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Trapping and reflection of eV deuterium ions by fusion materials

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Abstract

The experimental results on the reflection of deuterium with energies of 2.5–30 eV per nucleon from niobium are presented. D_3^+ and ArD^+ ions and various conditions of irradiation were used. Influence of the surface binding energy on the particle reflection coefficient is discussed. The possibility of using the heteronuclear ions in the investigations of the low energy hydrogen reflection and trapping is discussed based on calculations with the binary collision approximation model.

Keywords: Recycling; Deutrium

1. Introduction

The eV primary energy range of hydrogen isotopes is of great interest for studying the processes contributing to the recycling of the fuel particles in fusion devices. At least two of them, namely the reflection and trapping of the bombarding particles, have not been investigated experimentally at the energies compared with the surface binding energy E_S. There are few measurements made with molecular hydrogen ions at energies as low as 10–30 eV per hydrogen or deuterium fragment of molecule [1–4]. Recently we have measured the value of particle reflection coefficient R_N at energy of 5 eV per deuteron using D_3^+ ions [5]. Most predictions of the R_N values in eV energy range are obtained by computer simulations [6–8] and they must be tested by experiments because of the uncertainty in the description of the processes at these very low energies [9].

In this paper we present experimental results on reflection and trapping of deuterium in a Nb target with different conditions on its surface at primary energies E_0 comparable with the binding energy of the hydrogen on the surface.

The experimental results on the particle reflection coefficient R_N are compared with the calculations. The computer simulation code is based on the binary collision approximation (BCA) model and was the same as in [7]. Despite the limitations of the BCA approach at low energies, the simulated $R_N(E_0)$ dependencies were shown to fit the experimental data when some adjusting surface binding energies were used.

Niobium was used for the investigations. It is proposed as a material for superpermeation membranes [10] in the pumping area of the fusion reactor. Besides, niobium is a proper material for modelling of hydrogen interaction with other hydrogen active materials like vanadium or its alloys widely discussed for fusion applications.

2. Experiment

The experiments were performed at the ‘Medion-2’ experimental set-up described in detail earlier [11]. Briefly, an energetic primary ion beam was separated in a magnetic field and then decelerated by a retarding system before coming to the target. After retarding, the ion beam remained ‘monoenergetic’ with a low energy spread. The target was surrounded by a screen made of Ni which could be heated up to 350°C to prevent the trapping of deuterium on the chamber wall. A

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high-sensitivity mass-spectrometer was used to monitor the partial pressure of HD and D_2 molecules. To control the residual gas spectrum and target surface contamination a sector magnetic spectrometer was used. It could operate in two modes: as a mass-spectrometer for gas composition with ionisation chamber near the target and as analyzer of reflected and sputtered ions. For surface analysis the target could be tilted at different angles of incidence. The angle of registration of reflected ions and recoils is equal to 60°.

The number of the reflected particles was determined by the thermal desorption technique. The target was irradiated at room temperature, where the re-emission of the implanted particles is known to be negligible. Then it was heated up to 1300°C to release the captured deuterium. The number of the deuterium atoms released was supposed to be equal to the number of the deuterium ions trapped. Then the particle reflection coefficient can be determined by

$$R_N = 1 - \lim_{\Phi_0 \to 0} \left(\frac{\partial \Phi}{\partial \Phi_0} \right),$$

where Φ_0 is the fluence, Φ is the amount of the captured particles. The measurements were performed in the so called "dynamic regime" [12,13] where the release rate of the particles is proportional to the partial pressure of HD and D_2 molecules.

The sample used was made of high purity niobium manufactured by Daido Steel. Before the experiments, a surface composition was investigated using secondary ion mass-spectroscopy, Auger spectroscopy, and low energy ion backscattering. At room temperature its surface is covered by niobium oxide that can be removed by annealing up to 1300°C within a few hours. It was found that after removal of the niobium oxide layer the near-surface layer was contaminated with C (about 10%), O (5%), and Fe (2%) impurities. Taking into account the residual gas pressure during the experiments (about 10^{-6} Pa), the oxide covers the surface within few minutes.

