Measurement of deuteron induced gamma-ray emission cross sections on nitrogen for analytical applications

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ARTICLE INFO

ABSTRACT

Deuteron induced gamma-ray emission cross-section values from the $^{14}N(d,p\gamma_{4-1})^{15}N$ ($E_\gamma = 1885$ keV), $^{14}N(d,p\gamma_{6-1})^{15}N$ ($E_\gamma = 2297$ keV), $^{14}N(d,p\gamma_{5-0})^{15}N$ ($E_\gamma = 7299$ keV) and $^{14}N(d,p\gamma_{7-0})^{15}N$ ($E_\gamma = 8310$ keV) nuclear reactions were measured in the 600–2000 keV range of the incident deuteron energies, in 30 keV steps at the laboratory angle of 90°, utilizing a thin $C_3H_6N_6$ target evaporated onto a self-supporting Ag film. The thick target gamma-ray yields were measured in the same experimental setup using a thick TiN target. These measurements were conducted using the deuteron beam of the 3 MV Van de Graaff electrostatic accelerator of Nuclear Science and Technology Research Institute (NSTRI) in Tehran. The obtained results were compared with the previously reported data. The validity of the measured differential cross sections was verified through a thick target benchmarking experiment. The overall systematic uncertainty of the measured values was estimated and reported.

1. Introduction

Ion-induced nuclear reaction analysis is widely applied to determine the concentration and depth profile of light elements in the near-surface layers of targets containing heavier elements, where the applicability of Rutherford Backscattering Spectrometry (RBS) technique is limited. For the analysis of nitrogen, when the nitrogen content in heavy matrices is not very low and/or there is not any signal interference from other light elements, Elastic Backscattering Spectrometry (EBS) can be considered as an efficient technique for simultaneous depth profiling of nitrogen and other light elements co-existing in the matrix [1].

In the Nuclear Reaction Analysis (NRA) technique, the $^{14}N(d,p)^{15}N$ and $^{14}N(d,\alpha)^{12}C$ reactions have been commonly used for nitrogen analysis due to their relatively high differential cross sections and positive Q-values [2–5]. However in these measurements, the particle spectrum becomes more complicated by interference between proton groups from deuteron-induced reactions with nitrogen as well as proton groups originating from deuteron-induced reactions on elements like oxygen and carbon which usually co-exist in the sample matrix or appear as surface contaminants [6–8].

Particle-Induced Gamma-ray Emission (PIGE) spectrometry is another favored ion-induced nuclear reaction analysis for the quantitative analysis of light elements. The most important PIGE advantages are: (a) the capability to provide depth profiling information, with better resolution than other Ion Beam Analysis (IBA) techniques for some isotopes, (b) the fact that it is highly isotope specific, (c) its enhanced detection sensitivity for many nuclides, (d) the capability of employing the external beam, (e) the accessibility to good energy resolution with a HPGe detector, and (f) the possibility of simultaneous measurement of several light elements in heavy matrices. In the case of some light elements such as C, N and O, the gamma-ray yields of PIGE at low proton energies are very small [9,10]. Thus, PIGE using a few MeV proton beam accessible by commonly available accelerators is not a suitable analytical technique for the quantification of small amounts of these elements. Deuteron Induced Gamma-ray Emission (d-PIGE or DIGE) can be used to solve this deficiency of PIGE due to the higher gamma-ray cross sections of deuteron induced reactions, even at energies of a few MeV, compared to the proton induced ones [11–13]. Although due to the absence of narrow resonances, DIGE is not an appropriate technique for depth profiling, it enjoys all the other mentioned advantages of PIGE. The nuclear reaction $^{14}N(d,p)^{15}N$ has a ground-state Q value of 8.61 MeV and as is evident from the simplified level scheme of the residual ^{15}N nucleus in Fig. 1, the relatively high-energy excited states $1/2^+$, $3/2^+$, $3/2^-$, $1/2^+$ and $5/2^+$ in the ^{15}N nucleus decay to the ground state $1/2^-$ through the emission of respective gamma-rays of 8310, 7299, 6322, 5298 and 5269 keV and three excited states $1/2^+$, $7/2^+$ and $5/2^+$ decay to the lower states $1/2^+$, $5/2^+$ and $5/2^+$ by emission of 3013, 2297 and 1885 keV gamma-
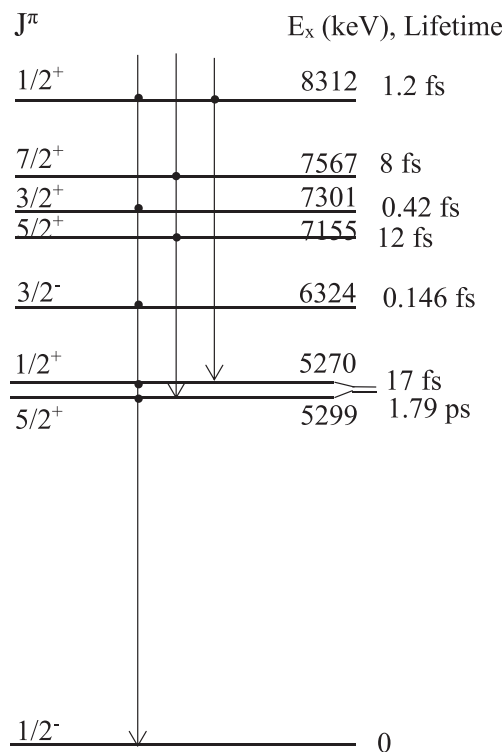
Fig. 1. The level scheme of ^{15}N with the most important gamma-ray transitions involved in the $^{14}N(d,p\gamma)^{15}N$ nuclear reactions at $E_d \leq 2000$ keV. The level scheme not drawn to scale.

