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PECULIARITIES OF STRESS RELAXATION OF NANOSTRUCTURED ALLOY Zr1Nb AFTER VARIOUS INFLUENCES

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Processes of relaxation of internal stresses in the nanocrystalline alloy Zr1Nb after various thermomechanical influences was studied. The relationship of structural states obtained as a result of the relaxation effects with the laws of plastic deformation during creep were established. Shown that heat treatments of nanostructured alloy Zr1Nb, which provide the increase of the plasticity, changed qualitatively the alloy structure, destructing the nanostructured state, that results to decrease of the mechanical characteristics of the material and the degree of resistance to subsequent deformation during the creep at the temperature of 700 K. Ultrasonic processing reduces the level of internal stress of nanostructured material by forming the equilibrium structure of boundaries without noticeable growth of grains. During subsequent creep deformation at 700 K of nanostructured alloy Zr1Nb, subjected to ultrasonic treatment, the relaxation of the internal stresses due to dynamic recrystallization and the formation of new stable nanostructure take place that leads to the marked increase of the material thermomechanical stability.

INTRODUCTION

Nanocrystalline metals and alloys, obtained by means of intensive plastic deformation (IPD) have unique functional and mechanical properties such as high strength, wear resistance, hardness, high fatigue properties due to a number of structural peculiarities. The large part of grain boundaries in the structure of nanocrystalline materials, which are in nonequilibrium state and contain the large free volume has the particular importance. Such boundaries create a high level of internal stresses and, consequently, nanomaterials have low plasticity and thermal stability [1–4], which reduces their processability.

For stress relaxation of nonequilibrium structures of nanomaterials obtained by the methods IPD is typically used annealing, the effectiveness of which increases with increasing temperature. However, it is known that due to high internal stresses in such materials the grain growth begins at lower temperatures [5], which may lead to loss of advantage of nanocrystalline state, such as high strength. Therefore, it is needed the search of relaxation effects, which do not destroy the nanostructure of the material in order to obtain the optimum combination of mechanical properties of them.

One of the promising methods for improving the properties of nanostructured materials can be ultrasonic impact treatment (UIT). The ultrasonic wave, passing through the material, interacts with various types of defects and causes the changes in the structure, which depend on the parameters of the ultrasound mainly on its power. By varying the parameters, the structure, having needed properties, can be obtained in the material. Previously we shown [6, 7], that as a result of influence of low-intensity ultrasound on the deformed nanostructure, the level reduction and equalization of the spectrum of internal stresses in the bulk of the material while maintaining the size factor and improving the uniformity of structure occurs.

Furthermore, an important issue is the stability of the obtained structural states in a wide temperature range, as strength and plastic characteristics of the material can considerably changed in the event of the appearance of structural instability [1–12].

The purpose of this paper is to investigate the peculiarities of creep and evolution of nanostructure alloy Zr1Nb, obtained with the use of rolling and subsequent relaxation influences. This will allow to develop an idea of the peculiarities of stress relaxation after different thermomechanical treatments, and also to establish the boundaries of practical use and the possibility of further improving the properties of nanocrystalline metallic materials obtained by methods of IPD.

MATERIAL AND EXPERIMENTAL PROCEDURE

The investigated material was the polycrystalline alloy Zr1Nb, obtained by electron beam melting. With the purpose to influence on the structure and properties of the alloy Zr1Nb the various treatments were conducted in the following modes:

1 – MT-1 – combined rolling at 77…300 K, the residual deformation ε was 3.9;

2 – MT-2 – MT-1 + UIT at 300 К;

3 – MTT 1 – MT-1 + annealing at 500 K, 1 h;

4 – MTT-2 – MT-1 + annealing at 570 K, 1 h;

5 – MTT-3 – MT-1 + annealing at 720 K, 1 h;

6 – MTT-4 – MT-1 + annealing at 800 K, 1 h;

7 – MTT-5 – MT-1 + annealing at 870 K, 1 h.

The part of rolled samples was subjected to low-intensity UIT (f = 20 kHz) at T = 300 K by the method described in [5]. The amplitude of the ultrasonic shear stresses was 80 mPa, the duration was 10 min. The selected mode of pre-processing ultrasound, as shown previously, produces the softening effect on the deformed material [5–7].

For studying the material defect structures, the electrical resistance (R) was measured by 4-th points scheme at T = 300 K after each treatment. A measuring error did not exceed ±0.05 %.

The structure evolution monitoring was carried out by electron microscopy.

