Dependence of Thermal Expansion of Zr-Based Products on their Crystallographic Texture

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Abstract. The accident at Fukushima Daiichi in 2011 highlighted the increased interest in predicting the materials properties under extreme conditions. In this work, it is considered the behaviour of cubic samples cut from the wall of channel tubes made of Zr-2.5% Nb alloy during heating and cooling in the temperature range of 20-1200 °C. The anisotropy of changes in the dimensions of cubic samples in different structural states has been studied. Thermal tests were carried out in three mutually perpendicular directions: radial, tangential and axial. The structural state of tubes was estimated from the features of their crystallographic texture. The significant anisotropy of the size change is observed in the Zr-2.5%Nb alloy product with an unstable crystallographic texture of deformation due to the \(\alpha\leftrightarrow\beta\)-phase transformation inhomogeneity.

In the case of complete recrystallization and orientation of the <11\(\overline{2}\)0> axes along the tube axis, the anisotropy of the dimensions change is minimal, due to uniform nuclei formation of new phases in different orientations.

1. Introduction

The tightening of requirements for structural materials in connection with emergencies in recent years at nuclear power plants (Chernobyl, Fukushima) explains the increased interest in predicting the materials properties under extreme conditions. The need to preserve the tightness of fuel elements under operating conditions with temperature drops and to ensure the possibility of subsequent repair work brings to the fore the issues of stabilizing the dimensions of products as a result of their cyclic heating and cooling. In this regard, it is necessary to know not only the thermal coefficients of linear expansion of fuel elements to operating temperatures, but also their behavior in the event of a sharp increase in temperature in an emergency.

According to numerous studies of the kinetics of changes in the dimensions of cladings and channel tubes [1-6], the thermal expansion of tubes during their heating in the region of existence of a low-temperature hexagonal phase is determined with the distribution of basal axes. This makes it possible to observe the proportional dependence of the coefficient of linear thermal expansion (LTEC), on the integral texture parameters [1-6]. The reduction in size as a result of cooling the tubes fully corresponds to the LTEC for the single-phase region if there were no phase transformations in the sample during heating. If the temperature interval of heating the sample is extended to the region of existence of the \(\beta\)-phase, for example, up to 1200 °C, then as a result of phase transformations (PTs) the crystallographic texture changes, which also leads to changes in the LTEC [5-6]. The repetition of the heating and cooling

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cycles in the future does not affect the changes in the crystallographic texture and, consequently, the LTEC [6].

This paper considers the regularities of PTs $\alpha \rightarrow \beta \rightarrow \alpha$ in tubes made of Zr-2.5% Nb alloy, which are implemented during the heating-cooling cycle of thermal tests in the temperature range of 20-1200 °C. The tests were carried out in three mutually perpendicular directions for cubic samples cut from the wall of the different channel tubes. This work is devoted to the study of the influence of the crystallographic texture on the process of thermal linear expansion of samples from the Zr-2.5% Nb alloy in the different directions of the studied tubes.

2. Samples and methods
Most of the works on the study of thermal expansion have been carried out on finished products, since researchers are mainly interested in their performance properties. However, the expansion of tubes ensures their volumetric changes in such a way that changes in linear dimensions in different directions are dependent on each other. In this regard, it is necessary to prepare free samples on which it is possible to establish a real change in dimensions in three mutually perpendicular directions.

For this purpose, in this work, samples for thermal tests have been cut from thick-walled channel tubes made of Zr-2.5% Nb alloy pre-annealed in different modes. Samples of the first series were annealed at a temperature of 400 °C for 24 hours (hereinafter samples # 1), and the second series - at a temperature of 580 °C for 3 hours (samples # 2). Then samples of a cubic shape were cut from the walls of the tubes, the cutting scheme is shown in figure 1. The edges of the cubes were oriented along the main directions of the channel tube: radial $R$, tangential $T$, and axial $L$. The final dimensions of the prepared samples were approximately 4x4x4 mm.

The study of samples expansion in three mutually perpendicular directions: $R$, $T$ and $L$, in the temperature range of 50-1200 °C was carried out on a Netzsch 402 C dilatometer in a flow of high-purity helium (99.9999%) with its additional purification by a Monotorr getter catalytic filter. An additional oxygen getter OTS manufactured by Netzsch was located next to the sample. The rate of heating and subsequent cooling of the test sample was 5°/min. The method of temperature experiments is described in detail in the works [5-6].

X-ray studies of the structure and crystallographic texture of the initial samples, as well as the samples after a heating and cooling cycle in the temperature range of 50-1200 °C, were carried out on DRON-3 and D8 Discover diffractometers using Cr K$_\alpha$ and Cu K$_\alpha$ radiation, respectively. Based on incomplete direct pole figures (DPFs) taken from three mutually perpendicular sides of a cubic sample, the complete DPFs of the sample for a plane perpendicular to the $R$-direction were reconstructed by the PF combination [7-8]. Based on the calculated complete DPF (0001), the integral texture parameters of
Kearns were estimated for three directions $R$, $T$ and $L$ [9]. The Kearns parameter is the sum of the effective fractions of the basal axes [0001] oriented along the chosen direction in the polycrystal material. This parameters for different directions are used to evaluate the properties of textured material with known properties for a hexagonal single crystal along the crystallographic axes $a$ and $c$ [9-11].

