

Effect of Thermal Exposure on Structure of the Ultrafine-Grained Zr-1Nb Alloy

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Abstract. Effect of annealing at temperature range of 573–823 K on stability of the ultrafine-grained structure of the Zr-1wt.%Nb alloy was studied by methods of transmission electron microscopy. Growth kinetics of grain–subgrain structure elements of alloy was investigated.

1. Introduction

An effective method of improving the strength and operational characteristics of metal polycrystals at low homologous temperatures is grain refinement to submicron dimensions (grain size $d < 1 \mu\text{m}$) [1–3]. All most widely spread methods of the ultrafine-grained (UFG) state formation in metallic materials are based on Severe Plastic Deformation (SPD). SPD methods allow obtaining the UFG state in volume billets, which is a structure of grain type (size of grain–subgrain structure elements 0.05–0.5 μm) and contains predominantly high-angle grain boundaries [1, 4]. However, UFG state formed in metallic materials by SPD methods has a high density of lattice and grain boundary dislocations, elastic lattice distortions and long-distance stress fields and, as a consequence, is metastable. One of the possible ways of increase in stability of the UFG state and its mechanical properties is formation of internal structure in alloys. Such structure combined two and/or some structural elements (or phases) with different dispersion is obtained by means of directing use of structural and phase transformations initiated by SPD, as well as subsequent thermal treatments [5, 6]. In this connection, study of development features of such process, as recrystallization, phase transformations, solid solution decay, and phase coalescence during UFG state forming by SPD methods and subsequent thermal treatments is of great relevance.

On the basis of the above reasoning, a purpose of this work was experimental investigation of development features of structural and phase transformations at the UFG Zr-1wt.%Nb alloy (further Zr-1Nb) during annealing and electron beam irradiation at the temperature range of 573–873 K.

2. Experimental procedure

Commercial zirconium Zr-1Nb alloy (E110 grade) was used as an initial material for study in this work. Previously, it was found [3] that used Zr-1Nb alloy in as-received condition has polycrystalline structure with the grain size of 3–5 μm . X-ray structural analysis indicates that the alloy contains not only the main α -Zr phase, but also small amount of β -Zr phase (total volume fraction $\sim 2.5 \text{ vol. } \%$), β -Nb (solid solution of Zr on the base of Nb) and solid solution of Nb in β -Zr phase. Secondary phases



in the form of particles, which dimensions vary from a few tens of nanometers to several microns, are present in the bulk and at grain boundaries of alloy.

UFG structure in alloy was formed using one of the SPD methods: pressing with the change of deformation axis and gradual temperature decrease at the interval of 973–623 K. Investigation of thermal stability of the Zr-1Nb alloy UFG structure was carried out by means of annealing at the temperature range of 573–873 K, including annealing under constant electron beam exposure. Energy of electron beam was 35 keV, current density was equal to 75 and 110 μA , and time of irradiation was 1 hour.

Alloy structure was investigated with the use of JEM-2100 transmission electron microscope. The size of structure elements of the UFG alloy was measured from the relevant dark-field image micrographs by means of the secant method. Data sample was not less than 200 elements. The volume fractions and lattice parameters of the phases were determined with accuracies of $\pm 1\%$ and 0.0001 nm, respectively, using a Shimadzu XRD7000 diffractometer with $\text{Cu-K}\alpha$ radiation source. PowderCell program was used during identification of diffraction patterns.

