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Effect of ultrasonic impact treatment on creep characteristics and evolution of Zr1Nb alloy nanostructure

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Effect of ultrasonic impact treatment on creep characteristics and evolution of Zr1Nb alloy nanostructure has been studied. Under the ultrasonic impact treatment the level of internal stresses in the nanostructured material decreases due to formation of the more equilibrium boundary structure without appreciable grain growth. Subsequent deformation of the nanostructured Zr1Nb alloy under creep conditions at 700 K induces the relaxation of internal stresses as a consequence of dynamic recrystallization. As a result, the new stable nanostructure is formed that appreciably increases thermomechanical stability of the alloy.

Keywords: Zr1Nb alloy, nanostructure, creep, ultrasonic impact, structural instability.

Изучено влияние ультразвукового воздействия на характеристики ползучести и эволюцию наноструктуры сплава Zr1Nb. Показано, что ультразвуковая обработка приводит к снижению уровня внутренних напряжений наноструктурированного материала за счет формирования более равновесной структуры границ без заметного роста зерен. В процессе последующей деформации в условиях ползучести при 700 К наноструктурированного сплава Zr1Nb, подвергнутого ультразвуковому воздействию, происходит релаксация внутренних напряжений вследствие динамической рекристаллизации и образование новой стабильной наноструктуры, что приводит к заметному повышению термомеханической устойчивости.

Вплив ультразвукової обробки на характеристики повзучості та еволюцію наноструктури сплаву Zr1Nb. *В.І.Соколенко, В.М.Горбатенко, Є.В.Карасьова, О.В.Мац, Є.С.Савчук, В.О.Фролов.*

Вивчено вплив ультразвукової обробки на характеристики повзучості та еволюцію наноструктури сплаву Zr1Nb. Показано, що ультразвукова обробка призводить до зниження рівня внутрішніх напружень наноструктурованого матеріалу за рахунок формування більш рівноважної структури границь без помітного зростання зерен. У процесі подальшої деформації в умовах повзучості при 700 К наноструктурованого сплаву Zr1Nb, підданого ультразвуковому впливу, відбувається релаксація внутрішніх напружень внаслідок динамічної рекристалізації і утворення нової стабільної наноструктури, що призводить до помітного підвищення термомеханічної стійкості.

1. Introduction

The method based on the usage of severe plastic deformation (SPD) is one of the most promising ways of producing nanocrystalline metallic materials [1–4]. Nanocrystalline materials with a characteristic grain size less than 100 nm, obtained by the SPD method, have extraordinary functional and mechanical characteristics, namely: high strength, wear resistance, hardness, high fatigue properties etc. [1, 2]. They differ from the macrocrystalline materials, besides the grain size, by some structural features such as high density of grain-boundary dislocations, high concentration of non-equilibrium deformation vacancies, high level of internal stresses and others. Because of the high internal stresses, induced by the non-equilibrium grain boundaries, such materials are characterized by low plasticity and thermal stability [1–4]. Therefore, in order to obtain a complex of high mechanical properties in the nanostructured materials it is necessary to select an appropriate non-destructive processing method.

Among a variety of approaches the use of ultrasonic impact treatment (UIT) for improvement of the nanostructure material properties is one of the most powerful tools. Ultrasonic waves propagating in the material, interacts with various defects: vacancies, dislocations, grain and subgrain boundaries, impurities, and cause the structural changes which depend on the parameters of ultrasound, mainly on its power. By varying the ultrasonic treatment conditions for the material under study it is possible to form the structure providing the attainment of desired properties. The low-intense ultrasonic impact on the deformation nanostructure decreases and equalizes the internal stresses in the material volume while the structure size factor remains constant and structure homogeneity increases [5,6].

Further study of ultrasonic influence on the nonequilibrium structure of the nanocrysrtalline metallic materials, in order to open up the new potentialities of ultrasonic treatment for improving the structure and properties of these materials, is an urgent and important task for practical work.

2. Учзукшьутефд

Zr1Nb alloy prepared by the method of electron-beam melting has been investigated. In order to reveal the influence on the structure and properties of the Zr1Nb alloy the following treatment procedures were used:

1. MT-1 — combined rolling at 77 K–300 K, finite deformation (ε) was 3.9;
2. MT-2 — MT-1 + UIT at 300 K;
3. MTT-1 — MT-1 + annealing at 870 K for 1 h.