3. Experimental results and discussion

According to diffraction spectra of tubes No. 1 and No. 2 shown in figure 2, the material of the tubes differs significantly in composition. The main phase of both tubes is the $\alpha$-phase, solid solution of Nb in the hcp-Zr. An additional phase in the tube # 1 was the $\omega$-Zr phase, which is typical for these alloys with this kind of heat treatment [4]. It may also indicate a small degree of preliminary plastic deformation of the tube # 1. In the tube # 2, the $\beta$-phase with a high niobium content (90 wt.%) is turned out to be an additional phase, which proves the equilibrium of the final state of the phases in this tube, corresponding to the equilibrium phase diagram of the Zr-Nb alloys [12].

The difference in the structural state of the studied tubes is also evidenced by the DPFs (0001) and $\{11 \overline{2} 0\}$ shown in figure 3-a, b. Both tubes are characterized by a pronounced axial component with a predominance of different texture components. In the tube # 1, the axes $<10\overline{1}0>$ are oriented along the rolling direction. This texture component is corresponded to the rolling texture of the $\alpha$-zirconium. But, in the tube # 2 the axes $<11\overline{2}0>$ are oriented along the rolling direction, confirming the occurrence of recrystallization in the tube through grain growth misoriented relative to the deformed matrix at an angle of 30$^\circ$. The distributions of basal axes in the R-T-section of the DPF (0001) are also different. In tube # 1, the basal axes are predominantly oriented in the T-direction, and in the tube No. 2, the basal axes are deflected from the R-direction towards T by 30-40$^\circ$.

As a result of high-temperature tests of the described samples, their crystallographic texture changes in different ways. DPFs (0001) and $\{11 \overline{2} 0\}$ after the heating-cooling cycle in the temperature range of 20-1200 °C are shown in figure 3-c, d. The given DPFs are characterized by irregularity of the contours, which is due to the large size of $\beta$-grains formed in the high-temperature region of the state diagram, a decrease in the number of reflecting grains, and violation of measurement statistics. Nevertheless, the main regularities of the texture of PTs are obvious.

As noted in [8, 11], the localization of PTs significantly depends on the distribution of stored energy in grains of different orientations. As a result of an inhomogeneous PTs in a sample with a less stable structural state, the $\alpha$$\rightarrow$$\beta$-PTs are localized mainly in grains, the basal axes of which are oriented in the tangential direction (figure 3-c). In this case,
in a sample #2, $\alpha \rightarrow \beta$-PTs flow more uniformly in grains of different initial orientations, i.e. distribution of axes along the R-T-section of the DPF (0001) is preserved (figure 3-d) [8, 11]. The appearance of additional texture maxima on the DPF (0001) is determined by the orientation of the prismatic axes, the symmetry of the cubic lattice, and a high repeatability factor of the $\{110\}$ planes, parallel to which the unique basal planes of the $\alpha$-phase are located at the inverse-PT. The theoretical arrangement of texture maxima is described in a large number of works [4, 8, 11, 13-14]. The obtained experimental results completely coincide with theoretical calculations. The reverse PT can be accompanied by the predominant growth of grains of different orientations and is determined by the level of external stresses [15]. However, in the context of this work, we will not dwell on the differences in the DPFs for the samples on which measurements were carried out in different directions. All samples cut from the same tube have the same type of final texture.

Figure 4 shows the thermal expansion curves for all studied samples, measured during heating (solid lines) and cooling (dashed lines) in the temperature range 20-1200 °C. The results of testing cubic samples in different directions $R$, $T$ and $L$ are shown in figure 4 by curves of different shades: black is corresponded $R$-direction, gray - $T$ and finally light gray - $L$. The dimensions of the tested samples were measured before and after thermal tests. At the same time, different size changes were recorded in different directions, which are shown in Table 1.

![Figure 4](https://example.com/figure4.png)

**Figure 4.** Curves of linear expansion and contraction in three main directions at the heating (solid lines) or cooling (dashed lines) of cubic samples of the tubes annealed at temperatures of 400 °C during 24 h (a) and 580 °C during 3 h (b).

The sizes of the tested samples of tube # 1 vary widely, from −1.3 to 2.3%, while the sizes of the samples of tube # 2 vary from −0.3 to 0.2%. Therefore, the anisotropy of the change in the dimensions of the sample of tube # 1 is 4-5 times greater than for the tube # 2.

The final change in the size of the samples is completely determined by the processes of $\alpha \rightarrow \beta \rightarrow \alpha$-PTs in the temperature range 600-850 °C, occurring both during heating and during cooling of the samples. Let us consider the main processes that can lead to a change in the dimensions of the samples under study. First of all, heating of samples in the temperature range of existence of the $\alpha$-phase is accompanied by their expansion, at which the LTEC is proportional to the integral texture parameter $f$ for the considered direction [1-5].