We have done three series of experiments. In the first one, the target was irradiated by D_3^+ ions with energies ranging from 15 to 90 eV. As the primary energy of the ions is greater than the energy of dissociation, we supposed that the energy of each deuteron is equal to one third of the energy of the primary molecular ion. The fluence ranged from 10^{14} to 4×10^{14} D/cm^2. Because of the very low intensity of the ion beam in this energy range, the time of irradiation was rather high (tens of minutes) to accumulate the fluence necessary for measurements of the number of captured particles with reasonable accuracy. As the energy of deuterium ions is under the threshold of sputtering, the surface was covered by NbO in this set of measurements.

In the second and third series, we bombarded the target by ArD^+ ions with the primary energies in the range of 50–400 eV. We supposed that the ArD molecule dissociates as it strikes the surface, and that the energy of D atoms is proportional to the ratio of D–ArD masses (and respectively the energy of Ar fragment is proportional to the ratio of Ar–ArD). Thus, using the heteronuclear ions, it is possible to achieve extremely low energies for the light fragments of the molecule.

The third series differed from the second one by the long annealing time of the target before the measurements at the temperature of 1400°C. The intensity of the ArD^+ in these two series was much higher than in the first one. Therefore an irradiation time of about 1 min was sufficient for thermal desorption spectra (TDS) measurements. The respective fluence ranged from 2×10^{14} to 9×10^{14} D/cm^2. Analysis of the surface composition by spectroscopy of recoils and reflected ions has showed that in the second series of experiments traces of oxygen were present on the surface. The third series of experiments represented the cleanest Nb surface.

3. Results

The experimental values of R_N as a function of the primary energy of deuterons for the three series of measurements are shown in Fig. 1. In this figure we represent also the results of the computer simulations obtained by using the SCATTER program [7] based on the binary collision approximation. Calculations were performed both for NbO (solid lines) and Nb (dashed lines) to match the data obtained at different surface conditions. The surface binding energy E_S was consid-

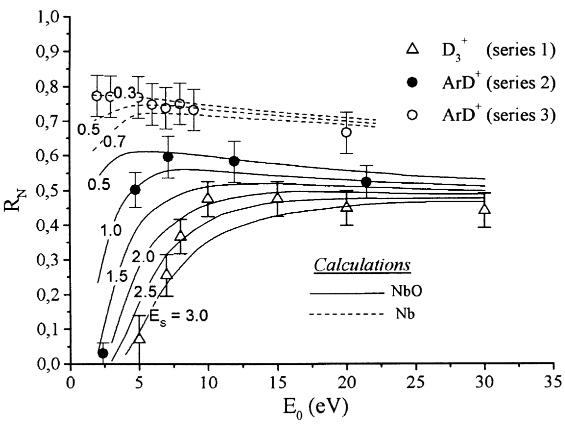

Fig. 1. The particle reflection coefficient of deuterium in Nb as a function of the primary energy of deuterium. Curves are computer calculations using a binary collision approximation: solid lines – calculations for NbO; dashed lines – calculations for Nb. Numbers at the curves are values of the "effective" surface binding energy E_S, used as an adjusting parameter in the computer calculations.

ered to be an additional potential retarding the reflected and the secondary particles leaving the surface and was used in the computations as an adjustment parameter.

It is clearly seen from the Fig. 1 that the experimental results in the three series differ significantly. For D_3^+ irradiation, a maximum of R_N was observed at $E_0 \approx 10$ eV. At lower energies, R_N decreases by about six times, and becomes equal approximately to 0.07 at the minimum energy achieved. The computer simulations fit the experimental data for $E_S = 2.5 \pm 0.5$ eV for the NbO covered surface.

The experimental values of R_N for the second series (ArD^+ bombardment) are higher than those for the first series and the maximum is shifted to the lower energies by about 3 eV. The lowest value, $R_N \approx 0.02$, is observed at an energy of about 3 eV lower than that of the first series of experiments made with D_3^+ ions. The ratio of the maximum value of the particle reflection coefficient obtained to the lowest value measured in the two experiments is about 25.

