THICK TARGET YIELD MEASUREMENT OF $^{32}S(d, p\gamma_1)^{32}S$ REACTION USING A DEDICATED PIGE EXPERIMENTAL CHAMBER

O. Kakuee[†], V. Fathollahi, M. Lamehi-Rachti, A. Jokar

Physics and Accelerators Research School
Nuclear Science and Technology Research Institute
P.O. Box 14395-836, Tehran, Iran

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In this research work, thick target yields for gamma-ray emission from the $^{32}S(d, p\gamma_1)^{32}S$ ($E_\gamma = 841$ keV) nuclear reaction were measured by bombarding pure-element sulfur target with deuterons in the energy range of 1300–2000 keV. Gamma rays were detected with a high purity germanium detector (HPGe) 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%. The measurements were conducted in a reaction chamber optimized for Particle Induced Gamma-ray Emission (PIGE) spectrometry. The main advantage of this reaction chamber is that the employed charged particle detector can be readily approached to or retracted from the target — along the preferred direction of RBS measurement — during the experiments to perform PIGE measurements with the least possible uncertainty in beam charge collection at a wide range of beam currents.

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1. Introduction

Ion Beam Analysis (IBA) are powerful techniques which make use of nuclear data for a wide range of analytical applications in materials science, art and archeology, geology, surface and interface engineering, and environmental studies. These techniques due to their non-destructive nature, wide range of elements accessible to analysis, the possibility of simultaneous measurement of several elements and their quick measurement are valuable

[†] Corresponding author: okakuee@aeoi.org.ir

investigation methods for the quantitative analysis of elements in surface region of solids [1, 2]. However, most common IBA techniques *i.e.* Particle Induced X-ray Emission (PIXE) and Rutherford Backscattering Spectrometry (RBS) are deficient in analysis of low-Z elements. PIGE as a powerful complementary IBA technique with high sensitivity in analysis of low-Z elements could overcome this shortcoming [3, 4].

Based on the Coordinated Research Project (CRP) recently conducted by IAEA [5], a reputable reference database for PIGE spectroscopy is being developed. The Van de Graaff lab in Tehran had a great contribution in PIGE cross section and thick target yield measurements of a number of light elements including sulfur in this project [6]. Sulfur can be analyzed by PIGE technique employing the $^{32}\mathrm{S}(p,p'\gamma)^{32}\mathrm{S}$ reaction via detecting the 2230 keV gamma rays. The $^{32}\mathrm{S}(p,p'\gamma)^{32}\mathrm{S}$ reaction exhibits two strong and isolated resonances at $E_p = 3379$ keV ($\Gamma = 1.0$ keV) and 3716 keV ($\Gamma = 1.5$ keV) [7] which are suitable for sulfur determination and depth distribution in near-surface layers of materials [8, 9]. The disadvantage of this reaction is its small gamma-ray yield at proton energies lower than around 3 MeV [4]. Therefore, PIGE using proton beams up to 3 MeV — accessible by commonly available accelerators — is not a suitable analytical technique for the quantification of small amounts of sulfur. One can solve this deficiency of PIGE by the use of $^{32}\mathrm{S}(d,p\gamma_1)^{32}\mathrm{S}$ ($E_\gamma = 841$ keV), a deuteron induced gamma-ray emission (DIGE) reaction [4]. To our knowledge, only a few systematic studies concerning the measurement of DIGE thick target gamma-ray yield data for sulfur have already been published in the literature in the deuteron energy range suitable for IBA purposes [10, 11]. Their data have been obtained at the detection angle of 135°.

Obviously, reliable performance of the IBA techniques is based on the utilization of the accurate nuclear data, implementation of particular analysis software for each of the IBA techniques and an appropriate geometry of the experimental setup. To meet the latter requirement, a dedicated experimental PIGE reaction chamber with the added capability of RBS/PIXE analysis was developed in this lab. The main design requirements and features of this home-made PIGE reaction chamber are outlined in Section 2.1.

The aim of the present research work is to provide reliable thick target yield data for the analysis of sulfur by the DIGE technique. In this work, we present absolute thick target yield data for gamma-ray emission from the nuclear reaction of $^{32}\mathrm{S}(d,p\gamma_1)^{32}\mathrm{S}$ ($E_\gamma = 841$ keV) in the energy range of 1300–2000 keV and at the detection angle of 90° in the laboratory frame of reference as well as the capabilities and special features of the recently-developed PIGE reaction chamber.

