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Differential cross section measurement of $^{16}O(d,p_{0,1})$ reactions at energies and angles relevant to NRA

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ABSTRACT

Nuclear Reaction Analysis (NRA) and more specifically d-NRA is a suitable technique for ^{16}O detection on or near surface of a compound containing heavy elements. The main problem of d-NRA is the limited number of NRA differential cross section data sets and their differences. The differential cross sections of $^{16}O(d,p_{0,1})$ reactions were measured in the present work for $E_{d,lab}$ = 700–1800 keV, at four scattering angles, namely at 90°, 135°, 150° and 165° which are suitable for NRA. Differential cross sections were collected with an energy step of ~10 keV. However, detailed measurements were carried out with an energy step of ~2 keV around resonances. The results are compared with previous studies and the similarities and differences are discussed.

1. Introduction

Oxygen is the third-most abundant element in the universe, a highly reactive nonmetallic element and oxidizing agent that readily reacts with most elements to form different compounds, mostly oxides. From material science and metallurgy point of view, determination of absolute concentration and depth profiling of oxygen at near-surface layers of different samples is important. Nuclear Reaction Analysis (NRA) and more specifically d-NRA is a suitable technique for oxygen isotopes detection [1]. The $^{16}O(d,p)^{17}O$ reaction cross section is one of the most demanded data in the IBANDL (http://www-nds.iaea.org/ibandl/) [2] and has been used for natural oxygen analysis for the following reasons:

a. It enhances sensitivity and accuracy of analysis due to its generally large deferential cross section.
b. It requires low energy deuteron beam which is accessible with low energy accelerators,
c. It has high *Q*-value (1.92 MeV).
d. The employment of deuteron beam allows simultaneous analysis of most light co-existing elements (e.g. B, C, N, F, Al, Mg and S) in the target.
e. At energies below the neutron emission threshold (1.83 MeV) the (d,n) reaction channel is closed which is important from radiation safety point of view [3,4].

Despite these advantages, the cross-section data of this reaction are only measured in a limited energy range and at a few scattering angles. However, the reported data in the literature are often inconsistent. Nuclear reactions at low energies and in light targets have a complex nature that nuclear physics theory cannot calculate them with an acceptable accuracy for IBA. These data can be obtained from experiment only with some limited of confidence. Since the differential cross section is dependent on the scattering angle, the available data can only be used for analysis in the same or a close geometry to the measurements one [3].

In this research, the differential cross section data for $^{16}O(d,p_{0,1})$ reactions in the range of $E_{d,lab}$ = 700–1800 keV and at scattering angles of 90°, 135°, 150° and 165°,suitable for NRA have been studied. The results have been compared with the reported data in the literature, as well as recently evaluated data of SigmaCalc (http://sigmacalc.iate.obninsk.ru/). The differences have been discussed in detail.

2. Experiment

Deuteron beam from the 3 MeV Van de Graaff accelerator of NSTRI was employed with an energy resolution of ±1 keV. The energy calibration of accelerator was done using the $^{7}Li(p,n)$ reaction threshold at 1880.4 keV. Deuteron beam with a spot size of 1×1.5 mm^2 was irradiated on the target with the current of 15–20 nA, resulting in a small dead time less than 10% during the data acquisition. The accumulated charge for each spectrum corresponded to ~10 μC. Differential cross sections was collected with an energy step of ~10 keV. However, detailed measurements

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were carried out with an energy step of ~2 keV around the resonances. The vacuum of reaction chamber was less than $\sim 2 \times 10^{-6}$ Torr during the measurements.

Solid ^{16}O target was prepared by anodizing tantalum substrate, a technique which is known to produce stable target with highly uniform stoichiometry and homogenous thickness [5]. Tantalum sample (a square with side length of 13 mm) was obtained from 0.3 mm thick tantalum foil (99.9% purity, confirmed by PIXE). The same sample was initially degreased with acetone and chemically polished for 15 s in a solution of H_2SO_4, HNO_3 and HF (5:2:2). The sample was then dipped in distilled water for 20 min. Anodization was carried out at room temperature with a constant current power supply at 2 mA/cm^2 in 0.1 mol (KI) electrolyte until the desired voltage of 70 V was achieved [5,6]. The purity of the prepared target was confirmed using PIXE and DIGE analysis. The stoichiometry of the target was analyzed with E_{lab} = 1.8 MeV and 2 MeV of 4He beam at a scattering angle of 165° in IBM geometry at incident angle of 60 ± 0.1°. Simulation was done using SIMNRA 6.06 code [7]. Ziegler/Biersack stopping power data and Chu and Yangs straggling model were adopted, as implemented in the code. Also pile-up and dead time corrections were considered. Based on the results of simulations, Ta and O content in the oxide layer are $(2.36 \pm 0.07) \times 10^{17}$ Ta/cm^2 and $(5.80 \pm 0.17) \times 10^{17}$ O/cm^2 respectively, with a ratio of 0.41 ± 0.02. The uncertainty of stoichiometry was estimated to be 3% due to stopping power and fitting procedure [8,9].

The detection system included four 300 μm-thick surface barrier detectors (located at 90°, 135°, 150° and 165° toward incident beam) which were covered with 26 μm-thick Kapton foil to suppress elastically scattered deuterons and alpha particles from (d, α) reaction, and another detector was used for beam monitoring (located at 165° toward incident beam). The target was tilted with respect to the beam by 15° to enable measurement at detection angles of 90°.