In the third series, the values of R_N were even higher than those in the second series. Argon bombardment along with thermal annealing produced a clean Nb surface in this set of experiments. It is seen from Fig. 1 that the experimental and calculated data for a Nb surface are in good agreement. This supports the suggestion that the third series of experiments corresponds to a clean target surface. For this case the decrease of R_N at low energies, typical of the two previous experiments, is not observed for energies as low as $E_0 = 2.5$ eV. The calculated data fit the experiment for $E_S = 0.4 \pm 0.2$ eV.

4. Discussion

First of all, the experiments show that $R_N(E_0)$ has a maximum. This means that the low energy component of hydrogen particles impinging on the wall of the reactor will be captured if favourable conditions on the surface arise. For oxidised and clean surfaces of Nb, the number of reflected particles at the minimal primary energies of 2–3 eV achieved in our experiments differ as much as 30 times.

Because of low flux in the first experiment (D_3^+ bombardment), the time of irradiation was rather long, and the surface became covered with oxide. In the second and third experiments, the flux was much higher, so the irradiation time was short. In addition, due to possible sputtering of oxides by heavy Ar, the surface could be considered to be less contaminated with hydrogen-active impurities. In the third experiment, we annealed the sample, and this can produce an even cleaner surface. Thus, the trend in the binding energy variation in the different experiments correlates with expectations. Therefore one can accept the BCA for modelling of low energy collisions.

Unfortunately, we do not know exactly the energy distribution of the fragments after the molecule dissociates when it strikes the surface. We simulated the influence of different distributions of fragments over energy on value of particle reflection coefficient and concluded that for low E_S (less than 0.5 eV) the variation of the energy distribution of fragments does not affect R_N noticeably. But averaging over all possible energy distributions of fragments for greater E_S values corresponds to the decrease of R_N. In fact, the direct experiment with deuterons and molecular ions of equal velocity is necessary to make clear the situation.

A serious question in the comparison of the experiment and the calculations is that ArD^+ has a heavy component that can cause some additional processes, such as sputtering of the retained D, defects production etc. We have made some calculations to investigate the possible influence of these processes on R_N. This is necessary for comparison of the results of the first experiments with the others and to investigate the applicability of ArD^+ irradiation for modelling of the low energy hydrogen atom interaction with metal. For this purpose calculations were made for D^+ and Ar^+ irradiation of Nb.

4.1. Sputtering

To evaluate the effect of D sputtering by Ar^+, we considered a two-component (Nb + D) multilayered structure. For this purpose we have calculated initially the depth profiles of D in Nb (Fig. 2) and constructed an NbD_x target consisting of several layers with the respective D content. The maximum value of the deuterium sputtering coefficient by Ar ions was found to be about 5×10^{-2} D/Ar. Therefore, sputtering leads to a small increase of the $R_N(E_0)$ dependence in the energy range below 20 eV/D if we compare it with the curves in Fig. 1. Thus, the effect of sputtering of trapped deute-

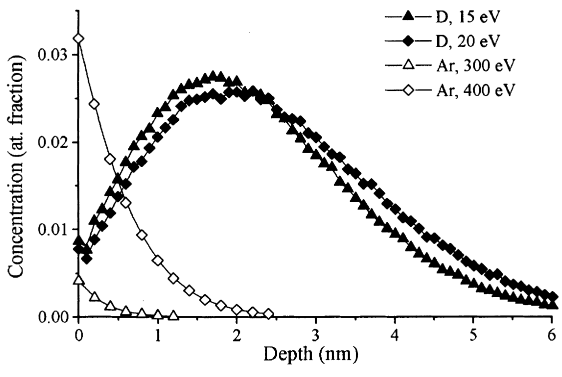

Fig. 2. Depth distributions of trapped D atoms (solid shapes) and the defects produced by Ar (open shapes).

rium by Ar^+ ions in these conditions can be considered to be negligible.

