Nuclear Inst, and Methods in Physics Research B 414 (2018) 166–169

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Reprint of: Effects of cold deformation, electron irradiation and extrusion on deuterium desorption behavior in Zr-1%Nb alloy☆

O. Morozov*, O. Mats, V. Mats, V. Zhurba, P. Khaimovich

National Science Center "Kharkiv Institute of Physics & Technology", 1 Akademicheskaya Str., Kharkiv 61108-UA, Ukraine

ARTICLE INFO

Peer-review under responsibility of the Scientific Committee of the Conference on the Application of Accelerators in Research and Industry.

Keywords:
Zr-1% Nb alloy
Cold deformation
Electron irradiations
Extrusion
Nanostructure
Thermal desorption spectroscopy (TDS)
TEM
Deuterium desorption

ABSTRACT

The present article introduces the data of analysis of ranges of ion-implanted deuterium desorption from Zr-1% Nb alloy. The samples studied underwent plastic deformation, low temperature extrusion and electron irradiation. Plastic rolling of the samples at temperature ~300 K resulted in plastic deformation with the degree of $\varepsilon = 3.9$ and the formation of nanostructural state with the average grain size of $\mathbf{d} = 61$ nm. The high degree of defectiveness is shown in thermodesorption spectrum as an additional area of the deuterium desorption in the temperature ranges 650–850 K. The further processing of the sample (that had undergone plastic deformation by plastic rolling) with electron irradiation resulted in the reduction of the average grain size (58 nm) and an increase in borders concentration. As a result the amount of deuterium desorpted increased in the temperature ranges 650–900 K. In case of Zr-1% Nb samples deformed by extrusion the extension of desorption area is observed towards the temperature reduction down to 420 K. The formation of the phase state of deuterium solid solution in zirconium was not observed. The structural state behavior is a control factor in the process of deuterium thermodesorption spectrum structure formation with a fixed implanted deuterium dose (hydrogen diagnostics). It appears as additional temperature ranges of deuterium desorption depending on the type, character and defect content.

1. Introduction

Zirconium alloys present a unique combination of nuclear-physical, mechanical and corrosion properties, namely, a very-low neutron absorption, high corrosion resistance, high strength and plasticity. The combination of such properties promotes the use of zirconium and its alloys in the reactor engineering. Thus, studying the irradiation-induced effect on the structural state of alloys is of scientific and practical interest [1].

Introduction of dislocations into the metal crystalline lattice has found to decrease the radiation effect on metals and alloys. The most commonly used method of defect introduction into a metal crystalline lattice is through plastic deformation. The intense plastic deformation at low homologous temperatures ($T < 0.2T_M$, T_M is the melting temperature) forms a wide spectrum of distorted structures. It is known that the lower plastic deformation temperature, the higher defect density can be introduced by this treatment method since the dynamical recovery processes are suppressed at lower temperatures. On the basis of these considerations, the researchers of the NSC KIPT have developed a technique for plastic deformation of metals and alloys, namely, the low-temperature extrusion at extrusion temperature of ~ 78 K and lower. First the efficiency of this method was confirmed by experiments on pure metals, in particular, on copper [2] which has shown a capacity for elastic deformation at room temperature up to the pressures 700–720 MPa. Further investigations were carried out on the austenite stainless steel Cr18N10 T at cryogenic temperatures under conditions close to the high-pressure hydrostatic compression (quasi-hydrostatic extrusion). It has been found that as a result of deformation a high-disperse martensite is formed in the steel and its mechanical characteristics are appreciably improved [2].

The method of thermal desorption spectroscopy (TDS) is usually used to determine the temperature ranges of gas retention in metals and has been applied for investigations of structural states in a wide range of materials. The processing of spectra obtained for the thermoactivated hydrogen release permits to calculate the thermodynamic desorption parameters (desorption activation energy, order of reaction etc.). Early experiments [3,4] studied the correlation between the peaks in the thermo-desorption spectra of deuterium from Pd and austenitic stainless stell and the phase transformations in the metal-hydrogen system. Furthermore, it has been shown that the hydrogen release spectrum

☆ This article is a reprint of a previously published article. For citation purposes, please use the original publication details; Nuclear Inst. and Methods in Physics Research, B Volume 410, November 2017, Pages 37–40.