rays, respectively [14–16]. Since most of the initial states in Fig. 1 have lifetimes in the order of femtoseconds (fs), Doppler broadening can be expected for the corresponding gamma-ray peaks in the gamma spectrum. The thick target gamma-ray yields reported in the literature show that the 8310, 7299, 2297 and 1885 keV gamma-lines have high enough yields and are recommended for analytical purposes at deuteron energies below 2000 keV [17,18].

For the deuteron reactions on light nuclei at low energies, both compound nucleus and direct reaction processes contribute to the cross section values [19,20]. This is why for each excitation function, there is a smooth systematic trend as a function of bombarding energy having fluctuations around an average energy. The energy fluctuations can be attributed to the superposition of several Breit-Wigner resonances in the compound system while the smooth systematic trend can be considered to be due to the direct processes [21,22].

To our knowledge, a few works have already been published on the determination of deuteron induced gamma-ray emission cross sections on light elements in the energy range suitable for IBA purposes [23–30]; among them only two published studies concerning the measurement of gamma-ray production cross-sections on nitrogen are presently available in the literature [29,30]. A dataset was also published in two different papers, but reporting the thick target gamma-ray yield data [17,18].

The purpose of the present research work is to provide reliable differential cross section values for the analysis of nitrogen by the DIGE technique. In this research work, we present differential cross section values for gamma-ray emission from the nuclear reactions of $^{14}N(d,p\gamma_{4-1})^{15}N$ ($E_\gamma = 1885$ keV), $^{14}N(d,p\gamma_{6-1})^{15}N$ ($E_\gamma = 2297$ keV), $^{14}N(d,p\gamma_{5-0})^{15}N$ ($E_\gamma = 7299$ keV) and $^{14}N(d,p\gamma_{7-0})^{15}N$ ($E_\gamma = 8310$ keV) in the energy range of 600–2000 keV at a detection angle of 90° in the laboratory frame of reference.

This study is a part of the coordinated research project organized by IAEA with the aim of developing a reference database for particle induced gamma-ray emission cross sections for ion beam analysis [31].

2. Experimental setup

The experimental work was carried out on the 45° right beamline of the 3 MV Van de Graaff electrostatic accelerator of 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 calibration, the uncertainty of the proton beam energy was found to be about 2 keV, similar to the deuteron one. 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 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. The features of the PIGE reaction chamber was described elsewhere [32]. Our experimental setup includes a coaxial type HPGe detector, a silicon charged particle 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. Gamma-rays were detected by a P-type HPGe coaxial detector with the crystal size of 6.58 cm × 6.58 cm and an active volume of 213 cm³. 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 gamma-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 from the lead. To avoid neutron background due to the earlier implanted deuterons in the target through D(d,n) reaction, the measurements started from higher (Ed = 2000 keV) deuteron energy and proceeded downwards. With descending energy, the incident beam current was increased from 5 to 15nA accordingly to compensate for the decreasing cross section values. The accumulated beam charge was chosen to be between 4 and 10 μC at each energy point depending on the statistics. In this way, the statistical error was found to be 1–3%. The detector employed for the detection of the scattered deuteron was an ion-implanted silicon detector with 25 mm² active area, 300 μm thickness and 13 keV energy resolution placed at an angle of 165° relative to the incident beam direction. Based on the geometrical method, the solid angle of the Si detector was calculated to be 3.65 ± 0.07 msr. The employed target with an area of 65 mm² was prepared by vacuum evaporation of $C_3H_6N_6$ (melamine) powder onto a self-supporting thin Ag film (about 920×10^{15} atoms/cm²). The number of N atoms in the target was determined by an EBS measurement with a proton beam of 2.7 MeV. The obtained spectrum was simulated with the SIMNRA code [33] and two experimental datasets for proton elastic scattering on N and C isotopes [34,35] downloaded through IBANDL (Ion Beam Analysis Nuclear Data Library) [www-nds.iaea.org/ibandl/]. Following this measurement, the number of nitrogen atoms in the $C_3H_6N_6$ film was found to be $(1640 \pm 100) \times 10^{15}$ atoms/cm². The energy loss of deuterons in the $C_3H_6N_6$ film of the employed target varied between 14.4 and 31 keV, based on the calculations using SRIM 2006 [36]. In order to remove the large systematic uncertainty due to the direct measurement of the absolute value of the collected beam charge, the procedure proposed by Ref. [37] was followed which recommends the use of high-Z elements in the target for normalization of the measured cross