4.2. Defect trapping

Radiation defects produced by heavy Ar can trap deuterium atoms. If the temperature of implantation is low, four opportunities of D trapping can be considered: (i) trapping of the preliminary implanted deuterium in defects produced by newly incoming Ar^+ ions; (ii) trapping of a slowed down deuteron by defects produced by the previously bombarded argon; (iii) trapping of slowing down deuterons by defects produced by Ar^+ ions from another ArD^+ pair when two pairs are passing close to each other; (iv) trapping of the deuteron by the defect produced by Ar^+ arising from the very same pair. Estimations show that none of these mechanism is significant. The concentration of implanted deuterons (Fig. 2) is rather low, and the defect trapping (i) is small. The concentration of the defects is less than the concentration of atoms and trapping (ii) is even less than (i). The ion flux is low and the distance between two cascades is large and mechanism (iii) is negligible. The only opportunity for a deuterium atom to be trapped is capture in to the defect produced by Ar^+ from the very same ArD pair. To estimate the possibility of an event of this kind we analysed the position of D and Ar species propagating into the solid. Fig. 3 shows the time dependence of the mean value of the depth where D and Ar can be found for various ArD^+ energies. One can see that Ar^+, which creates the defects, is always behind the D atom. Therefore the deuteron is passing the undamaged region. Note also that Fig. 3 shows the projection to the normal, while the real distance between D and Ar is even larger.

An important parameter in the calculations of the defect production is the displacement energy E_d necessary to produce a vacancy-interstitial pair. In the calculations discussed we used $E_d = 5$ eV to show a possible effect. But the real value is higher. For Nb, it is estimated to be $E_d = 28$ eV [6]. According to the calculations no defects are formed at this displacement energy. Therefore one can expect no influence of defects in these calculations.>

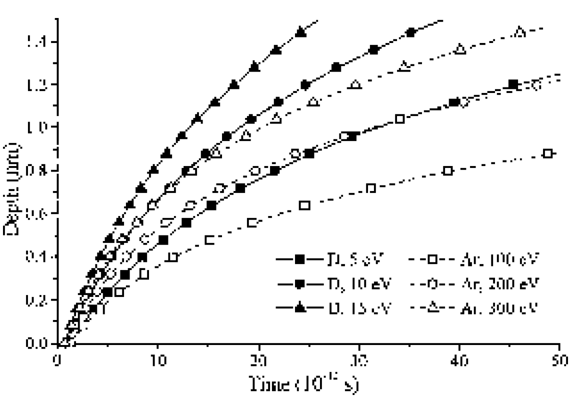

Fig. 3. The time dependence of the mean value of the depth of D (solid shapes) and Ar (open shapes).

4.3. Pushing deuterium into the bulk

According to Fig. 3, Ar always moves behind D. Therefore, the pushing of hydrogen atoms into the bulk seems to be a rare event. Summarising the model calculation discussed, one can state that within the accuracy of about of 5% (corresponding to the sputtering of implanted D atoms by Ar projectiles), the heavy Ar fracture of the primary ArD^+ ion doesn't influence the propagation of the deuterium fracture. Therefore the experiments with D^+ and ArD^+ bombardment can be compared directly.

5. Conclusions

1. Experimental investigations of low energy deuterium reflecting and trapping by niobium at normal angle of incidence and ion energies of 2–30 eV were performed. It is shown that in the eV primary energy range, the dependence of the particle reflection coefficient on the primary energy has a maximum, whose position strongly depends on the surface conditions. Computer simulation based on the BCA model show good agreement with the experimental results if the surface binding energy E_S is used as an additional adjusting parameter. Values of E_S for different surface conditions were evaluated. For the clean and oxidised surfaces, $E_S \approx 0.4$ and 2.5 eV, respectively. The difference of the particle reflection coefficient at the very low energies (about 2 eV) can be as high as 30 times. This must be taken into account in the evaluation of hydrogen isotope capture and retention during interaction of a cold plasma with plasma facing materials.

2. It has been shown that ArD^+ ions can be used for modelling of low energy (several eV) D^+ interaction with the solid. According to the calculations, after dissociation of the ArD^+ ions, Ar does not affect the light D-component for the primary ArD^+ energies of 50–500 eV. The heavy Ar atom in the beam influences only the sputtering of the retained light D-component, but the sputtering coefficient does not exceed 0.05 D/Ar. Defect production in the solid and kinematics pushing of deuterium into the bulk are negligibly small effects.

3. Good agreement of BCA model calculations with experimental results has been obtained at the eV primary energy range, contrary to the common opinion that the BCA models are not valid in this energy range.

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