Defect structure of the Zr1Nb alloy was investigated by the method of electrical resistance measurement after each SPD treatment. Electrical resistance (R) was measured by the four-point scheme using the compensation method. The error of R measurements did not exceeded ±0.5 %. The structure evolution control was carried out using the electron microscopic method.

Creep tests were conducted in the step loading mode at temperatures 300 K and 700 K that corresponds to the reactor operating temperature. The elongation measurement accuracy was ~ $5 \cdot 10^{-5}$ cm.

A part of the rolled samples were subjected to UIT ($f = 20$ kHz) at $T = 300$ K using the technique described in [5]. The amplitude of ultrasonic tangential stresses was 80 MPa, the UIT duration was 10 min. Selected mode of preliminary ultrasonic treatment [5, 7] exerts a softening effect on the deformed material.

3. Results and discussion

In order to reduce the internal stress level and to increase the plasticity of the nanostructured Zr1Nb alloy we have carried out two treatment procedures: ultrasonic impact treatment (MT-2) and, for comparison, standard thermal treatment (MTT-1), at temperature of the corresponding stage of primary recrystallization of the material being studied which, as our previous experiments [8] have shown, leads to the appreciable increase of plasticity.

By measuring the residual electrical resistance ($R_{300\ \mathrm{K}}/R_{77\ \mathrm{K}}$) it is possible to control the changes in the material defect state induced by deformation after rolling and different treatments. Results of $R_{300\ \mathrm{K}}/R_{77\ \mathrm{K}}$ measurement are given in Table.

It is evident from the table that both the treatments of the rolled Zr1Nb alloy (MT-2 and MTT-1) increase the residual electrical resistance i.e. reduce the internal stresses level.

The creep characteristics of the Zr1Nb alloy subjected to different treatments were investigated within the temperature range from 300 K to 700 K. The creep rates of the Zr1Nb samples, subjected to different treatments within the stress range being studied at the test temperature of 700 K (reactor operating temperature) are presented in Fig. 1.

Table. Residual electrical resistance of nanostructured Zr1Nb alloy after different treatments

Measured characteristics	Treatment type		
	MT-1	MT-2	MTT-1
$R_{300\ \mathrm{K}}/R_{77\ \mathrm{K}}$	3.03	3.47	3.57

Analysis of the experimental results show that the rolled ($\varepsilon \approx 3.9$) samples tested at 700 K have the high strength and, at the same time, the low plasticity and creep rate because of the high level of internal stresses. After the annealing (MTT-1) the ultimate stress of the samples is twice less than that of the rolled samples (MT-1) and the plasticity increases by 20 %. The ultrasonic treatment (MT-2) also decreases the strength properties of the material by ~ 15 % in comparison with the rolled samples, and its plasticity increases by ≈ 16 %.

Our previous experiments [9, 11] have shown that the plastic flow of submicro- and nanostructured materials, obtained by the SPD method, is conditioned by the joint action of several mechanisms: cross sliding, climb and annihilation of dislocations near the grain boundaries, as well as, diffusion creep and grain-boundary dislocation sliding along the grain boundaries. The contribution from each of these mechanisms into the material deformation depends on the test temperature, applied stress and grain boundary state.

It is important to note that the nanomaterials specificity, in the view of the defect theory, consists in the fact that the plastic deformation is strongly localized and the main processes, controlling the nanomaterial behavior and properties, are realized in the grain boundaries [1–4, 9–11], not in the crystalline lattice (in grains) as it occurs in the normal materials. The main type of defects, determining the process history are inner interfaces, not dislocations and vacancies. Features of the behavior of the processes taking place in the grain boundaries are due to interaction of the boundaries with dislocations point defects, coming into them from the lattice, i.e. the diffusion processes play a decisive role in the realization of unique properties of the nanomaterials. Indeed, as the grain size decreases, the volume fraction of the material near the grain boundaries and boundary regions is increasing. The sizes of structure elements approach the diffusion path lengths being characteristic for different processes. Investigations on diffusion in the nanostructured metals and alloys have shown that the diffusion coefficients in such materials exceed, by several orders of magnitude, the corresponding values in coarse-grained materials [1].

Quantitative and qualitative difference in the properties of these materials is determined, in general, by the nonequilibrium state of the grain boundaries formed in the SPD process. Consequently, by changing the boundaries defect structure the strength and creep properties of the nanocrystalline materials can be changed.