Creep tests were carried out in the step loading regime, the elongation measurement accuracy was $5 \cdot 10^{-5}$ cm.

RESULTS AND DISCUSSION

Values of the relative electrical resistance (R) of Zr1Nb alloy samples after rolling deformation and different treatments were determined. The results are shown in Table.

Measured charac-teristics	Modes of treatment						
	MT-1	MT-2	MTT-1	MTT-2	MTT-3	MTT-4	MTT-5
$R_{300\,K}/R_{77\,K}$	3.03	3.47	3.38	3.45	3.57	3.57	3.57

Mechanical characteristics (yield strength, tensile strength, plasticity) after various treatments were examined in the temperature range 300…700 K under creep conditions (Fig. 1).

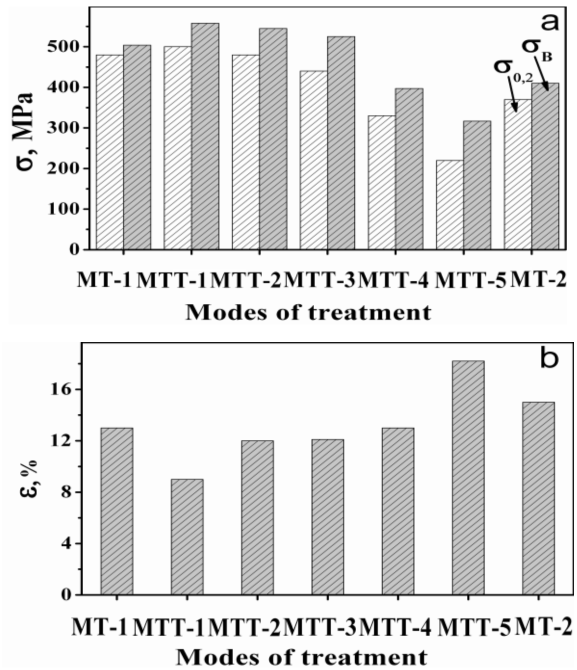


Fig. 1. Mechanical properties of nanostructured Zr1Nb alloy after various treatments: a – yield strength ($\sigma_{0,2}$) and tensile strength (σ_B); b – plasticity (ε)

Dependences of the creep rate at 700 K (operating temperature of reactors) of alloy samples after all treatments from the applied stresses is shown in Fig. 2 .

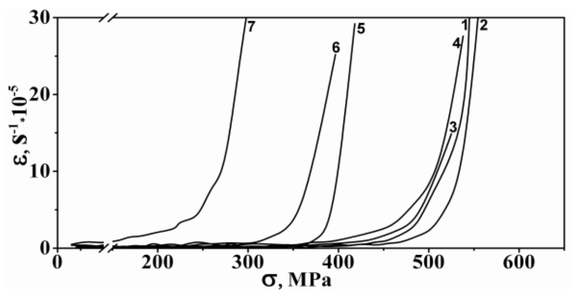


Fig. 2. Dependence of creep rate of nanostructured alloy Zr1Nb at 700 K from the applied stress after all treatments: 1 – MT-1; 2 – MTT-1; 3 – MTT – 2; 4 – MTT-3; 5 – MT-2; 6 – MTT-4; 7 – MTT-5

Analysis of the results shows that after annealing at 500 K the small hardening effect of Zr1Nb alloy is observed. Similar effect was observed after annealing in the temperature range before recrystallisation in other metals [8–10]. Since during annealing the substructure of nanomaterials tend not formed, the traditional points of view cannot be used for explaining the effect. The assumptions about the relationship the hardening effect in nanostructure metals after annealing in the temperature range before recrystallisation with the processes of formation of impurity atmospheres around dislocations, or the peculiarities interaction of grain boundaries with impurities, or with the accumulation of defects in the migrating grain boundaries of metals were made in the works [8–10]. However, the question of the hardening mechanisms is open until now and requires detailed experimental studies and construct of corresponding physical models.

Annealing at temperature of 500…720 K leads to some change in strength characteristics of the material when compared with the initial state (see Fig. 1,a), however, do not affect the value of plasticity (see Fig. 1,b) and creep speed (see Fig. 2). It should be noted that changes in the mechanical characteristics are accompanied by the constant increase of the residual resistivity, indicating the decrease in the level of internal stresses and the redistribution or reduction of the overall level of defects. It is also known that the temperature of the beginning of the dynamic effect of aging for the alloy Zr1Nb is about ~ 400 K and at the same temperature, the maximum of diffusion mobility of oxygen atoms is observed [11]. As shown in [12], when annealing at the third stage of recovery in deformed niobium the oxygen-vacancy complexes precipitate on dislocation sinks. This leads to the lattice crystal cleaning and to decrease of residual resistance compared to the undeformed condition. It can be assumed that in the alloy Zr1Nb, subjected to IPD, there are similar processes when annealing in this temperature range.