When the temperature of the monotectoid transformation 610 °C reaches, the $\alpha$-$\beta$ transformation begins, at which the sizes of the samples begin to decrease in all directions, as shown in [1-4, 6] and can be seen in figure 4. Such a decrease is due to a reduction in the size of the unit cell of the bcc phase as compared to the hcp. Moreover, in the direction of the $c$ axis, this decrease is less than along the $a$ axis, as was shown in [6] ($\Delta c = -3.26\%$, $\Delta a = -5.5\%$). The sharp crystallographic texture explains the difference in size variation in the $R$- and $T$-directions. The largest decrease in size occurs in the radial direction, and the smallest decrease is experienced by the sample in the axial direction.
Depending on the initial structural state, the onset of the PT shifts relative to the temperature corresponding to the onset of the PT in the equilibrium phase diagram. If the structure is less stable, as observed in the case of tube # 1, the PT process proceeds inhomogeneously and depends on the orientation, then the effect of size reduction in different directions will be recorded at different temperatures. It should not be forgotten that PTs occur throughout the entire volume of the material and affect changes in dimensions in all directions. But, since these changes are anisotropic, different directions are sensitive to different volumetric rearrangements. For example, in the case of tube # 2, for almost all directions, the sample dimensions change simultaneously (synchronously) (figure 4-b). For the L-direction of both tubes, the smallest reduction in sample size is recorded.

In the region of existence of the β-phase, the coefficient of linear expansion is the same for all directions, and within the limits of error, it can be assumed that the samples have the same dimensions at a temperature of 1200 °C.

At the stage of cooling the samples, the main changes in their dimensions are noted, which are then recorded at room temperature (Table 1). The largest increase in dimensions is observed for the T-direction of tube # 1, while in the L- and R-directions there is a reduction in dimensions. What processes can lead to size changing in the two-phase region of the phase diagram of Zr-Nb alloys?

With a decrease in the temperature of the single-phase Zr-2.5% Nb alloy to the two-phase region (α + β), the formed crystals of the α-phase are characterized by larger sizes in comparison with β-crystals. Since (0001)α || [110]β, the change in interplanar distance is εc = d_{110} - d_{0001} = 1 - d_{0001} / d_{110} = 1 - c_a / √2a_β, and for directions <1120>α || <111>β εa = d_{111} - d_{1120} = 1 - d_{1120} / d_{111} = 1 - 2a_a / √3a_β. With the substitution of specific values, it was shown [6] that an increase in the size of the cells of the hcp phase compared to the bcc phase should be observed both in the basal plane and perpendicular to it (Δc = + 1.8%, Δa = + 4.2%). The magnitude of changes in the parameters during forward and reverse phase transformations depends most of all on the concentration of niobium in the β-phase, the amount of which differs significantly and is estimated by the position of the β-phase line in the patterns (figure 2). Then all temperature dependences should increase with decreasing temperature. However, in figure 4-a, b it can be seen that along with an increase, a decrease in size is also possible (for example, for the R-direction). An additional process with decreasing temperature is the redistribution of niobium between phases. Moreover, in accordance with the phase diagram [12], the increase in the niobium content in the bcc phase occurs at a higher rate than niobium in the hcp phase. In this case, the parameter of the crystal structure of the bcc phase in accordance with Vegard’s law decreases by an order of magnitude larger than due to the thermal decrease in the size of the β-phase in accordance with its LTEC [12].

These two described processes occur simultaneously in the material, and the diffusion process is more likely to occur at higher temperatures of the two-phase region, which can be seen from the temperature dependence of the cooling of the sample in the R-direction (figure 4-a). With a decrease in the temperature of the two-phase region from 860 to 610 °C, the curve of decreasing dimensions in the

### Table 1. Changing the size of cubic samples in different directions as a result of a cycle of thermal tests in the temperature interval 20-1200 °C

| Tube number | Test direction | Resizing the sample, % | Initial state |
|-------------|----------------|------------------------|--------------|
| 1           | R              | -1.0                   | Anneal 400 °C - 3 h |
|             | T              | -0.3                   | Anneal 580 °C - 5 h |
|             | L              | -0.2                   |              |
R-direction changes the angle of inclination near temperatures of 780-800 °C, which indicates a change in the prevailing mechanism. For the T-direction, which was predominant in the original tube for the orientation of basal axes, the process of formation of the α-phase is of priority, which ensures a significant increase in dimensions in this direction with the predominant orientation of the basal axes in the T-direction.

Thus, it has been shown that in the presence of a sharp crystallographic texture in the product, significant anisotropy of the size change is observed. This can lead to the development of significant macro stresses in the case of heating the pipe to the two-phase region and subsequent cooling: high compressive stresses should develop in the tangential direction due to the need for the pipe to increase its dimensions in this direction, while a decrease in dimensions is observed in the radial direction.

4. Conclusions
1. It is shown that in the presence of an unstable crystallographic texture of deformation in a product made of the Zr-2.5%Nb alloy, a significant anisotropy of thermal change in product dimensions is observed as a result of the α↔β-phase transformation inhomogeneity.
2. If the material is characterized by a recrystallization texture, then the formation of nuclei of new phases occurs uniformly in different orientations, which ensures a significantly smaller change in the dimensions of the product as a whole.

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