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In-situ ERD Analysis of Hydrogen Isotopes during Deuterium Implantation of Pd

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Abstract

The elastic recoil detection (ERD) analysis is successfully applied to *in-situ* measurements of hydrogen isotope distributions formed in Pd during deuterium ion implantation aiming at observation of peculiar phenomena in connection with the so-called cold fusion. The beam-target D(d,p)t reaction yield during the implantation is found dependent on the beam current or the deuterium flux. This is interpreted in terms of a temperature dependence of the deuterium concentration that is measured *in situ* with the ERD method. When both surfaces of the Pd sample are coated with 7.5-μm thick films of aluminum oxide, the reaction yield is observed to increase by a factor of about 5, and the ERD spectra show distributions of D more localized near the surface.

1. Introduction

A trial is made to find anomalous phenomena in both charged particle spectra and yield under bombardment of deuterated Pd targets with keV/MeV ion beams. One of the key factors for the appearance of the excess heat in the electrolysis experiments has been found to be the concentration n_D of D atoms in Pd[1]. An abnormality has also been induced by a mechanical stress in the lattice of a deuterated Pd with a surface coating[2].

In view of the somewhat transient nature of these anomalous phenomena, it is very important to know the space- and time-dependent $n_D(x,t)$ in relation to the appearance of the reaction products. Moreover, since hydrogen isotopes are easily movable in metals, it is desirable to make the analysis of $n_D(x,t)$ *in situ*.

A keV-D ion implantation/irradiation system equipped with an *in-situ* elastic recoil detection (ERD) analysis system has been constructed, and the effectiveness of the *in-situ* measurements has been demonstrated[3]. In this paper we discuss a correlation between the d-D reaction yield and $n_D(x,t)$, and an effect of a surface coating with aluminum oxide both on the d-D reaction yield and $n_D(x,t)$.

2. *In-situ* Analysis System

A schematic of the system is shown in Fig.1. The system is the same as that described in detail in ref.3, except for the target. The target located at the center of the chamber is a 5 μm thick Pd film or a similar Pd film coated with Al. The Al layer was established using a vacuum evaporation, and exposed to atmosphere for oxidization.

The layer was later analyzed with RBS to find the thickness of 7.5 μm and the composition of Al_2O_x, where x≅3.5.

D_2^+ ions produced in a duoplasmatron ion source with an maximum voltage of 30 kV are used exclusively in the present work. They are injected into the target chamber through a transport chamber having a Wien filter for the mass selection. It has an exit aperture of 4 mm in diameter. The beam enters the target at an incident angle of 15° with a current density ranging from 1 to 10^2 μA/cm^2.

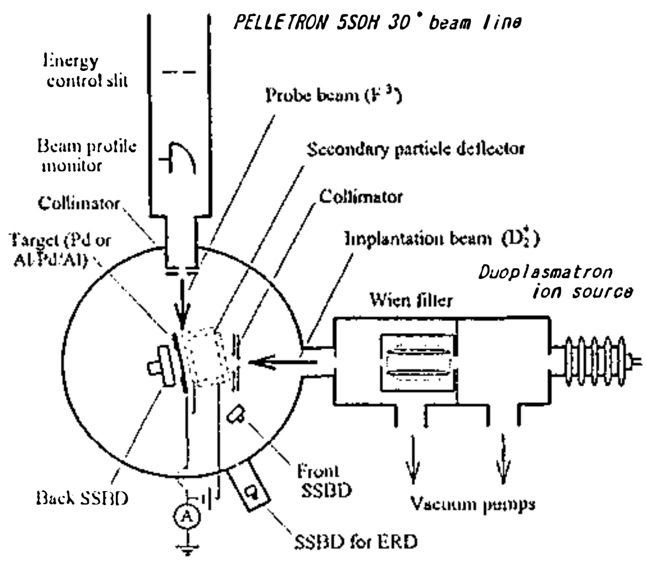

Fig 1. Schematic of experimental apparatus

The spatial distribution of the implanted D together with that of adsorbed hydrogen is measured with the ERD method occasionally; after interrupting the implantation or during the implantation. The accelerator beam for the ERD analysis is incident on the target with an incident angle of 75° through a collimating aperture of 0.5 mm in diameter. 6-MeV F^{3+} ions are used for the analyzing beam with a current of the order of 1 nA.

Three charged particle detectors, Si surface barrier detectors (SSBDs), are prepared around the target. The largest one with an effective area of 300 mm^2 is located just behind the target to detect nuclear reaction products with a large solid angle of 3.4 sr, and covered with a 15-μm Al foil to stop delta electrons and recoil particles. Another SSBD located at an angle of 30° with respect to the analyzing beam serves as an ERD detector. A 3-mm diameter aperture in front of it defines the solid angle to 8.2 $\times10^{-4}$ sr. To stop scattered (or recoil, if any) particles heavier than ^{4}He a 6.2-μm thick Mylar film covers the aperture. The third detector having an effective area of 25 mm^2 and a similar Mylar film is placed at an angle of 60° and used for measurement of scattered particles during the ERD analyses.

3. d-D Reaction Yield during Implantation

During the implantation of the targets with D ions the usual beam-target reactions D(d,p)t and D(d,n)^{3}He produce 3.02-MeV p, 1.01-MeV t and 0.82-MeV ^{3}He. For comparison with measurements, the reaction probability is calculated for a uniformly saturated composition of $PdD_{0.85}$ using a TRIM code with a modification to include the nuclear reaction[3].

