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# PRODUCTION OF HIGH ENERGY CHARGED PARTICLES DURING DEUTERON IMPLANTATION OF TITANIUM DEUTERIDES

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**Abstract** Implantation experiments using 300-keV deuteron beams are performed to study the 3-body reaction in metal deuterides with full use of *in situ* analyses of the target. The  $\Delta E$ -E telescope and the angular correlation measurements of the reaction products are made for  $TiD_x$  samples prepared with various methods. A portion of the  $\alpha$ -particle spectra with a yield ratio of  $10^{-7}$  to D(d,p) protons, which is difficult to explain by reactions with impurities and the sequential reactions, is ascribed to the 3D  $\rightarrow \alpha + p + n$  channel.

*Keywords* TiD<sub>x</sub>, implantation, 3-body reaction, ΔE-E telescope

#### 1. Introduction

Three-body nuclear reactions,  $D + D + D \rightarrow \alpha + p + n + 21.6$  MeV [1],  $\alpha + d + 23.8$  MeV [2] and  $t + ^3$ He + 9.6 MeV [3], have been claimed to take place with greatly enhanced rates during deuterium implantation of titanium deuterides. The  $\alpha$ -particle yield of the  $\alpha$ -p-n channel, for example, exceeded the theoretically evaluated value taking account of effects of electron screening [4] by 12 orders of magnitude. Theoretical models such as the formulation based on the optical theorem [5] and the multibody fusion model [2] have been presented to explain the observed enhancements from refined points of view. However, the angular correlation of the reaction products have never been confirmed yet. Moreover, it is curious that these reactions have never been observed to occur simultaneously, since the branching ratio of the final states seems to be of the same order of magnitude.

We are studying the reaction probability under various conditions of the beam and the target to clarify the phenomena. In these experiment,  $\Delta E$ -E coincidence method is employed to simultaneously measure the mass and the energy of the reaction products with special attention to helium isotopes, and the angular correlation of reaction products is also examined. After implantation experiments, these samples are analyzed *in situ* using Elastic Recoil Detection Analysis (ERDA), Nuclear Reaction Analysis (NRA) and/or Rutherford Backscattering Spectroscopy (RBS) by proton [6].

## 2. Experimental procedure

A schematic of the experimental set-up is shown in Fig. l. The target chamber is evacuated by a turbo-molecular pump to a base pressure of  $5 \times 10^{-5}$  Pa. 300 keV deuteron beams from Pelletron 5SDH-2 accelerator bombard a target with the incident angle of 30° through an aperture of 2.4 mm in diameter. The typical current density is 13  $\mu$ A/cm. A positive bias of 240 V is applied to the target to absorb secondary electrons from the target sample.

The  $\Delta E$ -E telescope is used for simultaneous measurement of the mass and the energy. A pair of solid state detectors (SSDs) with the depletion layer thickness of 20  $\mu$ m and 500  $\mu$ m is prepared around the target for detection of  $\Delta E$  and E, respectively. The solid angle is defined to  $4.3 \times 10^{-5}$  sr by an aperture of 7 mm in diameter. Output signals from each SSD are analyzed with a dual multi-channel analyzer (DMCA) with a coincidence time of 2.4  $\mu$ s, to measure the mass and the energy simultaneously.

On the other hand, angular correlation of the reaction products,  $\alpha$  and d, or t and  $^3He$ , is measured with two SSDs positioned opposite with respect to the target. One is the  $\Delta E\text{-SSD}$  used for the telescope and the other is SSD#3 whose depletion layer is 500  $\mu m$  thick. Then a  $\Delta E\text{-E}$  anti-coincidence circuit is prepared to reject possible chance coincidence caused by the D(d,p) protons. The output signals from the anti-coincidence circuit gate the angular correlation measurement.