2. Experimental setup

2.1. The dedicated PIGE reaction chamber

The experimental setups for PIGE analysis in some distinguished IBA labs are outlined in Table I. As it can be seen in this table, in PIGE measurements, high efficiency HPGe detector is preferred and the incident ions are measured either directly by the Faraday cup or indirectly by simultaneous RBS analysis.

TABLE I

Experimental setups for PIGE analysis in some distinguished IBA labs.

Laboratory	CFNUL [12]	CMAM [6]	Demokritos [13]	U. Helsinki [14]	HAS ATOMKI [15]	INFN LABEC [16]	Ruđer Bošković [17]
Gamma-ray detector type	HPGe 45% nominal efficiency	Reverse Electrode Coaxial Ge	HPGe 100% relative efficiency	HPGe 38% relative to 4 × 4 NaI(Tl)	HPGe 40% relative efficiency	Two HPGe with 50% and 25% nominal efficiency	HPGe 20% relative efficiency
Gamma-ray detector to target distance [cm]	5.65	21.06	25	2	9.5	20	6.6
Gamma-ray detector angle to beam direction	135°	135° 150°, 165°	0°, 15°, 40°, 55°, 90°, 105°	55°	55°	90° and 45°	135°
Beam charge collection	Target and chamber as a Faraday cup	Polarized target	Target and chamber as a Faraday cup	Faraday cup after target	Faraday cup	Faraday cup after target	Faraday cup beyond target, suppressor around target
Beam charge normal-ization	RBS (90°)	—	RBS	—	RBS (135°)	RBS (150°)	—

Based on the type of gamma-ray transition, the intensity of the reaction product in PIGE is either isotropic or anisotropic which depends on the scattering angle. Therefore, the preferred detector angles to beam direction are chosen to be 55° and 125° [4]. Moreover, the scattering angle of 90° is the accepted angle of PIGE detector setup by the IBA community. It should be mentioned that scattering angle of 135° is commonly used for PIXE detector setup by this community. One can obviously observe the inconsistency of the preferred gamma-ray detector angle to beam direction with some of those indicated in Table I.

Having this inconsistency in mind and the need for accurate measurement of PIGE reaction products at the preferred detection angles as well as our desire for simultaneous analysis of PIGE, PIXE and RBS led us to design and fabricate a reaction chamber with the following specifications.

The chamber is made of an aluminum alloy lined with 1 mm-thick tin (Sn) to minimize the PIGE background radiation. In fact, for measurement of low concentration elements in the sample, minimized background radiation is a serious requirement. In the design of our PIGE reaction chamber, the following points have been taken into account:

— Having the maximum attainable solid angle for measurement of PIGE reaction products with the minimum physical volume of the chamber for rapid evacuation.

— The possibility for measuring products of nuclear reaction at the scattering angles of 55° and 90° using gamma-ray detectors.

— Equipping the chamber with a charged particle detector at backward angle of 165° so that RBS analysis and measurement of incident beam current are feasible.

— Equipping the chamber with an X-ray detector for PIXE analysis of the sample at 135°.

— Equipping the chamber with an isolated target holder accommodating a reasonable number of samples at the same time.

It should be noted that variation of cross-section values of nuclear reactions of interest and natural isotope abundance of the target elements impose PIGE measurements to be conducted in a wide range of beam currents. In PIGE measurements with high-beam currents, one should be able to retract the charged particle detector from the target along the direction of RBS measurement which could not only reduce the pile up and dead-time effects but also save the life-time of the detector. In contrast, in measurements with low-beam currents, one should be able to readily approach the charged particle detector to the target to have an appropriate count rate and conduct the experiment in a reasonable time. In practice, based on this capability, our PIGE cross-section measurements have been conducted in a wide range of beam currents from 5 nA up to 600 nA [18, 19].

The main housing of the PIGE reaction chamber (Fig. 1) is designed to accommodate gamma-ray detectors at 55° and 90° and X-ray detector at 135°. In our design, the HPGe detector could be placed close to the target and isolated from the vacuum of the chamber using a 1-mm thick Al sheet. Moreover, one could mount a charged particle detector at 165° with the possibility of changing its solid angle without breaking the vacuum. In addition, required ports for connecting vacuum measuring equipment, beam entrance and beam exit accessories to the chamber are foreseen.

