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Thick target yields of proton induced gamma-ray emission from Al, Si and P

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ABSTRACT

Thick target excitation yield curves of gamma-rays from the reactions ^{27}Al(p,p′γ)^{27}Al (E_γ = 844 and 1014 keV), ^{27}Al(p,αγ)^{27}Al (E_γ = 1369 keV), ^{28}Si(p,p′γ)^{28}Si (E_γ = 1779 keV), ^{29}Si(p,p′γ)^{29}Si (E_γ = 1273 keV) and ^{31}P(p,p′γ)^{31}P (E_γ = 1266 keV) were measured by bombarding pure-element targets with protons at energies below 3 MeV. Gamma-rays were detected with a High Purity Ge detector placed at an angle of 90° with respect to the beam direction. The obtained thick target gamma-ray yields were compared with the previously published data. The overall systematic uncertainty of the thick target yield values was estimated to be better than ±9%.

1. Introduction

Particle induced γ-ray emission (PIGE) is an isotope sensitive analytical technique to quantify the concentration of elements in surface regions of solids. This technique is based on the detection of characteristic prompt gamma-rays arising from nuclear reactions produced during the bombardment of targets by a few MeV proton beam. PIGE is particularly suitable for measuring light elements as compared to heavy ones since the Coulomb barrier increases with increasing atomic number.

For the implementation of quantitative PIGE analysis without utilizing reference materials, adequate and reliable experimental cross section data are required. To meet the needs of the IBA community for PIGE measurements, a Coordinated Research Project (CRP) was initiated by the IAEA in 2011 [1]. In this CRP, besides the compilation of existing experimental cross section data, new measurements were performed; the resulting data were assessed and finally disseminated as a reference database to be used for PIGE analysis. Since at present the requirements for a standardless PIGE technique are not fully met, thick target yield data should be made available for current analytical applications. In fact at this stage, quantitative PIGE analysis of thick samples is either performed relative to proper standard samples or deduced from experimental thick target gamma-ray yields of pure-element samples. Thick target excitation yield curves of the gamma-rays can also be utilized for extracting resonance parameters [2–4] as well as benchmarking experiment in order to test the validity of the measured cross sections [5–10]. PIGE bulk analysis of light elements with a proton beam is commonly performed by prompt gamma-rays due to inelastic scattering (p,p′γ). Moreover, the narrow and isolated resonances of (p,γ) and (p,αγ) nuclear reactions are used for obtaining depth profile information [11].

Aluminum can be analyzed by the low energy gamma-rays of nuclear reactions ^{27}Al(p,p′γ)^{27}Al (E_γ = 844 and 1014 keV), ^{27}Al(p,αγ)^{27}Al (E_γ = 1369 keV) and ^{27}Al(p,γ)^{28}Si (E_γ = 1779 keV). Among them, the 844 keV and 1014 keV lines are the most applicable ones for aluminum analysis due to their high gamma-ray yields. To our knowledge, only few studies concerning the measurement of thick target gamma-ray yields for aluminum have been published in the literature for proton energies of 1.5–3.0 MeV [12]; 1.77–4.07 MeV [13]; 2.5–4.0 MeV [10] as well as at individual proton energies of 1.7, 2.4 MeV [14]; 1.7, 2.4, 3.1, 3.8, 4.2 MeV [15]; 7.0 and 9.0 MeV [16]. Silicon has two significant gamma-lines of 1779 keV from the ^{28}Si(p,p′γ)^{28}Si reaction and 1273 keV from the ^{29}Si(p,p′γ)^{29}Si reaction, which are the most appropriate for PIGE analysis. Six datasets of silicon thick target gamma-ray yields are available in the literature for the energy ranges of 3.0–4.09 MeV [13] and 2.65–4.13 MeV [10] as well as at several selected proton energies of 2.4 MeV [14]; 2.4, 3.1, 3.8 MeV [15]; 7.0, 9.0 MeV [16]; 4.0, 4.5, 55, 6.0 and 6.5 MeV [9]. Phosphorus can be analyzed by the PIGE technique employing the ^{31}P(p,p′γ)^{31}P reaction via detecting the 1266 keV gamma-ray which has the highest yield among other gamma-rays emitted during the proton beam bombardment of a phosphorus target. The reported thick target yields of the 1266 keV gamma-ray are measured for the energy ranges of

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1.75–2.56 MeV [17] and 2.2–4.08 MeV [13] as well as at some selected proton energies of 1.7, 2.4 MeV [14]; 1.7, 2.4, 3.1, 3.8, 4.2 MeV [15]; 7.0 and 9.0 MeV [16].

The aim of the present study is measurement of detailed reliable nuclear datasets of the absolute thick target gamma-ray yields at relevant angles and energies suitable for PIGE analysis of aluminum, silicon and phosphorous.