3. Results and discussion

The typical electron micrographs of the UFG structure of the Zr-1Nb alloy after SPD is shown in Fig. 1. It is evident (Fig. 1, a) that an entangled deformational contrast is observed and some structure elements are poorly distinguished in the bright-field image of the structure. Electron diffraction patterns of such structure obtained for an area of $1.4 \mu\text{m}^2$ show diffraction rings formed by the reflections from individual crystallites (Fig. 1, a). At the same time almost all of the reflections exhibit azimuthal smearing. Such kind of electron diffraction patterns is characteristic for the UFG state obtained by SPD method and indicates high-angle misorientations of the structure elements and presence of elastic stresses at the individual grains [1, 4]. Grain–subgrain structure elements, which dimensions vary between 0.1 and 0.5 μm , are clearly distinguished in the dark-field image (Fig. 1, b). The average size of grain–subgrain structure elements determined from the dark-field image is $(0.3 \pm 0.12) \mu\text{m}$. Obtained UFG structure contains only $\alpha\text{-Zr}$ and $\beta\text{-Nb}$ phases. Volume fraction of the $\beta\text{-Nb}$ phase does not exceed 0.5 vol. %. This phase is observed in the form of particles at the grain boundaries and volume of main $\alpha\text{-Zr}$ phase. This is indicative that under mentioned conditions of SPD process partial dissolution of secondary phases and formation of supersaturated solid solution occur in the Zr-1Nb alloy.

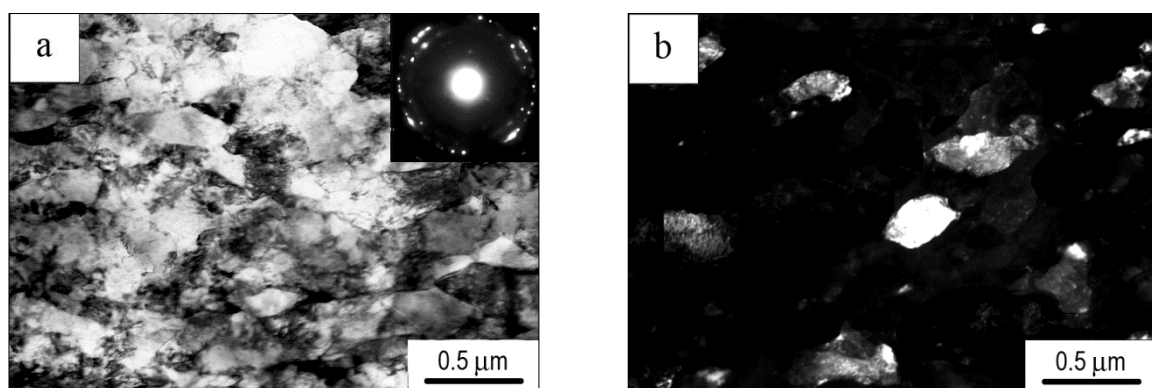


Figure 1. Electron microscope image of the UFG structure of Zr-1Nb alloy: (a) bright-field image and microdiffraction pattern, (b) dark-field image.

Electron microscopic studies of Zr-1Nb alloy microstructure after hour annealing at the temperature range of 573–873 K showed that UFG structure of alloy is stable up to a temperature of 723 K. However during annealing at the temperature range of 573–673 K relieving of defect UFG structure is observed: dislocation density at the grain volume decreases and boundaries of individual grains reveal. At the same time the fringe contrast is detected at some grain boundaries that is inherent

in the equilibrium state of grain boundaries (Fig. 2, a). After annealing at temperature of 723 K individual recrystallized grains with dimensions of 1–2 μm appear in structure (Fig. 2, b). Density of such grains is low, so the average size of the structural elements of the alloy found from the dark-field image increases slightly.