An example of the evolution of the D(d,p)t reaction yield during the implantation is shown in Fig.2, where the measured yield of p, Y_m, normalized to the calculated one, Y_c, is plotted against the fluence of deuterons injected in the form of 18-keV D_2^+ molecular ions. During the run an intentional change in the beam current is cyclically made in expectation of the Takahashi effect[4].

We note almost the same dependence of Y_n on the current density as that found in our previous work[3]; the normalized reaction yield $Y_n = Y_m/Y_c$ changes following the cyclic change in the beam current. The lower is the current density, the larger is the reaction yield.

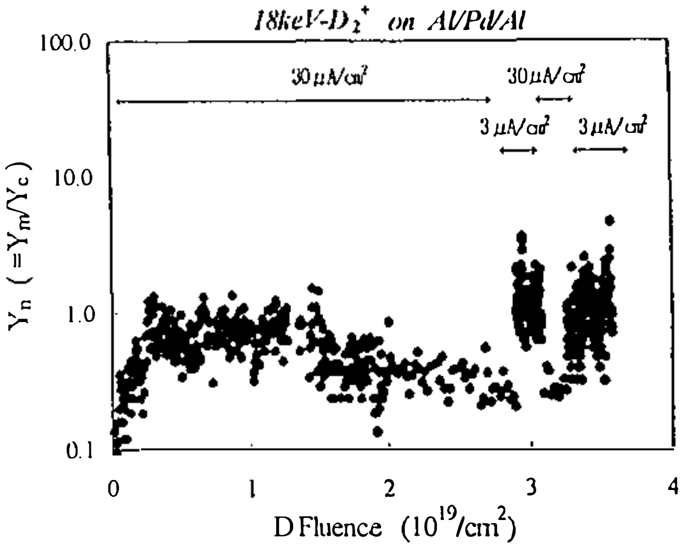

Fig.2. Evolution of normalized reaction yield Y_n during implantation of the 5μm-thick Al/Pd/Al target.

This phenomena was interpreted[3] in terms of the temperature dependence of the saturated amount of D atoms in Pd. The current density was varied in a range from 14 μA/cm^2 to 110 μA/cm^2, when Y_n changed from 1 to 0.1. In the case of the largest current density the target temperature becomes higher than the phase change temperature of Pd around 100°C, where the atomic fraction of D in Pd decreases down to 0.03. This explained the behavior of Y_n observed experimentally.

Below about 20 μA/cm^2, however, the temperature remains almost at the room temperature, and therefore n_D should remain constant at the highest composition of 0.85. Nevertheless Y_n increases above 1 as shown in Fig.2. This may be due to an effect of surface plugging by the Al_2O_x coating.

4. Deuterium Distribution in Pd and Al/Pd/Al Samples

The changes in n_D have been observed with ERD measurements made at several phases of the implantation history. In the case of Pd without Al_2O_x the D distribution has its maximum at a depth 10-20 nm into the bulk[3], which is much shallower than 55 nm, the stopping range of 8-keV deuterons in Pd. This could be interpreted qualitatively as follows. Deuteriums prefer a region having more damage as their permanent abode. On the other hand, they escape from the bulk out of the surface due to recombination into D_2 molecules. The former effect moves the distribution peak toward the surface, while the latter into the bulk. The peak is therefore determined by a compromise between the two effects.

An example of the D distributions in the Al_2O_x-covered Pd is shown in Fig.3. The distribution peak is even at a shallower depth than that in the bare Pd; near the surface or near the boundary of the Al_2O_x layer and Pd bulk. The FWHM of the peak is approximately 20 nm, which should be compared with 34 nm of that in the bare Pd. This seems to contradict the above interpretation for the shallow location of the distribution peak in Pd, since we expect a suppressed loss of D for the Al_2O_x layer.

The reaction yield Y_m should be reduced a little by the reduction of D concentration near the surface compared with the case of the fully saturated distribution, since the reaction cross section decreases rapidly with decreasing energy. As shown in Fig.2, however, the maximum value of Y_n exceeds 1.

The implantation was done up to a fluence such that the whole Pd film could become over-saturated. The target structure seems therefore to have been completely amorphised near the surface region. The value of Y_n exceeding 1 might suggest that the amorphization could cause an increase in the saturation concentration of D in Pd.

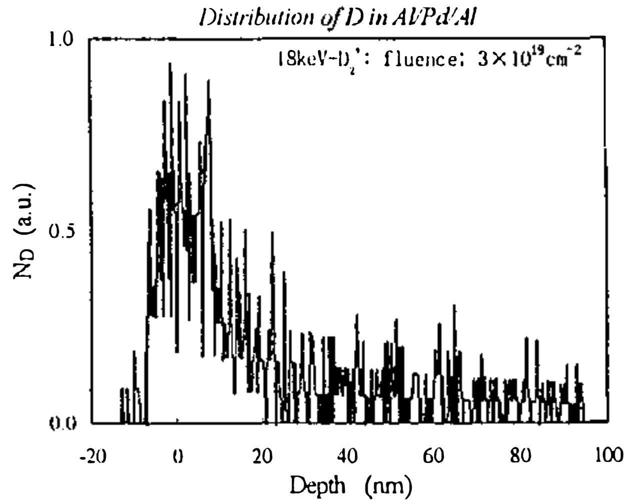

Fig.3. Typical distribution of D atoms in the Al/Pd/Al target.

5. Conclusion

The evolution of the D(d,p)t reaction yield during the D implantation of Pd with cyclically changing fluxes has been observed both on the bare Pd and on the Al_2O_x-coated Pd, and discussed in terms of the temperature dependence of the saturated concentration of D in Pd. The Al_2O_x coating is found to have an effect to localize the D distribution near the boundary.

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