2. Experimental procedure

2.1. Experimental setup

The experimental work was carried out at the 45° right beamline of the 3MV Van de Graaff electrostatic accelerator of the Nuclear Science and Technology Research Institute (NSTRI) in Tehran. The beam energy was calculated based on the field strength of the analyzing magnet, as measured by an NMR fluxmeter. The accelerator beam energy was calibrated using the 991.88 keV resonance of the $^{27}Al(p,\gamma)^{28}Si$ reaction and the 1880.44 keV threshold energy of the $^{7}Li(p,n)^{7}Be$ reaction. The targets used for these measurements were a 60 μm-thick Al foil and a LiF pellet, respectively. After the calibration, the uncertainty of the proton beam energy was found to be about 2 keV. The beam was collimated by means of two fixed apertures, 5 mm and 3 mm in diameter, positioned at 250 cm and 87 cm from the target (center of the chamber), respectively. The beam spot size on the target was about 4 mm in diameter. The target was oriented inside the reaction chamber so that the incident beam direction made an angle of 45° with the normal to the target. The reaction chamber which is recently designed and fabricated in our lab can be used for simultaneous measurements of PIGE, RBS and PIXE. It is made of an aluminum alloy with a lining of tin (Sn) to minimize the gamma radiation background caused by backscattered particles in the aluminum wall of the reaction chamber. Our experimental setup includes a coaxial type HPGe detector, a silicon detector, an isolated target holder and a Faraday cup electrically connected to the target to measure the incident beam current. During the measurements, the vacuum in the chamber was about 5×10^{-5} mbar. The gamma-rays were detected by a P-type HPGe coaxial detector with a crystal size of 6.58 cm × 6.58 cm and an active volume of 213 cm^3. The detector was placed at a right angle with respect to the beamline direction at a distance of 51.9 mm from the target center and subtending an angle of 65°. The nominal efficiency and resolution of the detector were 50% and 1.95 keV for 1.33 MeV, respectively. To reduce the γ-ray background, the detector was protected by a 5 cm-thick cylindrical lead shield as well as by lead bricks around the apertures. Moreover, the inner wall of the cylindrical lead shield was covered with a 3 mm-thick copper (Cu) lining to reduce the X-ray production from the lead. The detector employed for the detection of the scattered protons was an ion-implanted silicon detector with 25 mm^2 active area, 300 μm thickness and 13 keV energy resolution placed at an angle of 165° relative to the incident beam direction. The solid angle of the charged particle detector determined from the geometry of the experimental setup, was found to be 1.65 ± 0.03 msr. During the experiments, the proton beam currents were adjusted within 1–40 nA depending on the beam energy and the target material to keep the counting rate of both detectors low enough so that the pile up effects were negligible and the dead time of both detectors less than 10%.

2.2. Targets

Thick targets of Al and Si were prepared from a 500-μm thick pure elemental aluminum sheet and a 300-μm thick silicon wafer, respectively; while thick targets of P were prepared by pressing red phosphorous powder into 1-mm thick pellets with a diameter of 10 mm using a hydraulic press. Moreover, a thin conducting Au film was evaporated on the surface of the Si and P targets to measure the incident beam charge on these targets.

2.3. Absolute efficiency of the HPGe detector

The absolute efficiency of the gamma-ray detector was determined using ^{133}Ba, ^{152}Eu, ^{137}Cs, ^{60}Co and ^{241}Am calibration radioactive sources which were placed into the exact position of the target. The absolute full-energy peak efficiency curve and the corresponding fitting parameters were obtained by fitting the experimental points with the function [1]:

$$\varepsilon_{abs}(E_\gamma) = a + \frac{b}{E_\gamma} + \frac{c}{E_\gamma^2} + \frac{d}{E_\gamma^3} \tag{1}$$

where E_γ is the energy of each gamma-ray line. The branching ratios of the calibration source gamma-ray lines were taken from Ref. [18]. The obtained absolute efficiencies for the gamma-rays at 844, 1014, 1369, 1273, 1779 and 1266 keV were $(11.9 \pm 0.8) \times 10^{-3}$, $(10.4 \pm 0.7) \times 10^{-3}$, $(8.3 \pm 0.6) \times 10^{-3}$, $(8.7 \pm 0.6) \times 10^{-3}$, $(6.9 \pm 0.6) \times 10^{-4}$ and $(8.8 \pm 0.6) \times 10^{-3}$, respectively.

2.4. Gamma-ray yield and proton beam charge

Yields related to the gamma-ray lines of interest and the incident beam charge values were determined from the simultaneous collection of gamma-ray spectra and backscattered proton spectra, respectively. Fig. 1 shows the typical gamma-ray emission spectra collected with 2650 keV incident proton beam on thick targets of Al, Si and P. For each run, the incident beam charge on the Si and P thick targets were determined by RBS measurement of the thin Au films deposited on the surface of the two targets. The uncertainty in the determination of beam charge was estimated to be about 5.5%, including mainly the uncertainties from thickness of the Au film (±5%), the solid angle (±2%), and proton Rutherford cross sections from Au due to uncertainties in the proton beam energy in the Au film and in the particle detector scattering angle (±1%). The thicknesses of the Au films on Si and P targets were found to be $(72 \pm 4) \times 10^{15}$ and $(55 \pm 3) \times 10^{15}$ atoms/cm^2, respectively by fitting measured RBS spectra of 2000 keV alpha particles using SIMNRA code [19]. The RBS spectrum along with simulated spectrum is presented in Fig. 2.

For the Al thick target, the incident charge for each run was determined by fitting the backscattered proton spectra by SIMNRA code. It should be mentioned that the experimental cross section data of proton elastic scattering on Al were taken from Ref. [20]. In this case, the uncertainty in the determination of beam charge was estimated to be about 5%, including the uncertainties from the stopping power (±4% [21]), the solid angle (±2%), and the experimental proton elastic scattering cross sections (±2.5% [20]).

3. Experimental results and discussion

The experimental thick target gamma-ray yields $Y_\gamma(E,\theta)$ as a function of the proton energy E and gamma-ray detection angle θ were determined from the following equation:

$$Y_\gamma(E,\theta) = \frac{N_\gamma(E,\theta) \cdot k(E)}{4\pi \cdot \varepsilon_{abs}(E_\gamma) \cdot Q} \tag{2}$$

where $N_\gamma(E,\theta)$ is the net area of the gamma-ray peak, $\varepsilon_{abs}(E_\gamma)$ is the absolute efficiency of the gamma-ray detector corresponding to the energy of each gamma-line, Q is the incident beam charge and k(E) is a correction factor used when the target is a chemical compound