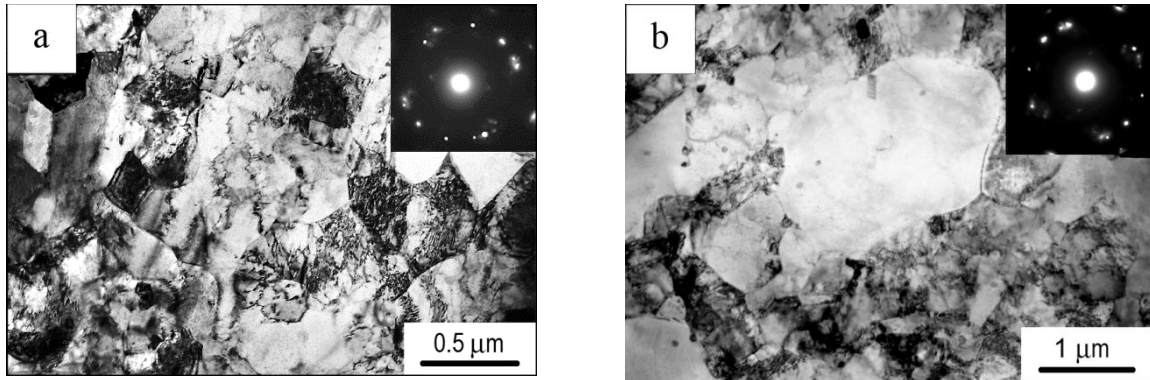


Figure 2. Electron microscope image and microdiffraction pattern of the UFG structure of Zr-1Nb alloy: (a) structure after annealing at 673 K for 1 h, (b) structure after annealing at 723 K for 1 h.

After annealing at temperature of 773 K recrystallization and grain growth are observed in the whole volume of samples. The average grain size increases up to 1.2 μm . At the same time during annealing at temperature of 873 K for 1 h collective recrystallization is actively developing and average grain size becomes equal to 2.5 μm (Fig. 3, b).

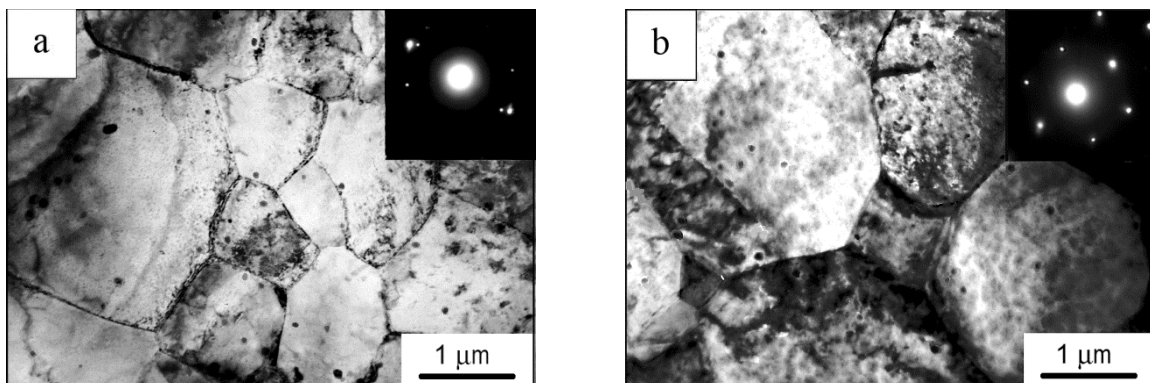


Figure 3. Electron microscope image and microdiffraction pattern of the UFG structure of Zr-1Nb alloy: (a) structure after annealing at 873 K for 1 h, (b) structure after annealing at 723 K for 1 h under the electron beam irradiation

Stability of the UFG Zr-1Nb alloy reduces when annealing is carried out under conditions of electron beam irradiation. In this case growth of structure elements is observed throughout the sample volume already at 723 K. At the same time average element size has increased from 0.3 to 1.5 μm for 1 hour of annealing (Fig. 2, b). Recrystallization and grain growth are accompanied by precipitation of nano-sized particles of the β -Nb phase at the UFG Zr-1Nb alloy at temperatures of 723 K and above (Fig. 2, b and 3). Volume fraction of the β -Nb phase increases up to 1–1.5 %.

Annealing at temperature of 753 K for 2, 3, 5, 7 and 9 hours were performed to study growth kinetics of average size of grain–subgrain structure elements during annealing of investigated alloy. Kinetic curve of growth of grain–subgrain UFG structure elements of Zr-1Nb alloy at temperature of 753 K is represented at the Fig. 4. It is seen that there are two stages on this curve: with high and low rate of the structure element growth. High rate of structure element growth is observed at the first hours of annealing (2–4 hours). Then the rate of the structure element growth significantly slows down.