Further increase of the annealing temperature to $T > 720$ K leads to decrease the strength properties and increase of plasticity and creep rate of nanostructured samples Zr1Nb alloy at the temperature test of 700 K (see Fig. 1). At the same time the residual electrical resistivity does not change (see Table), which may be related to the restructuring and ordering of the dislocation structure [13, 14].

As a result of ultrasonic processing (MT-2) the strength characteristics of the material decreased by ~ 15% in comparison with the deformed specimens, at the same time the plasticity increased by ~ 20% (see Fig. 1).

It should be noted that the specifics of nanomaterials, from the viewpoint of the theory of defects, lies in the fact that plastic deformation is highly localized and the basic processes that control their behavior and properties are not realized in the crystal lattice (in grains), as in traditional materials, but take place in grain boundaries [1–4]. Therefore, the main type of defects, determining the nature of these processes, are not the dislocations and vacancies (as in traditional materials), but are the internal interface boundaries. Moreover, the qualitative and quantitative properties of these materials is determined primarily by non-equilibrium state of grain boundaries. Therefore, we can affect characteristics of nanocrystalline materials by changing the defective structure of the boundaries.

It is known that as a result of low-intensity ultrasound treatments (UST) occurs relaxation of internal stresses in the material volume due to the number of factors. At the high-frequency alternating influences the large number of vacancies is generated, which are stimulate of non-conservative slipping [5–7]. Moreover, the dissipation of vibrational energy occurs mainly at the interface boundaries, which can lead to the formation of the equilibrium state of boundary structure, as well as to the local heatings, reducing of the level of local stresses and the revitalization of the dislocation sources. The imposition of ultrasonic field starts the process of dislocation multiplication through two channels [6]. One of the channels is the activation of Frank-Read sources, but due to the action of the ultrasound, the activation of each source occurs at the lower value of shear stress (the effect of ultrasound act as the trigger). The second channel is the increase of the number Frank-Read sources.

The effect of all these factors leads to the active movement, interaction and annihilation of dislocations at grain boundaries, and to the rehabilitation of the structure of boundaries. Grain boundaries become more equilibrium, resulting in the reduced level of long-range stress and the certain reduction of the strength characteristics of the alloy. Moreover the equilibrium grain boundaries are more resistant to subsequent thermal-mechanical influences.

Investigations of the structure have shown that after the combined rolling of Zr1Nb alloy on the value of true strain $\varepsilon \sim 3{,}9$ nanostructure with the grain size of ~ 60 nm is formed (Fig. 3,a). The dislocation density in the body of grains is about ~ $3.4 \cdot 10^{10}$ cm^{-2}. Most of the dislocations are concentrated at the grain boundaries and triple junctions. The average size of disorientation, caused by boundaries, is about ~ 6^0. At the same time there are the big concentrations of the highangle boundaries ($8…30^0$), as well as the ragged dislocation boundaries. The sharp heterogeneity of contrast on electron microscopic images testify of the high level of internal stresses and the presence of the peak stresses in the junctions of the boundaries.

Annealing nanostructured alloy Zr1Nb in the temperature range 500…570 K does not cause the significant change in the character of the microstructure. All the peculiar properties of the microstructure, which are characteristic for the nanostructured state, are stored. Some redistribution and reducing of the dislocation density at the grain boundaries and especially at triple junctions are observed. The distribution of the dislocations in the body of grains becomes more uniform.

After annealings at 720…800 K the structural changes indicative of the return process in the grain boundaries are observed.