* Corresponding author.
E-mail address: morozov@kipt.kharkov.ua (O. Morozov).

https://doi.org/10.1016/j.nimb.2017.11.006
Received 28 June 2017; Received in revised form 22 July 2017; Accepted 24 July 2017
Available online 10 November 2017
0168-583X/

depends on the type and degree of structure-changing effect, implying the usefulness of TDS for "hydrogen diagnostics" of the structural state of materials. At this stage of method developing the results of various actions on the structural changes in materials were qualitatively evaluated.

The present paper reports the results of investigations on the temperature ranges of ion-implanted deuterium desorption from the Zr-1% Nb zirconium alloy in different structural states: after the melt crystallization; nanostructuring with deformation by plastic rolling at room temperature; electron irradiation of samples pre-deformed by plastic deformation; and after low-temperature extrusion at extrusion temperature of ~78 K.

2. Experimental

Desorption temperature ranges of the ion-implanted deuterium from Zr-1%Nb samples upon different structure-changing treatments were investigated by the TDS using the facility "Skif" [5]. The samples of Zr-1%Nb alloy were investigated in the initial (cast) state; after deformation by plastic rolling at 300 K with a degree of compression $\varepsilon = 3.9$; after irradiation with 10 MeV electrons at a fluence of $\sim 6 \times 10^{17}$ cm^{-2} and a temperature of < 370 K; and after low-temperature extrusion by 15% at extrusion temperature of ~78 K [2].The samples were mounted onto the zirconium heater of $40 \times 5 \times 0.3$ mm^3. The temperature was measured with the tungsten-rhenium thermocouple W5Re/W20Re fastened on the sample. To decrease the influence of background hydrogen contained in the samples and the target chamber, deuterium (a hydrogen isotope) was used in the experiment. Deuterium was implanted into the samples cooled to the temperature of ~100 K by a 24 keV D_2^+ ion beam. After the implantation of a given deuterium fluence, the beam was switched off and the sample heating was started. The sample temperature was elevated to approximately 1600 K using an average heating rate of ~3.5 K/s. Deuterium release in the measuring chamber was recorded using the mass-spectrometer for m = 4 amu (D_2^+). The sample microstructure was investigated using the transmission electron microscope (TEM) under the accelerating voltage of 100 kV.

3. Results and discussion

The characteristic thermodesorption spectra of deuterium implanted into the initial Zr-1%Nb samples by different deuterium ion doses are shown in Fig. 1. Deuterium thermodesorption spectra represent the sample structural states and their changes as a function of the implanted deuterium dose. Implantation of low deuterium doses results in formation of a deuterium solid-solution phase state in the zirconium alloy that appears in the thermal desorption spectrum as two peaks with maximum temperatures approximately 1080 K and 1400 K (Fig. 1, curve **a**). Such a spectrum pattern is explained by the presence of the phase transition **α**-Zr ⇒ **β**-Zr in zirconium. A peak with a maximum temperature of ~1080 K is formed due to the decomposition of the deuterium solid solution phase state in **α**-Zr. Almost simultaneously with beginning of this phase state decomposition in zirconium the phase transition **α**-Zr ⇒ **β**-Zr occurs. In the spectrum we can see that the deuterium desorption decreases and the phase state of deuterium solid solution in **β**-Zr is formed. At a temperature near 1400 K the phase state of deuterium solid solution in **β**-Zr is decomposed. The increase in implanted deuterium dose stimulates the zirconium hydride formation that is evidenced by the peak intensity increase in the deuterium thermodesorption spectrum with a maximum temperature ~900 K (Fig. 1, curves **b** and **c**). Note that according to the Zr-H system phase diagram [6] the temperature of zirconium hydride decomposition practically coincides exactly with the temperature maximum of a corresponding peak in the deuterium thermodesorption spectrum. For further investigations of samples deformed with the use of different structure-changing treatments a testing dose of 1.3×10^{17} D/cm^2 (hydrogen diagnostics) was chosen [7,8].