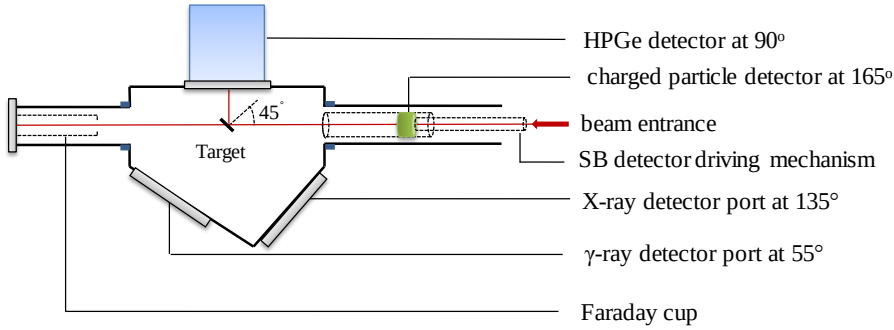

Fig. 1. Schematic diagram of the developed PIGE reaction chamber and experimental geometry.

The Faraday cup could also be used for the beam monitoring in thin film analysis where the incident beam is transmitted through the target [20]. The graphite Faraday cup was installed far enough from the HPGe detector to avoid additional gamma-ray background due to the suppression of high-current beams. The impinging charge on the Faraday cup could be directly measured by a digital current integrator and recorded by a data acquisition system. For thick targets, however, the beam stops at the target and since the target holder is electrically isolated, the total charge impinging on the target could be measured similarly. This approach could lead to a considerable error in the beam charge measurement [21]. In Fig. 2, a photo of the introduced PIGE reaction chamber is shown. In Fig. 3, a photo of the PIGE reaction chamber after installation in the beamline is shown.

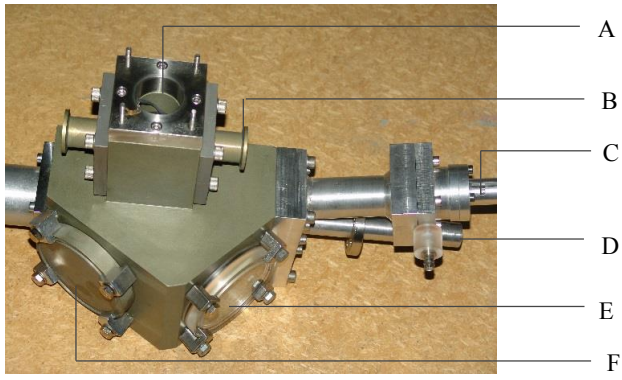

Fig. 2. A photo of the developed PIGE reaction chamber, (A) sample introducing port, (B) vacuum gauge connecting port, (C) charged particle detector driving mechanism, (D) beam entrance tube, (E) X-ray detector port at 135°, (F) gamma-ray detector port at 55°.

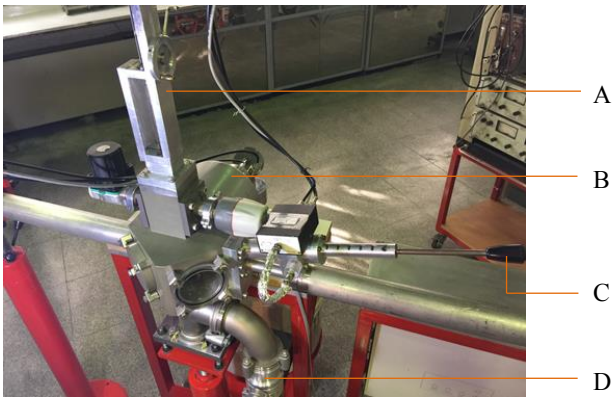

Fig. 3. A photo of the PIGE reaction chamber after installation in the beamline, (A) sample interchange mechanism, (B) cylindrical lead shield of the HPGe detector at 90°, (C) handle for driving mechanism of the charged particle detector, (D) turbomolecular vacuum pump.

2.2. Experimental details

The experimental work was conducted on the PIGE beamline (45° right) of the 3 MV Van de Graaff electrostatic accelerator of Nuclear Science and Technology Research Institute (NSTRI) in Tehran. The ion beam energy was determined based on the field strength of the analyzing magnet, as measured by a Nuclear Magnetic Resonance (NMR) fluxmeter. The beam energy calibration of the accelerator was performed through the $^{27}\mathrm{Al}(p,\gamma)^{28}\mathrm{Si}$ reaction at $E_{\mathrm{resonance}} = 991.88$ keV and through the $^{7}\mathrm{Li}(p,n)^{7}\mathrm{Be}$ reaction at $E_{\mathrm{threshold}} = 1880.44$ keV. The employed targets for this aim were a 60 μm-thick Al foil and a LiF pellet, respectively. After calibration, the uncertainty of the deuteron beam energy was estimated to be about 2 keV. The incident beam was collimated using two fixed diaphragms, 3 mm and 5 mm in diameter, placed at 87 cm and 250 cm from the target, respectively. The spot size of the deuteron beam on the target was kept around 4 mm in diameter. The orientation of the target inside the reaction chamber was adjusted so that the incident beam made an angle of 45° with the normal to the target. The experimental setup includes a coaxial type HPGe detector, a silicon charged particle detector, an electrically isolated sample holder and a cylindrical Faraday cup electrically connected to the target to quantify the incident beam current. The vacuum of the reaction chamber was checked during the measurements and found to be about 5×10^{-5} mbar. The employed gamma-ray detector for PIGE measurements was a p-type HPGe detector with crystal size of 6.58 cm × 6.58 cm and an active volume

