Influence of dispersion of ion irradiation D + on distribution of deuterium in titanium

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Abstract. In single crystals of titanium was studied deuterium segregation induced by ion irradiation. Segregation created at room temperature deuterium ion implantation energy of 700 keV and analyzed by the nuclear reaction D (d, p) T. During the experiment changed the duration of continuous exposure and the duration of stops. It was found that with increasing radiation dose continuous average concentration of the implant in the irradiated volume gradually increases. It has been found that a long interruption of ion bombardment leads to an abrupt increase of the concentration. Analysis of the depth distribution of deuterium showed that during stopping the accumulation of radiation near the surface of the implant due to the redistribution of the target volume. The obtained results are discussed in terms of the evolution of the defect structure of the target in the course of continuous exposure and stop time of implantation.

1. Introduction

In recent years it was established that upon the operation of different-type nuclear reactors the rate of generation of hydrogen proved to be higher than it was expected. [1]. This experimental fact gave rise to the reinterpretation of the approach to the problem of hydrogen accumulation in reactor materials and brought about a rebirth of interest in it. The accumulation and retention of hydrogen in reactor materials can be to some extent simulated using radiation-induced segregation (RIS) of hydrogen, when high concentrations of radiation-induced defects and the accompanying high concentrations of hydrogen are formed in small volumes. Under the conditions of possible levelling-off of hydrogen concentrations, the magnitude of hydrogen trapping during RIS is an indicator of the defect state of materials. In other words, the application of RIS as a tool for studying the behaviour of hydrogen in irradiated materials allows one to investigate the evolution of the defect structure in the segregation regions from the dynamics of hydrogen trapping under irradiation.

It is evident that information on the magnitude of hydrogen trapping and evolution of a radiation-induced defect structure can be used to predict the level of radiation damage that is sufficient to nucleate critical elements of steels, such as gas pores. For identification of influence of interphase surfaces on the capture of hydrogen it is necessary to attract as objects of studying the pure model materials with initial sink of point defects. The material modelling a role of surfaces is the sample with perfect crystal structure - single crystal titanium.

The existence of deuterium segregation induced by ion implantation in titanium at room temperature was determined for the cases of irradiation of titanium target by deuteron of low and high energies [2]. In the first case the nuclear reaction of deuterium atoms with 'He' ions was used to...
measure the concentration of implanted deuterium. In the second case the radiation-induced segregation (RIS) of deuterium was created and analyzed using one and the same beam of deuterons with the energy of 700 keV with the help of nuclear microanalysis method NRA. The measurements were carried out in situ using reaction $D(d,p)T$, where $p$ is the protons, $d$ is the deuterons, $T$ and $D$ are the atoms of tritium and deuterium correspondingly.

It is well known that accumulation and distribution of implanted deuterium in metals depends on the existence of effective sinks of point defects and their distribution in target and also on the existence, distribution and capacity of hydrogen traps [3-5]. The surface of sample is the only effective sink in single crystal target. To study the formation and evolution of RIS such target presents a real interest since it helps to minimize a number of uncontrolled factors during the experiment.

The aim of the present work is to study the formation and stability of segregation of deuterium in single crystal titanium implanted by deuterium ions at room temperature.

1. Experimental

2.1. Methods
The work was carried out on the nuclear-physics accelerator complex of the Institute of Metal Physics of the Ural Division of RAS using the reaction $D(d,p)T$ with deuteron energy of 700 keV. The use of deuteron beam as a probe gave the opportunity to measure the deuterium concentration during implantation and simultaneously to control the composition of the analyzed zone regarding oxygen and nitrogen (according to reaction $D(^{16}O,p)\,^{17}O$ and $D(^{14}N,\alpha)^{12}C$). The choice of reaction $D(d,p)T$ ensured the coincidence in time and space of implantation processes and measurement of concentration of implanted deuterium in the irradiated volume. Besides, it ensured experimental purity of the introduction of additional component to the target (helium) and excluded the appearance of ion-induced damages beyond the irradiation zone.

