Application of external (in air) Particle Induced Gamma-ray Emission using low energy proton beam for non-destructive quantification of low Z elements in reactor materials

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Introduction

Particle Induced Gamma-ray Emission (PIGE), an accelerator-based nuclear analytical technique, is based on the detection of prompt gamma rays mainly from inelastic scattering when a target is bombarded with low energy charged particles (usually 2 to 4 MeV proton beams from tandem particle accelerator). This method has been utilized for quantification of low Z elements like Li, Be, B, F, Na, Mg, Al, Si and Ti. in different matrices ranging from biological, environmental, archaeological to reactor materials. Due to its isotope specific nature, isotopic composition determination of low Z elements like B ($^{10}B/^{11}B$ atom ratio) and Li ($^{6}Li/^{7}Li$) could also be done. We had developed and standardized in situ current normalized PIGE method (using vacuum chamber) at FOTIA, BARC using 4 MeV proton beam (~10-15 nA beam current) for non-destructive quantification of low Z elements and isotopic compositions (wherever applicable) in different boron based ceramics, reference materials etc [1,2]. Recently we had extended this system for direct analysis of non-standard geometry samples by extracting the proton beam in air using a thin Ta window. This paper describes the applications of this external PIGE set up for non-destructive quantification of low Z elements in 'as received' samples (reactor materials) and the advantages of the external PIGE method compared to the conventional PIGE set up.

Experimental

External PIGE set up at FOTIA, BARC is shown in Fig.1. This external PIGE set up facilitates easy sample manipulation i.e. handling and positioning of the objects and hence, high sample throughput with less turnaround time. Here the proton beams are extracted in air through a 25 μm Ta window. For the initial 5 MeV proton beam (in vacuum energy), the energy on the target was ~3.5 MeV after loss of energy in the window, air gap between window to sample and Mylar foil (1mil thickness) used to wrap the samples as calculated using SRIM 2013. Direct solid samples or powder samples wrapped in Mylar were irradiated with 3.5 MeV proton beam of current ~10-15 nA and prompt gamma rays were measured online using HPGe detector system. Each sample was irradiated sufficient time (~10-15 minutes) to gather sufficient statistics. Both samples and standards were irradiated under similar experimental conditions for elemental quantification. Beam current fluctuations are monitored using prompt gamma rays from Ta window material.

Fig. 1 External PIGE set up at FOTIA, BARC showing the HPGe detector and beam extraction window

Results and Discussion

This external PIGE method was successfully applied for rapid non-destructive quantification of isotopic composition of boron

in various ceramic/ refractory boron based neutron absorbers [3]. Both total boron mass fraction and isotopic composition of boron (IC, $^{10}B/^{11}B$ atom ratio) in natural and ^{10}B enriched boron carbides, di-borides of refractory metals (TiB_2, ZrB_2 etc.) and hexa-borides of rare-earths (LaB_6, CeB_6, GdB_6 etc.) were determined using external PIGE just by wrapping the powder samples in thin Mylar foils and irradiating with 3.5 MeV proton beams of ~5-10 nA current. B and Fe were quantified simultaneously in different ferro-boron alloys important for various applications including nuclear waste storage. Prompt gamma rays at 429, 718 and 2125 keV of $^{10}B(p,\alpha\gamma)^{7}Be$, $^{10}B(p,p'\gamma)^{10}B$ and $^{11}B(p,p'\gamma)^{11}B$, respectively, were measured for boron quantification and 847 keV of $^{56}Fe(p,p'\gamma)^{56}Fe$ was measured for Fe quantification. Typical PIGE spectra of B_4C and ferro-boron alloy are shown in Fig. 2.

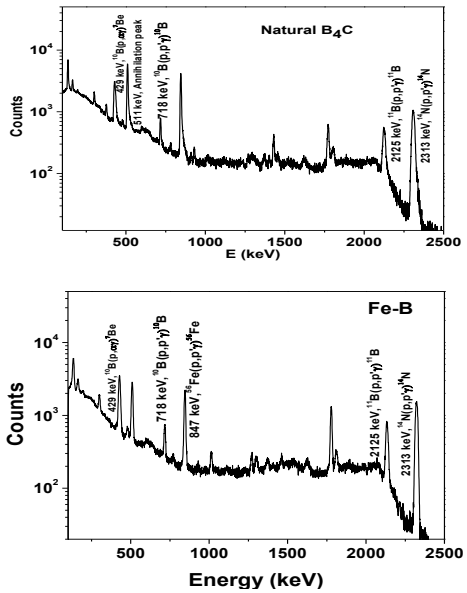

Fig. 2 External PIGE spectra of natural B_4C and ferro-boron alloy irradiated using 3.5 MeV proton beam (external beam)

Low Z elements like Al ($^{27}Al(p,p'\gamma)^{27}Al$, 1014 keV) and Si ($^{28}Si(p,p'\gamma)^{28}Si$, 1779 keV) in radioactive U_2Si_3 and U_2Si_3 dispersed in Al matrix (uranium matrix) samples were determined by external PIGE. Here powder samples were doubly sealed in thin Mylar films and irradiated using external PIGE facility at FOTIA. Low Z elements in uranium matrix could be determined easily due to feasibility of analyzing "as received" samples in external PIGE, as no pellet preparation was required. Beam current normalization was carried out using either 135 keV from $^{181}Ta(p,p'\gamma)^{181}Ta$ (tantalum from beam exit window) or 2313 keV from $^{14}N(p,p'\gamma)^{14}N$ (nitrogen from air).

Conclusions

External (in air) PIGE using proton beam (3.5 MeV energy, 10-20 nA current) from FOTIA, BARC was utilized for non-destructive quantification of low Z elements in "as received" reactor materials. Because of high thermal neutron absorption cross section of boron, boron based samples have various applications in nuclear industry as control/ shut-off rods, shielding materials, neutron poison and sensors for neutrons and nuclear material storage etc. They are difficult to be analyzed by conventional wet chemical methods. Chemical quality control of these materials is required before their uses in reactors. Utilizing external PIGE method they could be analyzed directly without any sample preparation. It is especially advantageous for quantification of low Z elements in radioactive samples as direct 'as received' samples are analyzed and there is no instrumental contamination as well as additional radioactivity generation. This method is a novel and greener approach for simultaneous rapid non-destructive quantification of low Z elements in different ceramic/refractory reactor materials.

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