of 213 cm^3 positioned perpendicular to the beamline direction and 5.2 cm from the target center. To conduct accurate gamma-ray detection, we had to decrease the disturbing effects. For this purpose, a 5 cm-thick cylindrical lead shield along with a 3 mm-thick copper (Cu) lining was used to protect the detector. Moreover, the Faraday cup was set farther and a number of lead bricks were set around the diaphragms as well as the Faraday cup tube.

The detector employed for the detection of the scattered deuterons was an ion-implanted silicon detector (25 mm^2 active area, 300 μm thickness and 13 keV energy resolution) positioned at the scattering angle of 165°. Based on the geometrical calculations, the solid angle of the silicon detector was found to be 1.65 ± 0.04 msr.

During the measurements, the incident beam current was varied within 2 nA $\sim$ 20 nA depending on the deuteron energy 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 was less than 10%.

2.2.1. Target

The thick target of sulfur was prepared by pressing sulfur powder into 1-mm thick pellets with a diameter of 10 mm using a hydraulic press. A thin Au film was then vacuum deposited on the surface of the prepared sulfur pellets to measure the incident beam charge and to prevent charging up by the incoming ion beam. Simulation of the collected RBS spectra of 1400 keV deuteron beam by SIMNRA code [22], indicated that the thickness of Au film was about$(50 \pm 3) \times 10^{15}$ atom/cm^2.

2.2.2. Absolute efficiency of the HPGe detector

The absolute efficiency (ε_{abs}) of the HPGe detector was derived by measuring the gamma-ray yields obtained by the calibrated sources of ^{133}Ba, ^{152}Eu, ^{137}Cs, ^{60}Co and ^{241}Am, placed at the exact target position. The absolute efficiency curve for the detector was determined by fitting the experimental data with a third degree polynomial function (1) [6]

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

where E_γ is the energy of each gamma-ray line. The branching ratios of the calibrated source gamma-ray lines were taken from Ref. [23]. By interpolating the fitted experimental calibration points, the absolute efficiency of the HPGe detector for the 841 keV gamma ray was found to be $(11.9\pm0.8)\times10^{-3}$.

2.2.3. Gamma-ray yield and proton beam charge

Yield of the gamma-rays 841 keV and the incident beam charge were determined from the simultaneous collection of gamma-ray spectra and backscattered deuteron spectra, respectively. Figures 4 and 5 display the gamma-ray and backscattered deuteron spectra collected at 2000 keV incident deuteron beam on the Au/S thick target. Collecting the gamma-rays and particle spectra at the same time interval has the advantage of removing large systematic uncertainty due to the direct charge integration. For each run, the incident beam charge on the S thick target was determined by RBS measurement of the thin Au films deposited on the surface of the target.

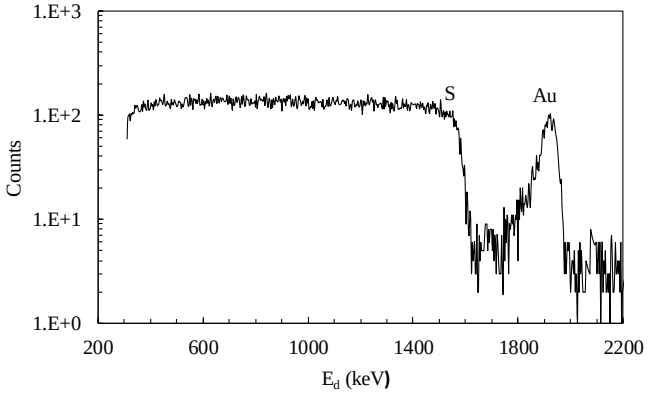

Fig. 4. The RBS spectrum of 2000 keV deuterons from the thick Au/S target at 165°.