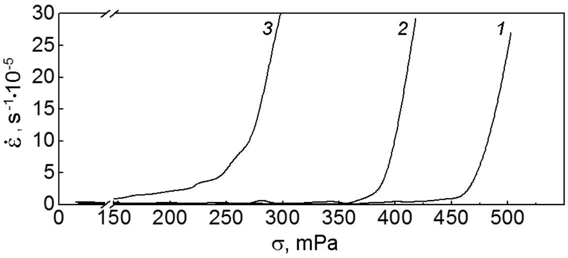

Fig. 1. Creep rates of nanostructured Zr1Nb alloy subjected to different treatments (1 — MT-1; 2 — MT-2; 3 — MTT-1 at T = 700 K) as a function of the applied stress.

As is known, the low-intense ultrasonic influence leads to relaxation of internal stresses in the material volume caused by some factors. Under the high-frequency alternating impact a great number of vacancies are generated that stimulates a nonconservative slip [5–7]. In this case dissipation of vibrational energy takes place, for the most part, at the interfaces that can lead to formation of the more equilibrium state of the structure boundaries, local heating to decrease of the local stresses and to activation of the dislocation sources [5–7]. It is common knowledge [7] that the ultrasonic field influence drives the process of dislocation multiplication by two channels. One of the channels is activation of the Frank-Read sources when each of them is activated under the ultrasonic impact with a lower value of the constant shear stress (the ultrasonic influence is reduced to action of the trigger mechanism). The second channel is the increase of a number of the Frank-Reader sources. The effect of all the above factors leads to the active displacement, interaction and annihilation of dislocations at the grain boundaries, as well as to recovery of the boundary structure. The grain boundaries become more equilibrium, the level of long-range stresses decreases that leads to the strength property deterioration and plasticity increasing. It is important to note that the equilibrium grain boundaries are more resistant to the mechanical-thermal effects under the creep conditions.

The structure investigations show (Fig. 2) that the combined rolling of the Zr1Nb alloy up to deformation value of ~ 3.9 results in the formation of the nano-grained structure with an average grain size of ~ 60 nm and polygonal nonequilibrium boundaries (Fig.

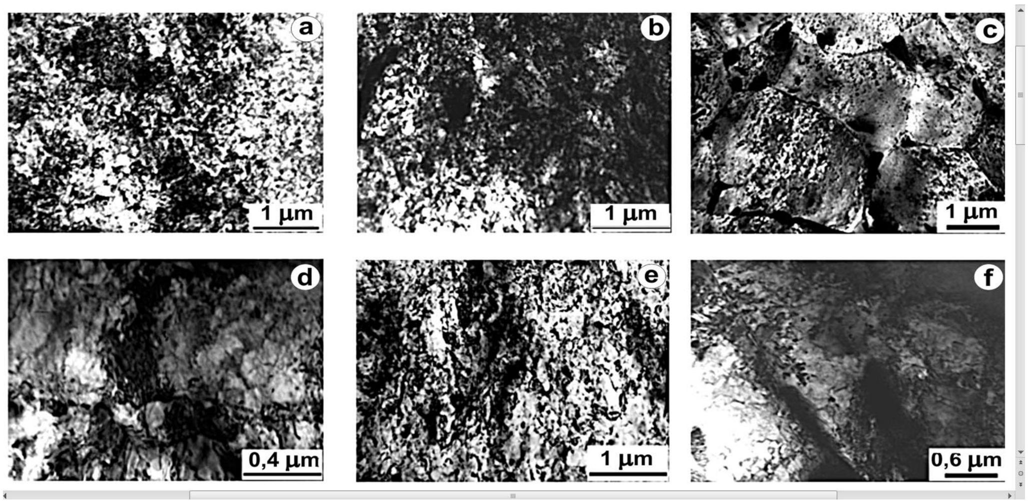

Fig. 2. TEM images of Zr1Nb alloy after following treatments: a) MT-1; b) MT-2; c) MTT-1; d) MT-1 + creep at 700 K; e) MT-2 + creep at 700 K; f) MTT-1 + creep at 700 K.

2a). The dislocation density in the grain body is ~ $3.4\ 10^{10}$ cm^{-2}. The most part of the dislocation mass is concentrated near the grain boundaries and triple junctions. The average size of misorientations caused by the boundaries is ~ 6°. There observed is a high concentration of large-angle boundaries (8–30°) as well as polygonal boundaries of dislocations. Sharp inhomogeneity of the contrast in the electron microscope images evidences on the high level of internal stresses and presence of peak stresses in the boundary junctions.