The differential cross section value for relative measurements was obtained using the following equation:

$$\left(\frac{d\sigma}{d\Omega}\right)^{(d,p)}_{\theta^\circ} = \frac{Y}{NQ\Omega_{165^\circ}\frac{\Omega_{\theta^\circ}}{\Omega_{165^\circ}}}$$

where Y is the experimental yield (area under the peak) and N is the areal density of the target atoms [10]. In addition, the product $Q \times \Omega_{165^\circ}$ was accurately measured by deuteron elastic scattering from Ta substrate at the scattering angle of 165° with an uncertainty of less than 3%. The solid angle ratio for every detection angle ($\frac{\Omega_{\theta^\circ}}{\Omega_{165^\circ}}$) was determined by the following formula, assuming Rutherford scattering and taking average of the two incident proton energies ($E_{p,lab}$ = 2.5 MeV and 2 MeV):

$$\frac{\Omega_{\theta^\circ}}{\Omega_{165^\circ}} = \frac{Y_{\theta^\circ}}{Y_{165^\circ}} \times \frac{\frac{d\sigma_R}{d\Omega_{165^\circ}}}{\frac{d\sigma_R}{d\Omega_{\theta^\circ}}}$$

where $\frac{Y_{\theta^\circ}}{Y_{165^\circ}}$ is the yield ratio of the proton elastic scattering on a thin layer of Au (60 nm) which is evaporated on a kapton foil [10]. The uncertainty of the Au peak area measurement was kept less than 1%. The solid angle ratios between the different detectors were determined with an uncertainty that did not exceed 1.4% in the worst case.

The first two proton levels corresponding to $^{16}O(d,p_0)^{17}O$ and $^{16}O(d,p_1)^{17}O$ were registered simultaneously and both differential cross sections were extracted from the spectra. A typical measured spectrum is shown in Fig. 1, for $E_{d,lab}$ = 1380 keV and detection angle of 135°. There was some carbon contamination on the surface of the target that was observed from $^{12}O(d,p_0)^{13}O$ reaction in the spectra. Energy correction of the target thickness and energy loss in carbon layer are calculated for each incoming deuteron beam using SRIM 2013 [11].

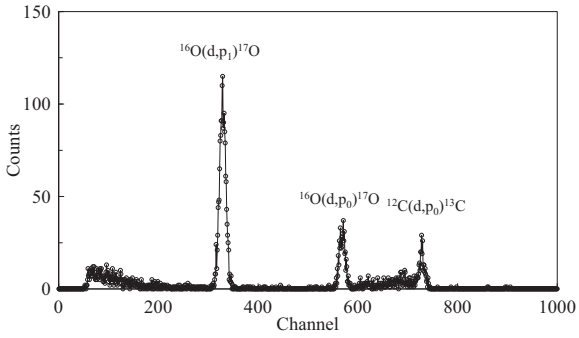

Fig. 1. A typical measured spectrum for $^{16}O(d,p)^{17}O$ reaction, $E_{d,lab}$ = 1380 keV at 135°.

3. Results and discussion

The measured differential cross sections are presented in Figs. 2 and 3, together with the available experimental data from the literature [1,4,13–19] and the SigmaCalc evaluated cross section for the $^{16}O(d,p_{0,1})^{17}O$ reactions .

The counting statistical error and background subtraction for the differential cross section varied between 3–10% for $^{16}O(d,p_0)^{17}O$ and 3–6% for $^{16}O(d,p_1)^{17}O$ reactions in most cases (at ±1σ accuracy). When cross sections are less than 1 mb/sr, consequent statistical uncertainty is ~10% which is dominant factor in determination of the total uncertainty. The total estimated systematic error does not exceed 4.5% in the least favorable case. The indicated combined experimental errors correspond to ±1σ accuracy.

Due to the co-existence of two reaction mechanisms (direct and compound), evaluation of (d,p) reactions at low energy is a theoretical challenge which has been rarely studied [12]. From NRA point of view, in the energy range of 700–1800 keV, the cross section of $^{16}O(d,p_1)^{17}O$ reaction is generally larger than that of $^{16}O(d,p_0)^{17}O$. The most often the plateau-like structure between 770 and 920 keV and a broad resonance peak at ~970 keV of the $^{16}O(d,p_1)^{17}O$ reaction is employed for natural oxygen analysis. In the targets containing ^{11}B and ^{14}N the p_1 peak of ^{16}O overlaps the peaks of $^{11}B(d,p)^{12}B$ and $^{14}N(d,p)^{15}N$ reactions. Therefore, the $^{16}O(d,p_0)^{17}O$ reaction can be used for oxygen analysis. In addition, due to higher Q-value of $^{16}O(d,p_0)^{17}O$ reaction, the p_0 peak has great chance to appear far from the backscattering background of the matrix. The $^{16}O(d,p_0)^{17}O$ reaction can also be used for analysis of thicker oxide layers [3,4].

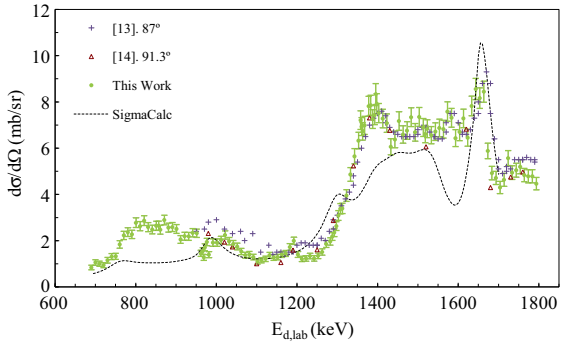

Fig. 2a. The excitation functions for $^{16}O(d,p_0)^{17}O$ reaction at 90°.