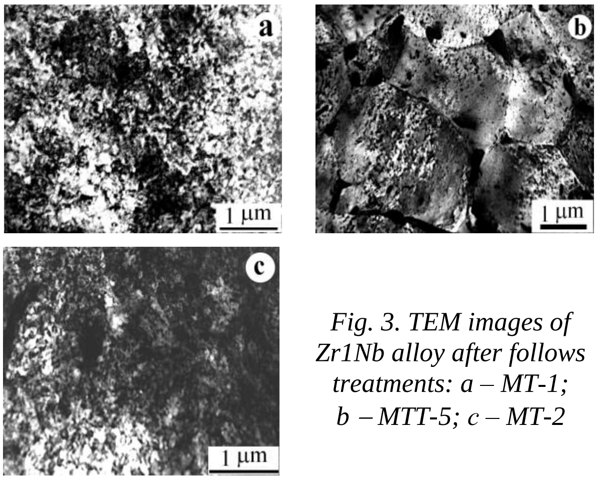


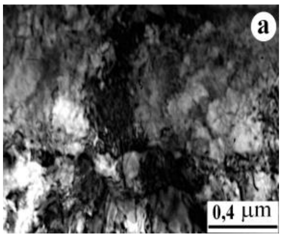
Fig. 3. TEM images of Zr1Nb alloy after follows treatments: a – MT-1; b – MTT-5; c – MT-2

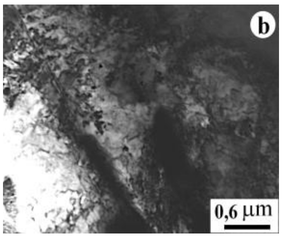
The heterogeneity of distribution of stresses in the nanostructure is preserved as evidenced by the non-uniformity of relaxation processes in the volume of material. We can observe In electron micrographs as the polygonal boundaries so and the embryos recrystallization grains size of ~ 30 nm.

Initial recrystallization comprise the entire volume of material (see Fig. 3,c) after annealing at 870 . The formations of new grains size up to 1 μm occur. The grains have the equiaxed shape, at the same time the structure inside the grains and the structure of the grain boundaries varies. The dislocation density within the grains is not exceed ~ 10^8 cm^{-2}.

Ultrasound treatments of the nanostructured alloy Zr1Nb, obtained by rolling, does not alter the morphology of the initial deformed nanostructure (see Fig. 3,b). However, it becomes more homogeneous and equilibrium, the average size of nanofragments is slightly increased (d = 67 nm). This is primarily due to the decrease in the number of fragments with low-angle boundaries, which have been so much in the deformed state. The range of internal stress is aligned, their level and scale is reduced. The grain boundaries become more smooth and thin, the angles at triple grain junctions are approached to equilibrium, the density of defects within the grains reduces.

After all treatments the samples of Zr1Nb alloy were studied under creep conditions at T = 300…700 K. We have previously shown [13–17] that the plastic deformation of the submicro grained and nanostructured Zr and alloy Zr1Nb, obtained with the use of IPD, occurs due to the restructuring of the defected structure in the process of creep and is accompanied by stress relaxation. This process involves the destruction of the original structural configuration, created as a result of rolling, and the formation of the new structure, which is less stressful and more resistant to subsequent deformation. The cause of the kinetic instability of the deformation structures is the change of the geometry of plastic deformation and the mode of the deformation.





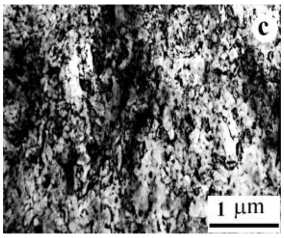


Fig. 4. TEM images of Zr1Nb alloy after follows treatments:
a – MT-1+ creep at 700 K ($\sigma \approx 0.9\sigma_B$);
b – MTT-5 + creep at 700 K ($\sigma \approx 0.9\sigma_B$);
c – MT-2 + creep at 700 K ($\sigma \approx 0.9\sigma_B$)

Structural studies have shown that the nanostructure created by IPD rolling, proved to be unstable to subsequent mechanical-thermal actions in creep conditions at 700 K (Fig. 4,a). Most of the boundaries were destroyed and in their place were formed the dislocation boundaries of polygon type. Sizes of polygons vary in ranges between 50…150 nm. The elongate boundaries with the large, above ~ 20^0, angles of disorientation partially preserved. The transformation of the original structure is due to the activation of the return processes by climb of dislocations near the grain boundaries, and also by the processes of generation and annihilation of dislocations at the boundaries, which leads to their disintegration [11–14].

Qualitative changes in the deformation structure during the annealings with the destruction of nanostructured states lead to its sharp transformation in the process of creep. So, the processes of dynamic recrystallization during the creep at 700 K begin to develop in the structure obtained after annealing at 800 K and the new grains of size 0.1…0.5 μm are formed. The structure of Zr1Nb alloy, completely recrystallized after annealing at 870 K, was unstable in creep conditions at 700 K. The new grain boundaries were destroyed and clusters of dislocations with a tendency to the formation of the cellular structure were formed (see Fig. 4,b).