The high concentration of boundaries and overlapping fields of the long-range stresses, induced by these fields, create conditions for the high plastic resistance and nanostructure evolution under the creep at 700 K.

The nanostructure formed as a result of the SPD was nonresistant to the subsequent mechanical-thermal action under the creep conditions at 700 K (Fig. 2d).

The figure shows that the most part of boundaries are destroyed and dislocation boundaries of a polygonal type are formed in their place. The polygon size is varying within the range from 50 to 150 nm. There the extended boundaries with large, more than 20°, grain-boundary angles are remained. The initial structure transformation occurs because of activation of the return processes, due to dislocation climb near the grain boundaries, as well as the processes of generation and annihilation of dislocations at the boundaries that leads to their crumbling.

The action of ultrasound with the tangential stress amplitude of 80 MPa does not provoke the change in the morphology of the Zr1Nb initial deformation nanostructure, however, it becomes more equilibrium and homogeneous, the number of 60 nm grains increases and the average grain size slightly increases to 67 nm (Fig. 3). This is because of in this case the number of grains with the small-angle boundaries is reduced compared to the deformed state.

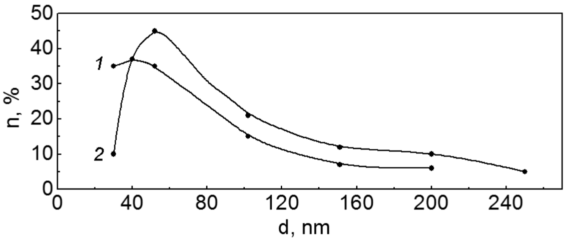

Fig. 3. Spectra of grain size distribution in the samples of nanostructured Zr1Nb alloy subjected to MT-1 (*1*) and MT-2 (*2*) treatments.

It is seen from the figure that the grain boundaries are more straight and thin, the angles in the grain triple junctions are close to the equilibrium one, the spectrum of internal stresses is even, and their level and scale are decreased (Fig. 2b).

The action of tensile stresses under the creep condition at 700 K leads to development of dynamic recrystallization in the samples processed with ultrasound (Fig. 2e). The average size of recrystallized grains is 100 nm. It can be concluded that as a result of the stress relaxation in the process of creep of the nanostructured Zr1Nb alloy subjected to UIT at 700 K the polygonization stage is absent. The nanostructure formed by the SPD is destructed and the new recrystallization nanostructure is formed which provides the high creep resistance and the high level of strength characteristics, the plasticity level being remained sufficient.

For comparison in Fig. 2c presented is the structure obtained using the thermal treatment (MTT-1), which decreases the internal stresses and increases the plasticity of the alloy but deteriorates its strength properties. This is a consequence of the recrystallization and formation of the new grains of about ~ 1 μm. The grains have equiaxed shape with the straight equilibrium boundaries. The intergranular structure is changing, the dislocation density decreases to ~ 10^8 cm^{-2}.

Furthermore, such a structure is unstable under the creep conditions at 700 K. The grain boundaries are fractured and the dislocation clusters takes place with a tendency towards the cellular structure formation (Fig. 2f). It has been found previously [2, 6–11] that the structural instability arising in the creep process is caused by the changes in the plastic deformation geometry and temperature-rate deformation mode.

4. Conclusions

The experimental results show that the ultrasonic impact treatment with amplitude of 80 MPa and frequency $f = 20$ kHz leads to relaxation of the internal stresses in nanostructured Zr1Nb alloy, produced by the SPD method, due to formation of the more equilibrium structure of boundaries without appreciable grain growth.

It has been found that in the nanostructured Zr1Nb alloy, subjected to the ultrasonic impact treatment, the creep at 700 K causes the relaxation of internal stresses as a result of dynamical recrystallization and formation of the new stable structure that significantly improves the thermomechanical material stability.

The effect of stress relaxation is observed also in the nanostructured Zr1Nb alloy subjected to the thermal treatment at 870 K providing the plasticity increase. However, in this case the alloy structure is changing qualitatively and the nanostructure state is destroyed. As a result, the strength characteristics are deteriorating and the creep rate increases. Moreover, the produced recrystallization structure possesses the low thermomechanical stability against the subsequent deformation under the creep conditions.

So, the ultrasonic impact treatment with optimum amplitudes can be used as an effective alternative or addition to the relaxation modes of metal and alloy treatments and extends the potentialities for modification of the structure and properties of nanostructured materials.

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