\frac{N}{N_s} = \frac{c}{c_s} \cdot \frac{Q}{Q_s} \cdot \frac{R}{R_s} \qquad (1)$$

where N is the number of 6.13 MeV gammas observed from the sample; Q is the charge collected; R is the range of incident protons in the sample and c the concentration of fluorine in p.p.m. in the unknown sample. The same notations with subscript s refer to the known standard for identical bombarding and detection conditions. The condition for the equation to be valid is that the matrices of both the unknown sample and the standard are the same or similar and that the fluorine is homogeneously distributed in the two. The unknown concentration c can then be obtained from direct experimental measurements (N,N_S) and from tabulated values of R and R_S [5]). The particle range in a compound or mixture of elements 1, 2, 3, ... is approximately given by

$$\frac{1}{R} = \frac{w_1}{R_1} + \frac{w_2}{R_2} + \frac{w_3}{R_3} + \ldots\ldots \qquad (2)$$

where w_1, w_2, w_3, ... are the weight fractions (gram per gram) and R_1, R_2, R_3, ... are the corresponding ranges (gram per cm^2) for each of the elements[6]).

3. Experimental Set-up and Method

In order to prepare thick usable targets the samples had to be powdered. The tea leaves were ground in ultra clean steel grinders while the rock samples were first crushed with steel grinders and then converted into fine powder using grindstones. The rabbit bones were ashed before being powdered. The powdered tea and bone samples were placed in 1 mm deep and 15 mm diameter groove of aluminium target holders, pressed into pellets with a hydraulic press and covered with a thin aluminium foil to avoid any possible spilling out of the target under bombardment. The powdered rock was first covered with a 20 μm thin aluminium before pressing. Ultra pure LiF, NaF and SrF_2 were used to make fluorine standards. These were converted into target pellets too by applying similar pressure conditions. These pressed targets were thick enough to stop 5 MeV protons completely. Each aluminium target holder contained an unknown sample and a known standard.

The targets were placed at the centre of an insulated chamber made out of a 10 cm diameter tube and were cooled with liquid nitrogen. The proton beam from the cyclotron was collimated by a number of carbon slits and had an area of 2mm x 2mm on the target. Beam currents from 0.2 to 0.5 μA were

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used for unknown target samples while much lower beams were required for the standards. The entire chamber assembly was used for measuring the collected charge.

The prompt gammas were detected with a 70 cc Ge(Li) detector placed at a distance of 30 cm from the target. The detector was shielded with paraffin wax to avoid any neutron damage and, therefore, could not be placed any closer to the target. The pulses were displayed on a PDP-11 multichannel analyser system after passing through conventional electronics. Using a biased amplifier only the region around three fluorine peaks (6.13, 6.92 and 7.12 MeV) was fed into and displayed on the analyser. The current digitizer, the ADC's and the computer-analyser system were operated in such a way that the pulses collected in the analyser corresponded to the live charge only. Therefore, no dead time correction was needed. In fact, even at the highest count rate the dead time was never more than a few percent.

4. Results and Discussion

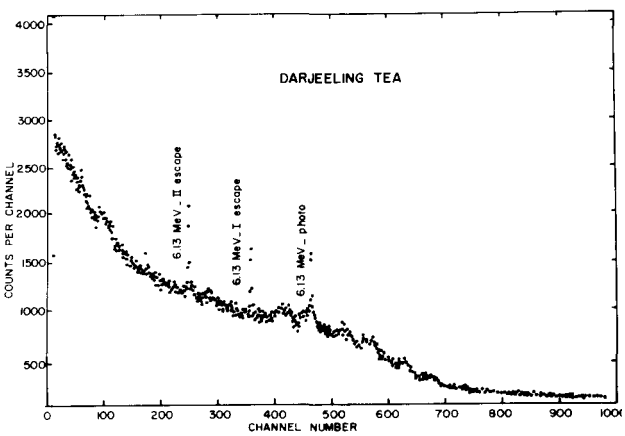

Fig. 1. Gamma spectrum in the region around 6 and 7 MeV from a thick tea target under bombardment with 5 MeV protons.

A typical gamma spectrum obtained from a tea target is shown in fig. 1. All the three peaks (the photo peak, and I and II escape peaks) corresponding to 6.13 MeV gammas are clearly resolved while those due to the other two gammas were Doppler-broadened. Though pulses due to all three gamma energies could be used for fluorine determination as the likelihood of gammas from any other element existing in this region was rather small, we confined ourselves only to the peaks for the 6.13 MeV gamma for calculations.

Using a tea matrix consisting of 6% H, 47% C, 4% N and 43% O [7]) the fluorine content in tea leaves from Darjeeling, Ceylon and Queensland are calculated to be 135 ± 5 p.p.m., 75 ± 5 p.p.m. and 60 ± 7 p.p.m. respectively. The rabbit bones (Ca_{10} $(Po4)_6$ $(OH)_2$) are found to contain 950 ± 50 p.p.m. of fluorine while the rock sample TB4 (major constituents: 48.7% O, 3.1% Na, 8.5% Al, 25.2% Si, 3.6% Ca, 7.1% Fe) has 295 ± 20 p.p.m. of fluorine. These values were reproducible and lie within experimental errors when different standards are used for calculations.

The amount of fluorine in tea leaves is of special significance. About 2.5 - 3.5 grams of tea (level or heaped teaspoon) is used for an average tea cup of about 180 cc volume. Around 80% of the fluorine gets into the tea-liquid from the leaves. This means that a cup of tea contains anywhere from about 120 - 380 µg of fluorine depending upon the type and amount of tea leaves used. A cup of fluoridated water (concentration 1 p.p.m.), on the other hand, contains about 180 µg of fluorine. People have been drinking tea for ages. Therefore our finding indicates that a cup of fluoridated water is no more poisonous than a cup of tea or it may also mean that persons in need of fluorine should be given tea rather than fluoridated water thus avoiding the need for fluoridating the water supply for the entire population.

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