Heat treatments that provide the growth of alloy plasticity, qualitatively changes the structure, destroying the nanostructured state that leads to the decrease in strength properties and to the increase in creep rate. Moreover, the such structure has the low thermomechanical resistant in relation to the subsequent deformation in the creep condition.

The action of tensile stresses in creep conditions at 700 K on the samples after ultrasounds leads to the restructuring of structure and to the development of dynamic recrystallization (see Fig. 4,c). Average size of recrystallized grains is 100 nm. We can conclude that in the process of creep at 700 K, nanostructured alloy Zr1Nb, subjected to the ultrasound, the stress relaxation is due to the restructuring of the structure so the stage of polygonize is absent. Dynamic recrystallization is developed and the new recrystallization nanostructure is formed, which is more adapted to the new conditions of deformation and provides the high resistance to the creep and the level of strength properties while maintaining the sufficient level of plasticity, ie with higher levels of thermomechanical stability.

CONCLUSIONS

Heat treatments that provide the growth of alloy plasticity, qualitatively changes the structure, destroying the nanostructured state that leads to the decrease in strength properties and to the increase in creep rate. Moreover, the such structure has the low thermomechanical resistant in relation to the subsequent deformation in the creep condition.

Ultrasonic impact treatment with the amplitude of 80 mPa and frequency of 20 kHz reduces the level of internal stress of nanostructured material by forming the equilibrium structure of boundaries without noticeable growth of grains. During subsequent creep deformation at 700 K of alloy nanostructured Zr1Nb, subjected to ultrasonic treatment, the relaxation of the internal stresses due to dynamic recrystallization and the formation of new stable nanostructure take place that leads to the marked increase thermomechanical stability of the material.

Thus, the ultrasonic treatment with the optimal modes may be an effective alternative or complement to the processing of relaxation type of metals and alloys and extends the possibility of modifying the structure and properties of nanostructured materials.

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ОСОБЕННОСТИ РЕЛАКСАЦИИ НАПРЯЖЕНИЙ НАНОСТРУКТУРНОГО СПЛАВА Zr1Nb ПОСЛЕ РАЗЛИЧНЫХ ВОЗДЕЙСТВИЙ

В.И. Соколенко, Е.В. Карасева, А.В. Мац, Е.С. Савчук, В.А. Фролов

Исследовали процессы релаксации внутренних напряжений в нанокристаллическом сплаве Zr1Nb после различных механико-термических воздействий. Установили взаимосвязь структурных состояний, полученных в результате релаксационных воздействий с закономерностями развития пластической деформации при ползучести. Показали, что термообработки наноструктурного сплава Zr1Nb, которые обеспечивают повышение пластичности, качественно изменяют структуру сплава, разрушая наноструктурное состояние, что приводит к снижению механических характеристик и степени устойчивости материала к последующей деформации в процессе ползучести при температуре 700 К. Ультразвуковая обработка приводит к снижению уровня внутренних напряжений наноструктурного материала за счет формирования более равновесной структуры границ без заметного роста зерен. В процессе последующей деформации в условиях ползучести при 700 К наноструктурного сплава Zr1Nb, подвергнутого ультразвуковому воздействию, происходят релаксация внутренних напряжений вследствие динамической рекристаллизации и образование новой стабильной наноструктуры, что приводит к заметному повышению термомеханической устойчивости материала.

ОСОБЛИВОСТІ РЕЛАКСАЦІЇ НАПРУЖЕНЬ НАНОСТРУКТУРОВАНОГО СПЛАВУ Zr1Nb ПІСЛЯ РІЗНИХ ВПЛИВІВ

В.І. Соколенко, Є.В. Карасьова, О.В. Мац, Є.С. Савчук, В.О. Фролов

Досліджували процеси релаксації внутрішніх напружень у нанокристалічному сплаві Zr1Nb після різних механікотермічних впливів. Встановили взаємозв'язок структурних станів, отриманих в результаті релаксаційних впливів з закономірностями розвитку пластичної деформації при повзучості. Показали, що термообробки наноструктурованого сплаву Zr1Nb, які забезпечують підвищення пластичності, якісно змінюють структуру сплаву, руйнуючи наноструктурований стан, що призводить до зниження механічних характеристик і ступеня стійкості матеріалу до наступної деформації в процесі повзучості при температурі 700 К. 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, що піддавали ультразвуковому впливу, відбуваються релаксація внутрішніх напружень внаслідок динамічної рекристалізації і утворення нової стабільної наноструктури, що призводить до помітного підвищення термомеханічної стійкості матеріалу.