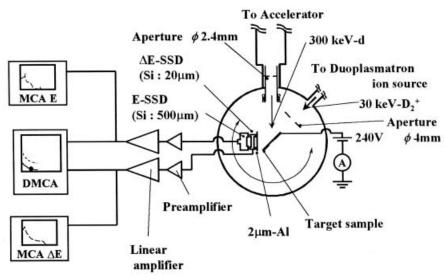


Fig. l. Experimental set-up of the  $\Delta E$ -E system.

All SSDs have been calibrated for energy using an <sup>241</sup>Am alpha source. Thin aluminum films with 2-µm thickness are mounted on all SSDs to shield thermal radiation from the target heated by the beam.

Three kinds of samples are prepared;  $300\text{-}1000~\mu m$  thick as-received titanium films,  $5\text{-}1000~\mu m$  thick titanium films deuterated by means of gas phase loading and those by ion implantation with 30-keV molecular deuterium ion beams. Some of the gas loaded samples are coated with 100-nm thick aluminum layer, which is expected to suppress deuterium loss caused by the beam heating.

#### 3. Results and discussion

A typical spectrum observed on the DMCA after 300-keV deuteron implantation of an as-received sample of Ti up to a fluence of  $4 \times 10^{18}$  cm<sup>-2</sup> is shown in Fig. 2. The horizontal and vertical axes show the energy measured at the E-SSD and the  $\Delta$ E-SSD, respectively. The D(d,p)T protons appearing around (E,  $\Delta$ E) = (2.6, 0.4) MeV predominate in the spectrum. Protons originating in (d,p) reactions with carbon, nitrogen and/or oxygen on/in the target are also distinguishable. For example, small peaks at (2.3, 0.4) MeV and (8.1, 0.2) MeV correspond to the  $^{12}$ C(d,p) $^{13}$ C and  $^{14}$ N(d,p<sub>0</sub>) $^{15}$ N (Q = 8.6 MeV) reaction, respectively. The impurity carbon is mainly introduced on the surface of the target during ion irradiation. Nitrogen and oxygen are easily introduced in/on the Ti as impurities.

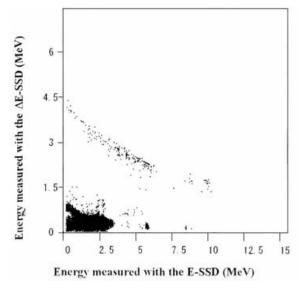


Fig. 2.  $\Delta E$ -E scatter plot of signals measured at 90° during 300-keV-d implantation of the as-received sample.

A distinct trace of  $\alpha$ -particles is observed in the energy range from 4.4 to 1.5 MeV on the  $\Delta E$ -SSD and from 0 to 10.0 MeV on the E-SSD. The  $\alpha$ -particles produced by the 3D reactions lie in this energy region and might be hidden in the trace. The total energy spectrum of only  $\alpha$ -particles deduced from the DMCA spectrum is shown in Fig. 3, and labeled as (a). For comparison a similar spectrum (b) obtained for a gas-loaded TiD<sub>1.6</sub> sample implanted with 300-keV deuterons up to a fluence of  $9 \times 10^{18}$  cm<sup>-2</sup> is also shown with shading. The  $\alpha$ -particles with energies below 4.4 MeV are lost by the  $\Delta E$ -E coincidence filtering. The peaks at 6.8 MeV and 10.2 MeV correspond to the α-particles originating in the  $^{14}N(d,\alpha_1)^{12}C$  reaction (Q = 9.1 MeV) and  $^{14}N(d,\alpha_0)^{12}C$  reaction (Q = 13.6 MeV), respectively. Subtracting the 6.8 MeV peak, a hump remains between 4.4 and 8.4 MeV in the spectrum (a), the yield of which was found to be almost independent of the fluence of the 300-keV deuteron beam. The spectral shape above 6.3 MeV differs from that of  $\alpha$ -particles emitted via sequential reactions; D(d,t)p followed by D(t, $\alpha$ )n together with  $D(d_{s}^{3}He)n$  followed by  $D(^{3}He_{s}\alpha)p$ . The energy spectrum of the latter has been calculated, and is found to be in fair agreement with the spectrum (b) with the maximum energy indicated by the dashed line.