The content of deuterium was determined from the energy spectrum of nuclear reaction products – protons. The sample of titanium with constant concentration of deuterium $TiD_{1.78}$ served as a standard sample. The value of RIS was estimated according to the average concentration of implanted deuterium in the irradiated region ($C_D$). The depth of analysis was conditioned by the length of projective range of deuterons (~ $4.7 \times 10^{-6}$ m) and by depth resolution of the used nuclear reaction (~ $0.5 \times 10^{-6}$ m). The analyzable volume was $4 \times 10^{-8}$ m$^3$.

Besides the $C_D$ measured discretely during the continuous deuterium implantation, $\alpha$ trapping of implanted deuterium of RIS was calculated as a fraction of the whole implanted deuterium trapped in the irradiated volume. The total of deuterium amount trapped all kinds of traps was calculated

$$\alpha_0 = C_D/C_D^{\text{theor}} \times 100\%,$$

where $C_D^{\text{theor}}$ is the average concentration of the implanted deuterium under the condition of its complete conservation in the irradiated volume.

The stability of deuterium segregation in titanium after irradiation was also studied. For that the beam of deuterons was shut off at different stages of ion irradiation. The duration of pauses in the process of implantation was from $1.8 \times 10^3$ c to $245.4 \times 10^3$ c. After the end of shutting the process of segregation measurement was continued.

2.1. Materials
The work was carried out with single crystals $\alpha$-Ti [6] made by electron-beam zone melting of titanium with the content of iron and silicon to 0.12 and 0.08 mass % correspondingly. The samples were cut of the cylinder single crystal (length 0.1 m and diameter 0.01 m) in such a way as to make the surface of sample perpendicular to the $c$–axis. The deformed in the process of cutting layer of crystal was removed by electro polishing. The content of oxygen in the samples was 0.05 at. %, the content of nitrogen, hydrogen and carbon was by the order lower.
Single crystals contain grains characterized 300-500 μm in size and density of dislocations less than $10^{11}$ m$^{-2}$. The neighboring blocks disorientation been not more than 0.1 degree, microstresses were absent.

3. Results and discussion

The results of measurements of dose dependence of the average content of deuterium and calculation of the corresponding trapping of deuterium in the analyzable zone during continuous irradiation and after the interruptions of deuterons beam are illustrated by figure 1 and table 1.

![Figure 1: Effect of total implantation dose $F$ on the average deuterium content in the analyzable zone $C_D$ and on the fraction of implanted deuterium $\alpha$, preserved in the analyzable zone. Dashed line and numbers show the stops 0, 1, 2, 3, 4 and 5. Duration stops see Table 1.](image)

**Figure 1.** Effect of total implantation dose $F$ on the average deuterium content in the analyzable zone $C_D$ and on the fraction of implanted deuterium $\alpha$, preserved in the analyzable zone. Dashed line and numbers show the stops 0, 1, 2, 3, 4 and 5. Duration stops see Table 1.

| Stops of irradiation | The dose of continuous exposure to the stop of radiation, $10^{20}$ m$^{-2}$ | The total dose of radiation, $10^{20}$ m$^{-2}$ | The duration of the pause, $10^3$ c | Q, appm |
|----------------------|-----------------------------------------------|-----------------------------------------------|-----------------------------------|--------|
| 0                    | 0                                              | 0                                             | 0                                 | 0      |
| 1                    | 15                                             | 15                                            | 1.8                               | 47     |
| 2                    | 2.6                                            | 21.6                                          | 13.2                              | 77     |
| 3                    | 6                                               | 31.6                                          | 245.4                             | 189    |
| 4                    | 40                                              | 73.6                                          | 77.4                              | 362    |
| 5                    | 20                                              | 95.6                                          | 2.6                               | 382    |

As it was expected the $C_D(F)$ increases in the parts of continuous growth of implantation doses. Long-term stops of irradiation lead to the jumping growth of this dependence.

The first part of measurement of $\alpha(F)$ deuterium trapping at continuous irradiation is italicized (Figure 1) with solid line which qualitatively coincides with the dose dependence of the implanted deuterium trapping into in-cascade clusters in the alloyed austenite without initial traps [3]. It gives us a ground to suppose that in titanium single crystal there are no initial deuterium traps or their capacity is extremely small. The part of $\alpha(F)$ dose dependence at continuous irradiation with total doses from $\sim32 \times 10^{20}$ m$^{-2}$ to $\sim74 \times 10^{20}$ m$^{-2}$ has more complicated character. In spite of the fact that $C_D(F)$
increases with the growth of irradiation dose a remarkable decrease implanted deuterium trapping in it is observed.

