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Proton induced gamma-ray production cross sections and thick-target yields for boron, nitrogen and silicon

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ABSTRACT

The excitation functions for the reactions ^{14}N(p,p′γ)^{14}N, ^{28}Si(p,p′γ)^{28}Si and ^{29}Si(p,p′γ)^{29}Si were measured at an angle of 55° by bombarding a thin Si_3N_4 target with protons in the energy range of 3.6–6.9 MeV. The deduced γ-ray production cross section data is compared with available literature data relevant for ion beam analytical work. Thick-target γ-ray yields for boron, nitrogen and silicon were measured at 4.0, 4.5, 5.0, 5.5, 6.0 and 6.5 MeV proton energies utilizing thick BN and Si_3N_4 targets. The measured yield values are put together with available yield data found in the literature. The experimental yield data has been used to cross-check the γ-ray production cross section values by comparing them with calculated thick-target yields deduced from the present and literature experimental excitation curves. All values were found to be in reasonable agreement taking into account the experimental uncertainties.

1. Introduction

Particle Induced Gamma-ray Emission (PIGE) is an analytical technique based on the use of MeV charged particle beams. It can be utilized to determine the composition of the surface region of solids for the light elements by measurement of characteristic prompt γ-rays.

Previous thick-target gamma-ray yield measurements on natural nitrogen (see e.g. [1]) have shown that there are practically only two γ-rays which can be utilized for analytical purposes. These are the 2313 keV γ-rays from the ^{14}N(p,pγ_{1-0})^{14}N reaction and the 4439 keV γ-rays from the ^{15}N(p,αγ_{1-0})^{12}C reaction. The available γ-ray production cross section and thick-target yield data for these reactions are scarce. In this work we concentrate on the utilization of the former reaction and restrict to proton energies ⩽7 MeV. Phillips et al. [2] have determined normalized cross sections within the energy range of 3.75–6.36 MeV. Dyer et al. [3] provide data for proton energies from 3. 7 MeV up to 23 MeV and H. Benhabiles-Mezhoud et al. provide cross section data for energies 6.55–26.2 MeV [4]. The available thick-target gamma-ray yields for this reaction are even scarcer. Only the individual data points at proton energies of 3.8 MeV [1], 7 MeV [5] and 3.537–4.13 MeV [6] can be found.

Silicon can be analysed by PIGE employing either the ^{28}Si(p,p′γ)^{28}Si reaction or the ^{29}Si(p,p′γ)^{29}Si reaction by detecting the γ-rays of 1779 keV and 1273 keV, respectively. Although a considerable body of data already exists in the nuclear physics literature, in relation to cross sections for nuclear reactions with gamma-rays in the exit channel, this has not been compiled aiming at ion beam based analytical use. The study of Ref. [3] provides γ-ray production cross section data for the reaction ^{28}Si(p,p′γ)^{28}Si for proton energies from 3.7 MeV up to 23 MeV, in the study of Ref. [7] data for energies 2.2–3.8 MeV is provided and in Ref. [8] for energies 2.0–3.2 MeV. For the reaction ^{29}Si(p,p′γ)^{29}Si no ion beam analysis relevant cross section data can be found in the literature for the presently considered energy range. Even more, the only available previous data is those of Jokar et al. for energies 2.0–3.2 MeV [8]. For silicon the following individual thick-target gamma-ray yield data points are available in the literature; at 2.4 MeV [9], 3.1 and 3.8 MeV [1], 7 MeV [5], 3.05–4.09 MeV [10] and 2.65–4.13 MeV [6]. A more systematic approach has recently been taken by Jokar et al. showing yield values from 2.4 MeV up to 3 MeV with 10 keV energy steps [8].

Boron isotopes can be detected by the reactions ^{10}B(p,αγ), E_γ = 429 keV; ^{10}B(p,p′γ), E_γ = 718 keV and ^{11}B(p,p′γ), E_γ = 2125 keV. The corresponding available thick-target gamma-ray yields are for energies 3.1–4.2 MeV [1]; 2.4 MeV [9]; 7 MeV [5]; 2.49–4.13 MeV [6] and 1.77–4.08 MeV [10].

Here we provide γ-ray production cross section data for the reactions ^{14}N(p,p′γ)^{14}N, ^{28}Si(p,p′γ)^{28}Si and ^{29}Si(p,p′γ)^{29}Si as well

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as thick-target gamma-ray yields for boron, nitrogen and silicon in the energy range of 3.6–6.9 MeV. It should be noted that the yield values provided in this work correspond to samples with natural isotopic abundance, i.e. they are not corrected to correspond to pure isotopic composition. The present study is part of an extensive effort, coordinated by the International Atomic Energy Agency (IAEA), to generate a reference database for PIGE spectroscopy.