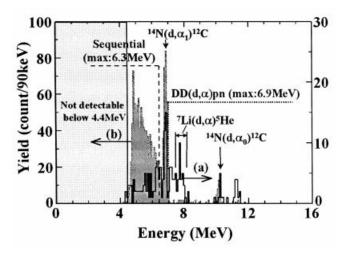


Fig. 3. Comparison of  $\alpha$  particle spectra obtained with 90-degree  $\Delta E$ -E telescope.

The reactions which could be induced by the present low-energy deuterons with a reasonable cross section and emit  $\alpha$ -particles observed in this energy region are the  $^7\text{Li}(d,\alpha)^5\text{He}$  and the  $^{17}\text{O}(d,\alpha)^{15}\text{N}$  reaction. If the latter were responsible for the hump (a), the more abundant isotope  $^{16}\text{O}$  must exist with the approximate areal density of  $2\times10^{16}/\text{cm}^2$  which is equivalent to 10 monolayers or more. And this amount of  $^{16}\text{O}$  should have produced more than  $10^4$  counts of 1.0-MeV protons via the  $^{16}\text{O}(d,p_0)^{17}\text{O}$  reaction, which we did not detect. Therefore the  $\alpha$ -particle spectrum from 6.3 to 8.4 MeV in (a) cannot be explained by the oxygen contamination alone.

We ascribe it mostly to the  ${}^7\text{Li}(d,\alpha){}^5\text{He}$  and the Li(d,2 $\alpha$ )n reaction, although we don't know the cross section of the reaction. The former produces 7.8 MeV  $\alpha$ -particles after passing through the Al shield foil, while the latter produces  $\alpha$ -particles with a continuum energy spectrum below 8.2 MeV through the Al foil. The reason why we do not have a sharp peak at 7.8 MeV is the uncertainty in the mass of the ground state of the  ${}^5\text{He}$  nucleus. The FWHM uncertainty of about 1.2 MeV [7] is calculated to result in the dispersion of  $\pm 0.4$  MeV in the energy of the  $\alpha$ -particles emitted from the  ${}^7\text{Li}(d,\alpha){}^5\text{He}$  reaction, accounting for the broad hump from 7.0 to 8.4 MeV in (a). It is not surprising that the counterpart of the  $\alpha$ -particle,  ${}^5\text{He}$  with energy around 6.1 MeV, have never been measured simultaneously, since the  ${}^5\text{He}$  travels only about 10 fm during its life time. The  $\alpha$ -particle energy was observed to decrease with increasing detection angle, as is expected for the  ${}^7\text{Li}(d,\alpha){}^5\text{He}$  reaction.

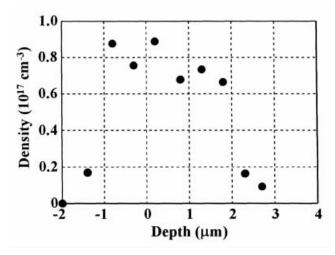


Fig. 4. Depth profile of the <sup>7</sup>Li density in the as-received Ti.

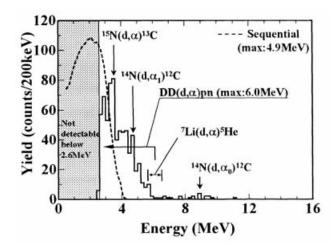


Fig. 5. The total energy spectrum of  $\alpha$ -particles during the 300-keV-d implantation of a TiD<sub>1.8</sub> implanted with 30keV-D<sub>2</sub><sup>+</sup> up to a fluence of 2.1  $\times$  10<sup>19</sup>/cm<sup>2</sup>.