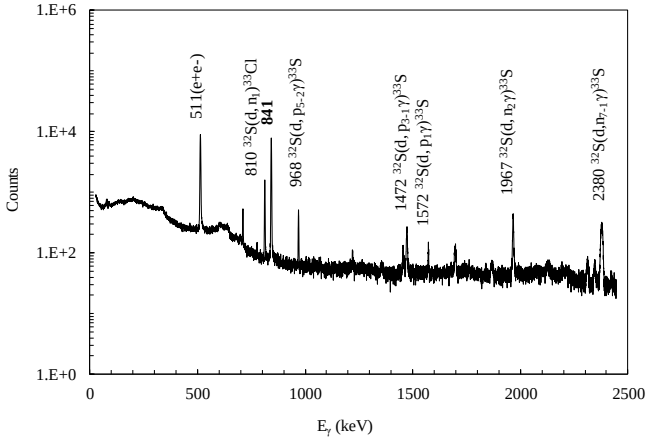

Fig. 5. The measured gamma-ray spectrum induced by 2000 keV deuterons from the thick Au/S target at 90°.

The uncertainty in the determination of the beam charge was estimated to be about 5.5%, including mainly the uncertainties from the thickness of the Au film (±5%), the solid angle (±2%), and deuteron Rutherford cross sections from Au due to uncertainties in the deuteron beam energy in the Au film and in the particle detector scattering angle (±1%).

3. Results and discussion

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

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

where $N_\gamma(E,\theta)$ is the net area of the gamma-ray peak, $\varepsilon_{\mathrm{abs}}(E_\gamma)$ 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 comprising the element under investigation.

The measured thick target yields of the reaction $^{32}\mathrm{S}(d,p\gamma_1)^{32}\mathrm{S}$ (E_γ = 841 keV) are shown in Fig. 6 for the deuteron energy range of 1300–2000 keV with an energy step of 50 keV at a detection angle of 90°, along with the published data in this energy range [10, 11]. It should be noted that the induced 841 keV gamma-ray emission has an isotropic angular distribution due to the $\frac{1}{2}$ spin value of the initial state. To avoid neutron background due

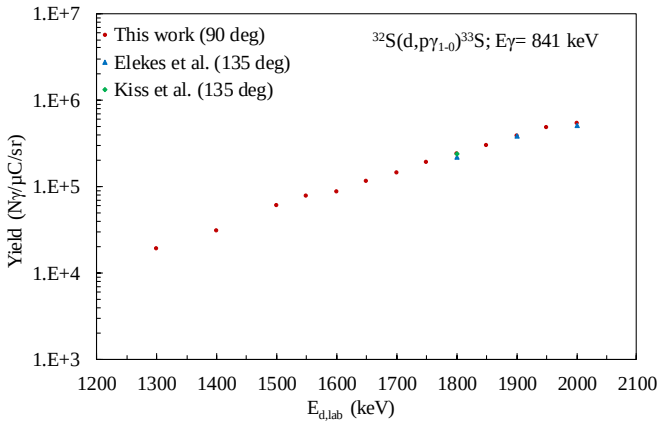

Fig. 6. The measured thick target yield data for the induced 841 keV gamma-ray emission due to $^{32}\mathrm{S}(d,p\gamma_1)^{32}\mathrm{S}$ reaction. For comparison, available data from the literature are also shown.

to the earlier implanted deuterons in the target through D(d,n) reaction, the measurements started from higher ($E_d = 2000$ keV) deuteron energy and proceeded downwards.

The uncertainty in the obtained thick target yield data was estimated to be less than ±9%, due to the uncertainties in the absolute efficiency of the HPGe detector (±7%) and beam charge normalization (±5.5%). As Fig. 6 shows, our results are in a very good agreement with the published data at the identical measured energy points with regard to the estimated uncertainty. It is worth mentioning that in DIGE measurements, neutrons produced from deuteron induced reactions on C and O nuclei encountered by the incident beam could lead to additional gamma-ray background. Therefore, to conduct accurate gamma-ray detection and to enhance DIGE sensitivity, especially in cases where the amount of the element of interest is small and/or the cross sections are low, it is necessary to minimize the level of gamma-ray background.

4. Summary

In the present study, thick target yield data for the ^{32}S$(d,p\gamma_1)^{32}$S (E_γ = 841 keV) reaction were obtained in the 1300–2000 keV energy range at $\theta_{\text{lab}} = 90^\circ$ in order to analyze sulfur in various samples by DIGE spectrometry. The obtained results are in a good agreement with the corresponding reported data in the literature. A versatile compact IBA reaction chamber optimized for PIGE measurements which was employed in this research work, is introduced. The measured thick target yields will be made accessible to the user community through Ion Beam Analysis Nuclear Data Library (IBANDL).

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