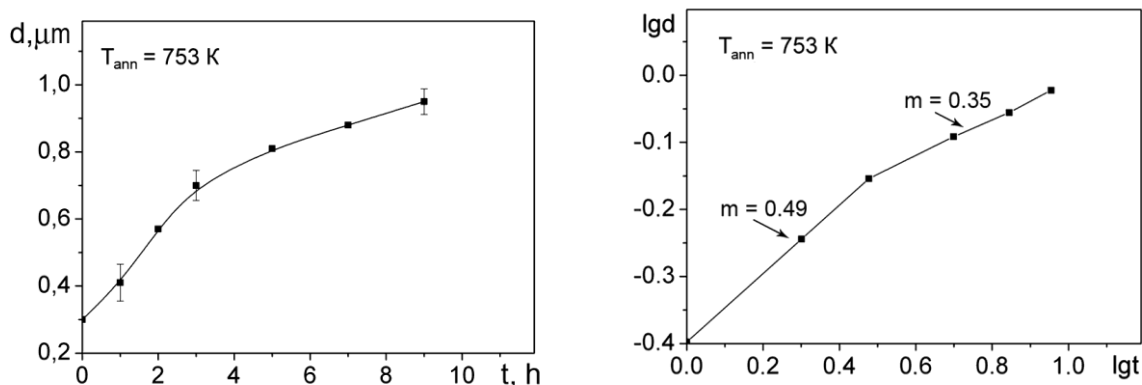


Figure 4. Kinetic curve (a) and logarithmic dependence (b) of change in the average size of the grain–subgrain UFG structure elements of the Zr-1Nb alloy during annealing at temperature of 753 K.

It is known [7] that depending on the development of the recrystallization process grain growth kinetics in materials with the UFG structure can be described by one of the following equations:

$$d = Kt^m \quad (1)$$

$$d = d_0 + Kt \quad (2)$$

where d – average grain size, K – constant factor, m – exponent of grain growth characterized dependence of grain growth on time at given temperature.

For the majority of fine metals grain growth during recrystallization is described by the equation of type (1) with m equal to 0.5. Rearrangement of d – t dependence for investigated zirconium alloy in logarithmical coordinates showed that kinetics of UFG structure element growth is described by an equation of type (1) on the both stages (Fig. 4, b). However, on the first stage the m value in the equation (1) is equal to 0.49, and on the second one – 0.35 (Fig. 4, b). Value $m = 0.49$ on the first stage of the UFG structure element growth for investigated heterophase Zr-1Nb alloy is closed to the m value for the fine metals. Apparently, this is due to a small volume fraction of the β -Nb phase particles, which are able to inhibit migration of grain boundaries. (It was noted above that content of secondary phases decreases to 0.5 vol. % in the process of SPD). With increasing of annealing time, solid solution decay and precipitation of nano-sized particles of the β -Nb phase occur in the Zr-1Nb alloy. These processes prevent grain boundary migration, so the rate of recrystallization process slows down.

4. Conclusion

Partial dissolution of secondary phases and formation of supersaturated solid solution are observed during UFG structure obtaining at the heterophase Zr-1Nb alloy with the use of pressing with the change of deformation axis and gradual temperature decrease at the range of 973–623 K. Hour annealing at the temperature range of 723–873 K result in growth of UFG structure elements and decay of supersaturated solid solution with the precipitation of nano-sized particles of the β -Nb phase. Growth kinetics of the UFG structure elements has double-stage character and is described by power dependence on the both stages. Slowing down the rate of element growth of the UFG grain-subgrain structure at the second stage is connected with precipitation of nano-sized particles of the β -Nb phase prevented grain boundary migration.

Acknowledgements

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