The existence of  ${}^{7}\text{Li}$  has been confirmed by means of the  ${}^{7}\text{Li}(p,\alpha)^{4}\text{He NRA}$ . The depth profile of  ${}^{7}\text{Li}$  density deduced from the  $\alpha$ -particle spectrum is shown in Fig. 4. The cross section has been cited from ref. [8]. Although the estimated resolution of 530 nm is rather poor and the cross section does not allow the analysis beyond 3  $\mu$ m, the profile indicates that the as-received Ti contains lithium as an impurity with concentration exceeding 1 ppm.

There might be hidden the  $\alpha$ -particles from the 3D  $\rightarrow$  d +  $\alpha$  and 3D  $\rightarrow$  n + p +  $\alpha$  reactions. In the former case the partner products, 15.9 MeV deuterons, should be measured together with the 8.0 MeV  $\alpha$ -particles. Unfortunately, both of the deposition energies of the deuterons, 0.2 MeV in the  $\Delta$ E-SSD and 5.6 MeV in the E-SSD, are not high enough to be separated from other peaks. Moreover, since carbon has been identified in the spectra, it is possible that the 15.9-MeV deuterons are hidden by the contour of 5.8 MeV protons from the  $^{13}$ C(d,p) $^{14}$ C reaction on the  $\Delta$ E-E plot.

The angular correlation was then measured to separate the  $\alpha$ -d channel products from other reaction products using the detector pair placed opposite to each other with

respect to the 300- $\mu$ m-thick as-received target. However, no coincidence between the deuteron and the  $\alpha$ -particle was observed up to a fluence of  $4 \times 10^{18} / \text{cm}^2$ .

This result does not always exclude the possibility that the  $\alpha$ -particles through the  $\alpha$ -p-n channel of the 3D reaction are hidden, which are expected to form a broad continuum from 6.9 MeV down to 0 MeV. To separate the  $\alpha$ -particles produced at different depth, the measurement of the  $\alpha$ -particles are made at 90° behind the 5- $\mu$ m-thick Ti implanted with 30-keV-D<sub>2</sub><sup>+</sup>. The result is shown in Fig. 5. The broken line shows the calculated spectrum of  $\alpha$ -particles emitted through the sequential reactions. The  $\alpha$ -particles from the  $\alpha$ -p-n channel are expected to appear below 6.0 MeV. We could attribute the portion of the spectrum between 5.0 and 5.5 MeV to the  $\alpha$ -p-n channel. The maximum ratio of the  $\alpha$ -particle yield to that of the proton from the D(d,p)t reaction reaches  $10^{-7}$ , which exceeds the theoretically evaluated value taking account of effects of electron screening [4] by 11 orders of magnitude.

The 3D  $\rightarrow$  t +  $^3$ He reaction is also expected. However, we could not identify the  $^3$ He peak in the DMCA, and the 4.8-MeV tritons, if any, are difficult to identify, since the energies of 0.7 MeV and 4.0 MeV deposited in the  $\Delta$ E-SSD and the E-SSD, respectively, lie in the pile-up spectrum of D(d,p) protons.

### 4. Conclusions

The  $\alpha$ -particles except those originating in deuteron induced reactions with impurities and the sequential reactions have been observed in the energy range from about 5 MeV to 8 MeV for the gas-loaded sample and the as-received sample. The result of NRA using the  $^7\text{Li}(p,\alpha)^4\text{He}$  reaction, together with the absence of the coincident partner, as well as the dependence of the yield on the deuteron fluence, suggests that the portion of the spectrum from 7.4 to 8.1 MeV can be ascribed to the  $\alpha$  particles originating in  $^7\text{Li}(d,\alpha)^5\text{He}$  reaction. For the 5- $\mu$ m thick sample implanted with the 30-keV-D  $_2^+$  beam,  $\alpha$ -particles have been observed in the energy range from 5.0 to 5.5-MeV with a yield ratio of  $10^{-7}$  to the D(d,p) proton. We speculate that these are the  $\alpha$ -particles from the 3D  $\rightarrow \alpha$  + p + n channel.

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