2. Experimental

2.1. Accelerator

The proton beam was generated by the 5-MV belt-driven tandem accelerator of the University of Helsinki. All the run parameters of the accelerator are controlled by industrial PLC's via an operator interface coded in NI *Labview*™. The energy of the proton beam at target is defined by a 90° analyzing magnet with a bending radius of 1500 mm. The flux density of the magnetic field is measured using a *Group*3™ teslameter with the Hall probe located between the magnet poles near the midpoint of the circular beam path. The beam energy spread (FWHM) as measured at E_p = 1 MeV was about 500 eV. The $^{14}N(p,p'\gamma)^{14}N$ reaction with resonances at 3903, 3996, and 5937 keV have been exploited in calibration of the absolute proton beam energy at 4–6 MeV to a precision of better than 1 keV. The essential factor in the procedure is the commercially available appropriate thin Si_3N_4 membrane target. The method is reliable and fast providing accelerator energy calibration at the level sufficient for practical ion beam applications. The procedure is described in detail in Ref. [11].

2.2. Detector

The γ-rays were detected with a 38%-efficient HPGe germanium detector positioned 20 mm from the target. The detection angle was fixed to 55° relative to the beam direction. The proton currents were adjusted to keep the detector count rate fixed and the dead time below 1%. The absolute efficiency curve for the detector was determined by using Eu-152, Co-60 and Co-56 calibration sources placed at the position of the target. The branching ratios of the calibration source gamma lines as provided in the IAEA compilation [12] were adopted in the calculations. The experimental data points were fitted by (beyond the maximum of the efficiency curve),

$$\varepsilon(E_\gamma) = a + \frac{b}{E_\gamma} + \frac{c}{E_\gamma^2} + \frac{d}{E_\gamma^3} \quad (1)$$

where E_γ is the energy of the γ-ray line.

2.3. Targets

As a target for the cross section measurements, a thin self-supporting Si_3N_4 membrane obtained from Silson Ltd was employed. The nominal foil thickness was 100 nm ± 10% and the window area of the membrane was 5×5 mm^2. Across a single membrane the thickness variation is much better than 1% and the membrane roughness is not considerably worse than 0.5 nm. The selected membrane thickness was chosen to ensure sufficient counting rate of the γ-ray detector, but so that the membrane can still be considered as a thin target in the present experiments. The estimated energy loss of 4 MeV protons in the Si_3N_4 target is about 2.5 keV and the energy straggling about 3 keV, based on SRIM calculations [13]. The thin Si_3N_4 membrane composition and areal density was determined accurately by Elastic Recoil Detection Analysis (ERDA).

For cross checking purposes also thick-target yields for boron, nitrogen and silicon were measured at various proton energies utilizing thick boron nitride, and Si_3N_4 samples. The sample compositions were determined by Elastic Recoil Detection Analysis (ERDA).

3. Measurements

3.1. Cross sections

The excitation functions for the reactions $^{14}N(p,p'\gamma)^{14}N$, $^{28}Si(p,p'\gamma)^{28}Si$ and $^{29}Si(p,p'\gamma)^{29}Si$ were measured at an angle of 55° by bombarding the thin Si_3N_4 target with protons in the energy range of 3.586–6.920 MeV with 100 keV energy steps except for the energy regions close to reaction $^{14}N(p,p'\gamma)^{14}N$ resonances where the energy steps were more narrow. The collected charge for each measurement point was ~12 μC with beam currents varying from 5 nA to 10 nA. A tantalum collimator (with a circular aperture of 4-mm diameter) was used in front of the target to ensure that the detected protons passed only through the target foil. The beam was finally stopped in a shielded Faraday cup with secondary-electron suppression. Carbon buildup at the membrane surface was regularly checked for and reduced by frequently changing the irradiation spot on the membrane.

3.2. Thick-target γ-ray yields for boron, nitrogen and silicon

Proton induced thick-target γ-ray yields were measured at an angle of 55° relative to the beam direction at 4.0, 4.5, 5.0, 5.5, 6.0 and 6.5 MeV using thick BN, and Si_3N_4 targets. In the thick-target yield measurements the collected charge was measured directly from the sample, using electron suppression. The employed beam currents were <5 nA. The uncertainty budgets of the cross section and thick-target gamma-ray yield measurements are provided in Table 1.

4. Results and discussion

4.1. Cross sections for the reaction $^{14}N(p,p'\gamma)^{14}N$

The resonance decay through the ^{14}N 2313-keV, $J^\pi = 0^+$, $T = 1$ first-excited state (mean lifetime τ_m = 98.7 ± 4.5 fs) was followed. The γ-ray decay to the ground state has an isotropic angular distribution. The γ-ray energy is sufficiently high to ensure rather low background contribution in the measured spectra. It should be also noted that the respective γ-ray peak is clearly Doppler broadened providing unambiguous interpretation.

The obtained cross sections are compared with the available previous cross section data in Fig. 1. Phillips et al. [2] used

Table 1
Uncertainty budget related to the absolute differential cross-section and thick-target gamma-ray yield measurements.

Quantity	Cross section	Thick-target yield
Beam energy spread (keV)	~2.5	~2.5
Charge collection	3%	6%
Target inhomogeneity	<1%	<1%
N_T number of target nuclei per square centimeter	3%	
Target stoichiometry	0.5%	0.5%
Stopping power correction		<5%
γ-Ray peak area	1.5*–3.0**%	1%
γ-Ray detector absolute efficiency	5%	5%
Final uncertainty	10%	10%

* At resonance.
** Off-resonance.