It is known [3] that trapping of deuterium during irradiation occurs into initial traps IT (existed before the beginning of irradiation) and radiation induced traps RIT, i.e. \( \alpha(F) = \alpha_{IT}(F) + \alpha_{RIT}(F) \).

Before overlapping of a scope of radiation traps \( \alpha_{RIT}(F) \sim F \). After filling of initial traps \( \alpha_{IT}(F) \sim 1/F \). The descending character of \( \alpha(F) \) dependencies between the stops speaks about the appearance of a large amount of deuterium traps and their filling during the stops 3 and 4. For the resumed irradiation such traps play a part of the initial ones, i.e. existed before this irradiation.

Considering the renewal of irradiation as an independent experiment with preliminary created defect structure of the sample we obtained \( \alpha(F^*) \) dependencies, where \( F^* \) is the dose of continuous irradiation (Figure 2). To determine the capacity of traps \( Q \) for each subsequent radiation, formed during the previous irradiation and stop time necessary to compare the initial portions of descending dependencies \( \alpha(F^*) \). It is here \( \alpha_{IT}(F^*) = Q / C_{D}^{emp}(F^*) \times 100\% \). Capacities of initial deuterium trap by the method described in [3] are after stops 0-5 are given in Table 1.

![Figure 2. Dose dependencies of deuterium traps in the initial titanium (0) and after stops 1-5 (see figure 1 and Table 1).](image)

Distribution of implanted deuterium without taking into account its diffusion mobility and also the mobility of radiation vacancies and SIs was calculated using SRIM program. It has a pronounced maximum near the depth \( 4.7 \times 10^{-6} \) m which is equal to the length of projective range of deuterons.

The obtained experimentally dependencies of concentration of the implanted deuterium on the depth of analysis differ from the calculated profiles. They are more diffused and the concentration of deuterium in the irradiated volume is rather less than the calculated one.

However, in spite of the low depth resolution of the used nuclear reaction the profile analysis of deuterium in titanium during the continuous irradiation showed the existence of the maximum of deuterium content near \( \sim 4.5 \times 10^{-6} \) m (Figure 3). The interruptions of irradiation resulted in the qualitative change of depth profiles that testifies first of all to the evolution of defect structure of the target during interruption and appearance of the “new” filled IT.

The depth dependencies of deuterium concentration in the irradiated target volume gave us the opportunity to detect the location of the “new” IT. If the continuous irradiation forms the distribution of deuterium with the maximum on the depth \( \sim 4.5 \times 10^{-6} \) m then the long-term stop of irradiation leads to the considerable redistribution of deuterium toward the surface of sample (Figure 3b).

The increase of implantation dose before the first beam shut off resulted in the growth of amount of implant along the whole analyzable zone but mainly in the region if this maximum (curves 1 and 2 in Figure 3a). At the time of \( 1.8 \times 10^{-6} \) c stop after implantation with the dose \( 15 \times 10^{20} \) m\(^{-2}\) the character of \( C_{D}(F) \) dependence did not change and the view of depth distribution of implant remained.
Figure 3. Distributions of deuterium on the depth of titanium sample before the first long-term stop (curves 1-3) and after interruption of irradiation for $2.45 \times 10^3$ c (curve 4). Measurements were carried out at irradiation doses: 1 - $7 \times 10^{20}$ m$^2$; 2 - $15 \times 10^{20}$ m$^2$; 3 - $28 \times 10^{20}$ m$^2$; 4 - $32 \times 10^{20}$ m$^2$.

4. Conclusions
The investigation of accumulation of implanted deuterium in the irradiated volumes of titanium by nuclear reaction method made it possible to observe in situ the development of defect structure of target.

The effect of accumulation of implanted deuterium near the sample surface during the isothermal aging of irradiated titanium single crystal at room temperature was determined for the first time.

The detected peculiarity in behavior of the implanted deuterium is explained by the evolution of defect structure at room temperature and the appearance of traps of large capacity.

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