Non-destructive quantification of boron and ^{10}B to ^{11}B ratios in neutron absorber materials by PIGE using proton beam from FOTIA, BARC

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Non-destructive quantification of boron in its compounds, used as neutron absorbers in nuclear reactor technology, is a challenge. Two nuclear analytical techniques namely Prompt Gamma ray NAA (PGNAA) and Particle induced gamma-ray emission (PIGE) are suitable for boron determination in solid as well as ceramic samples. PIGE using proton beam is a powerful tool for non-destructive quantification of low Z elements like Li,Be, B, F, Na and Al. A PIGE method using 4 MeV proton beam from FOTIA was standardized for boron quantification in samples like B4C and borax using (p,p'γ)and/or (p,αγ) reactions of ^{10}B and ^{11}B. The method gives total B as well as ^{10}B and ^{11}B concentrations and thus keeps promise for evaluating 10B/11B ratio in solid/ceramic samples.

Introduction

Particle induced gamma-ray emission (PIGE) is an isotope specific nuclear analytical technique used for non-destructive determination of mainly light elements in various matrices. In principle all elements are amenable to PIGE, however the cross section decreases with Z. For practical purposes, PIGE using p and d beams (in the approx. energy range of 2-6 MeV) is capable of determining light elements (Z<17) like Li, Be, B, C, N, O, F, Na and Al [1, 2]. PIGE is often used as a complementary technique to PIXE and NAA. It utilizes inelastic scattering or nuclear reactions after the bombardment of a sample with an energetic ion beam and measurement of prompt gamma rays using high resolution HPGe detector.

Amongst the low Z elements, boron is one of the most important elements, which finds applications in many fields including nuclear technology. Since its neutron absorption (n,α) cross section is very high (764 barn for B and 3837 barn for ^{10}B), it is used as nuclear reactor control rod and as neutron absorbers in several nuclear applications. For chemical/nuclear quality control it is essential to know the exact total boron contents as well as concentrations and ratios of two isotopes namely ^{10}B and ^{11}B. Since contamination and evaporation is a problem in wet chemical techniques, Prompt gamma ray neutron activation analysis boron estimation PGNAA [3] is one of the best methods for total as well as ^{10}B quantification. In the absence of PGNAA facility, PIGE using Proton beam is the best method for quantitative estimation of B and its isotopes ($^{10,11}B$). PIGE utilizes measurement of prompt gamma rays of 429 keV from ^{10}B (p, αγ) ^{7}Be and 718 keV from ^{10}B (p, p'γ) ^{10}B for 10B and total B and 2125 keV from ^{11}B (p, p'γ) ^{11}B for 11B and also total B. Thus it gives a scope to determine total B as well as its isotopic composition in samples.

In this work, a PIGE method for boron was standardized at FOTIA, BARC using 4 MeV proton beam using boric acid as the standard. Samples analyzed were boron carbide (B_4C as a ceramic sample) and borax ($Na_2B_4O_7.10H_2O$ as a salt). The existing PIGE set up at FOTIA (Fig. 1) by Radiochemistry Division, BARC was used in our work. Samples and standards were prepared in cellulose matrix in pellet form. The element F (mixed as CaF_2) was used as an insitu current normalize due to its high sensitivity. The samples (stoichiometric chemical compounds) analyzed were also used for validation of PIGE method and their isotopic composition.

Fig. 1: PIGE set up at FOTIA, BARC

Experimental

Five boron standard pellets were prepared by mixing H_3BO_3 (B amount 10000-70000 mg kg^{-1}) and constant amount of CaF_2 (~ 10 mg) in cellulose (Aldrich-Sigma) matrix. For method validation as well as application of PIGE, duplicate samples were prepared by mixing CaF_2 (~ 10 mg) and B_4C (50 mg) and $Na_2B_4O_7.24H_2O$ (200 mg) in cellulose matrix. Samples and standards, fixed in ladder, were irradiated with 4 MeV proton beam from FOTIA, BARC and 10 nA beam current. Depending upon the boron concentration in the targets, irradiation time (about 30 minutes) was decided. The prompt gamma-rays were measured using 30% HPGe at a distance of 6 cm from the sample position. A typical gamma ray spectrum of B_4C (mixed with F and Cellulose) is shown in Fig. 2, indicating both ^{10}B and ^{11}B. The experimentally determined boron sensitivity (from average of corresponding boron counts per ppm) were in good agreement (with in ±2%). The current normalized sensitivity (corresponding to the gamma ray of interest for

^{10}B and ^{11}B) was used for concentration calculation using following formula,

$$C_B \text{ (mg kg}^{-1}) = (CPS_{B,F})_N / (S_{B,F})_N$$

where $(S_{B,F})_N$ and $CPS_{B,F})_N$ are current normalized sensitivity and count rate for B. This formula helps in calculating isotopic concentrations as well as total concentration, since they are all of natural isotopic abundance.

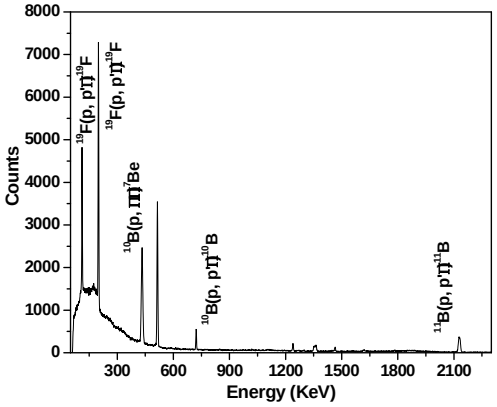

Fig. 2. Typical gamma-ray spectrum of B_4C with CaF_2

Result and Discussion

The gamma ray spectrum (Fig. 2) of B4C clearly indicates the presence of ^{10}B and ^{11}B, however the PIGE method is more sensitive for 10B due to lower energy (higher detection efficiency) and higher yield (one order) for (p,αγ) reaction. The activity ratios of gamma rays corresponding to ^{10}B and ^{11}B for boric acid, B_4C and borax are in good agreement, indicating they are of natural isotopic composition. The concentrations of boron in B_4C (76wt % w.r.to expected boron content of 78.3 wt%) and borax (11.26 wt% w.r.to expected boron content of 11.34 wt%) are found to be in good agreement. The method is being applied to other boron based compound like TiB_2, borosilicate glass, boral and hexaborides of rare earth elements and also in hydrogenous matrix.

Conclusion

PIGE method, thus standardized, is a suitable non-destructive approach for quantification of total B and its isotopic composition. It is interesting to note that enrichment of boron can easily be estimated by this method. This method can be applied to quantify boron diverse matrices, even in liquids in special cases. It is interesting to know that no gamma-ray interference was observed in boron quantification.

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