Setting up of PIGE facility at FOTIA, BARC and its application to Borosilicate Glass Samples for Determination of Fluorine by an Internal Standard Method

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Introduction

Particle Induced Gamma-ray Emission (PIGE) [1] is an important nuclear analytical technique used for determination of light elements like Li, Be, B, F, Na, Mg, Al and Si in diverse matrices. In this technique, beam of charged particles fall on sample surface and nuclear reactions like (p, p'γ), (p, γ) and (p, $\alpha\gamma$). Gamma rays emitted in this process are the signature of a particular nucleus and the numbers of counts are proportional to the concentration of the element in the sample. PIGE is advantageous over PIXE, since it uses gamma-rays, thus experiences less matrix effect. This property helps in applying PIGE for thick samples. It is non-destructive, simultaneous capable and high sensitive some of the light elements like Li, B and F. Determination of F in complex matrix like borosilicate glass is difficult by many chemical methods, involving dissolution and quantitative separation, and also by NAA since it contains B and Na. Fluorine is determined by on-line measurement of prompt γ rays of 110 and 197 keV from $^{19}F(p, p'\gamma)^{19}F$ as well as 6.1 MeV from $^{19}F(p, \alpha\gamma)^{16}O$ by high resolution γ-ray spectrometry [2].

In beam experiments current normalization is an important aspect. This can be done in two ways: either by taking count rate normalized to the backscattered protons from a thin target like gold kept before the sample or by counting them relative to one of the components (external / internal) which is not an element of interest in the sample to be investigated. The limitation of the first method is the inhomogenities of the gold foils used for samples and standards which in turn reflect in the inaccuracy of F concentration. The second method of internal standard (IS) is advantageous over the first method as it is an in-situ monitor of current variation and does not depend on the sample-to-detector geometry. The sensitivity (cps / ppm) of IS is used for the normalized count rate of gamma-ray peak of interest.

In the present work, we have standardized an internal standard particle induced gamma-ray emission (IS-PIGE) method to determine fluorine in glass matrix. The element Li, which is almost equal sensitive like F was used as an internal standard. The work was carried out using 4 MeV proton beam obtained from Folded Tandem Ion Accelerator (FOTIA), BARC, Mumbai.

Experimental

Calibration curve was obtained for standards of F containing Li as internal standard which were prepared using NaF (varying amount) and Li_2SO_4(fixed amount of 34 mg) mixed with cellulose in pellet (net weight 750 mg) form. For validating this method, synthetic standards were prepared by mixing fixed amount of base glass containing varying amount of F and fixed amount of Li_2SO_4(25 mg) with cellulose in pellet form. The sample and standard pellets were of 2 cm dia and 2 mm thick. The samples were irradiated with 4 MeV proton beam from FOTIA@BARC with beam current around 5 nA. Irradiation times were kept in the range of 30 min - 2 h depending on F concentrations. The prompt gamma-rays of 197 keV from $^{19}F(p, p'\gamma)^{19}F$ reaction were measured using a HPGe detector placed at 90^0 w.r.t. the beam. The

detector to sample distance was kept minimum (~5 cm). The energy calibration of the detector was done using a standard source of ^{152}Eu.

Results and discussion

Table 1: Details of F and Li standards

Std ID	Mass (mg)			
	NaF	$Li_2SO_4.H_2O$	Cellulose	Pellet
S-1	3.4	34	712.7	750.1
S-2	8.4	34.1	707.7	750.2
S-3	13.6	34.2	702.7	750.5
S-4	17.1	33.9	699.3	750.3
S-5	34	34.2	682.8	751
S-6	50.8	34	665.6	750.4
S-7	67.6	34.3	648	749.9

The calibration plot of CPS of 197 keV of ^{19}F normalized with sensitivity of Li is given in Fig. 1. The concentrations of F were in the range 1000 – 40,000 mg kg^{-1} and the concentration of Li (IS) was kept almost constant (0.5%). The slope of the plot was found to be 1.25 CPS/ppm of F, which was used for concentration calculations of F in five synthetic borosilicate glass samples. Control glass sample did not show any Li peak. The results of F concentrations are given in Table 2. The %deviations of F determined in synthetic samples are in the range of ±0.5-8% (Table 2). The method thus standardized was found promising and is being applied to several samples of barium borosilicate glass, to be used for vitrification of nuclear waste obtained from AHWR.

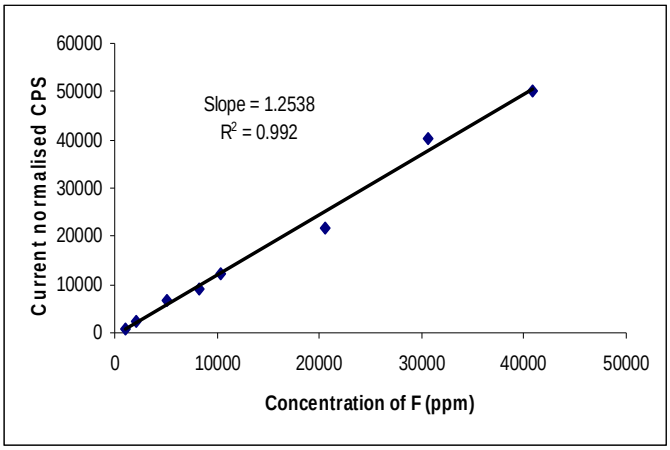

Fig 1. Calibration plot of CPS of 197 keV of F vs. concentration of F

Table 2: Validation of IS-PIGE method using synthetic borosilicate glass samples

Sample ID	F (mg kg^{-1}) obtained	F (mg kg^{-1}) expected	%Error
Syn-1	916	1000	-8.4
Syn-2	2153	2000	7.6
Syn-3	5231	5000	4.6
Syn-4	7785	8000	-2.7
Syn-5	10052	10000	0.5

References

[1] M. Volfinger, J.L. Robert, J. Radioanal. Nucl. Chem., 185 (1994) 273.
[2] HR Verma, *Atomic and Nuclear Analytical Methods* Springer-Verlag, Berlin (2007).