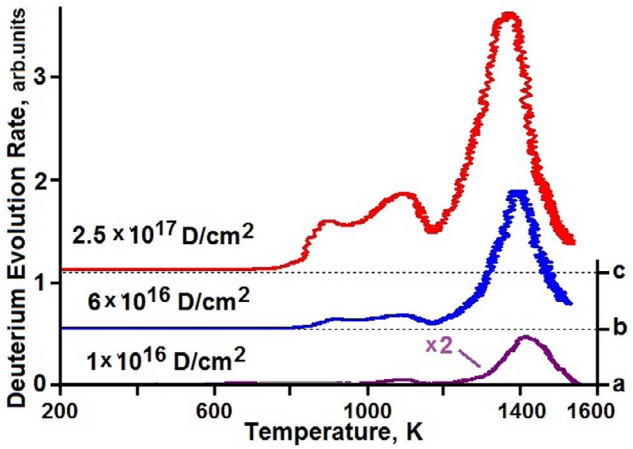

Fig. 1. Thermodesorption spectra of deuterium implanted into the zirconium alloy with Zr-1%Nb doses: (a) ~1×10^{16} D/cm^2; (b) ~6×10^{16} D/cm^2; (c) ~2.5×10^{17} D/cm^2.

The plastic deformation of Zr-1%Nb alloy performed by rolling at 300 K to the degree $\varepsilon = 3.9$ has created a nanostructured state with an average grain size of 61 nm. The chosen temperature-force mode of rolling has provided favorable conditions for the creation of uniformly distributed structure submicroinhomogeneities with formation of an optimum boundary ensemble configuration. In such a nanostructure a high volume concentration (3.4%) of grain boundaries and a high microstress level are observed (Fig. 2b). Probably, due to the high degree of defect content there is a low-intense deuterium desorption region, extended on the temperature scale in the range 700–1000 K, in the ion-implanted deuterium thermodesorption spectrum (see Fig. 3, curve b). Note that in this case the deuterium desorption begins at room temperature.

In the Zr-1%Nb sample, subjected to the plastic deformation and subsequently exposed to the electron irradiation, the new polygonal boundaries are formed, the average grain size decreases (**d** = 58 mm), the grain boundary concentration increases (~8.8%), and the dislocation density in the grain body increases (see Fig. 2c). Apparently, an intense dislocation displacement occurred in the peaks of broken boundaries and near-boundary zones i.e. places of the maximum stress concentrations. Besides, the microslip and climb of dislocations caused by their interaction with directed flows of radiation-origin point defects also occurred. In the thermodesorption spectrum of ion-implanted deuterium, such an effect becomes apparent with the occurrence of an additional desorption temperature range of 700–800 K (Fig. 3, curve **c**).

The investigation of Zr-1%Nb samples deformed by extrusion at 78 K with percent reduction $\delta = 15\%$ discovered the presence of a deuterium thermodesorption region, widely extended on the temperature scale, in the temperature range of 420–850 K and low-intense high-temperature deuterium desorption regions with peak temperatures of 1100 and 1400 K (Fig. 3, curve d). It should be noted that the most important factor accompanying the extrusion at 78 K is the presence of uniform compression forces simultaneously with deformation temperature decreasing in the region of cryogenic temperatures. This factor inhibits the microcracking, creates the precondition for a high fragmentation of the defect structure being formed and stimulates the formation in the material a high interface concentration. Thus in the material a high defect density is reached without tendency to the brittle fracture, and there formed are the high-regularity defect structures not observed in the case of deformation at room temperature or higher temperatures [2]. An important feature of deuterium thermodesorption spectra for Zr-1%Nb samples deformed by extrusion at 78 K is, practically, a total lack of deuterium desorption in the regions with peak temperatures of 1100 and 1400 K (Fig. 3, curve d), the presence and intensity of which evidence on the formation of a deuterium solid-