Regularities of crystallographic texture formation in cladding tubes from Zr-based alloys during their production

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Abstract. This paper summarizes researches of the authors, which are directed on the development of the methodological basis of X-ray studies in the materials science of zirconium and on the systematization of new experimental results obtained using developed methods. The paper describes regularities of the formation of the crystallographic texture and the substructure inhomogeneity of cladding tubes from zirconium alloys at various stages of their manufacturing, i.e. during hot and cold deformation, recrystallization, phase transformations and interactions of the above processes.

1. Introduction

Zirconium stands out among all other metals by extreme importance of the crystallographic texture for the service properties of products from the Zr-based alloys [1-12]. Zirconium-based alloys are used for manufacturing of critical structural elements of nuclear thermal neutron reactors. Their behavior under exploitation is controlled by anisotropy of the radiation growth and creep and by the development of hydride formation, which are predetermined in the significant degree by the features of crystallographic textures formed in products [11-14]. The correlation of corrosion behavior of cladding tubes with their crystallographic texture determined by a misorientation of neighboring grains was also observed in papers [15-17]. Therefore, optimization of manufacturing technologies of products from zirconium alloys requires a knowledge of regularities of the texture formation in them under different types of a plastic deformation and a heat treatment. Only the scientific support and the justification of technological developments at the field of zirconium production can ensure its real progress and effectiveness. In particular, this applies to the systematic study of the texture formation in products at successive stages of their production, and to the analysis of all other crystallographic aspects of the structure formation, one way or another related to the development of the texture.

The development of the deformation texture in a metallic material is consist in following: grains, initially had different orientations, as a result of activation of micromechanisms undergo regular rotation of the crystal lattice and at the orientation space are moving to certain final orientation, stable in relation to the used deformation scheme. At the same time, the substructure forming in a grain depends on its "biography", consisting of initial and final orientations, the trajectory of the grain at the orientation space, strain micromechanisms acting on different parts of its trajectory, the resulting distribution of dislocations and the level of the strain hardening. So, the grains corresponding to different components of the deformation texture obviously differ in their "biographies" and substructure, whereby the metal material with a multi-component texture is, in fact, a composite. The
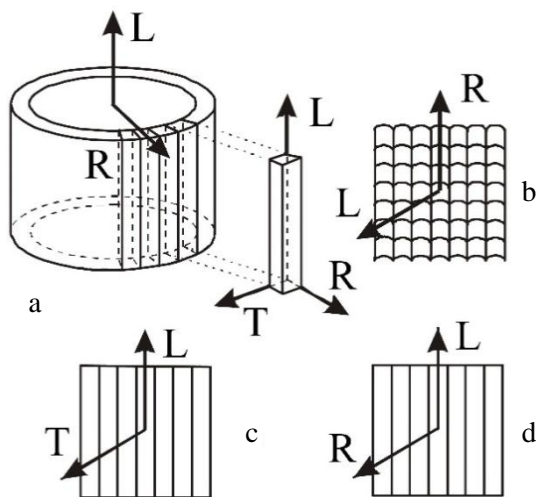
character of the deformed material substructure determines the inhomogeneous development in its recovery processes, recrystallization and phase transformation (PT) during the subsequent heat treatment [10, 18-19].

In this paper, the authors have summarized the results of own researches directed at systematization of the new experimental results, obtained using the developed X-ray methods for investigation of the substructure of textured materials. The processes of structure forming of products are considered in connection with development of their crystallographic texture, what allows to deviate from traditional one-dimensional description of product structure and to characterize it by using of distributions of recorded parameters, giving a much more complete understanding of the studied material.
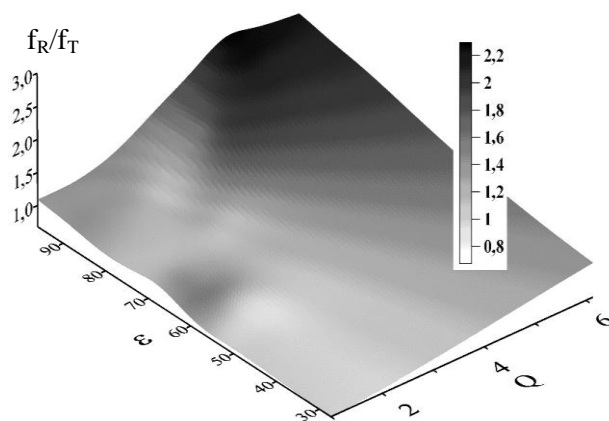
2. The used methods for studying of the crystallographic texture and substructure of textured materials

Analysis of the crystallographic texture and structure of products from Zr-based alloys was carried by using domestic and foreign diffractometers: DRON-3, DRON-3M, D500/TX (manufactured by Siemens) and D8 DISCOVER with LYNXEYE position-sensitive detector (Bruker's production). Filtered or monochrome radiations of chromium, cobalt and copper were used for measuring texture. The detailed description of the method of crystallographic texture analysis by means of direct pole figures (DPF), features automated recording and calculation of partial and complete DPF, taking into account defocusing of X-ray beam, the estimation of measurement errors and DPF construction are presented in [20]. As a rule, samples by dimensions 3×3 mm^2 or composite samples 17×17 mm2 were made (Figure 1). In the case of measuring the texture of the sample of small size it is assumed that the entire surface is within the primary beam, that allows correcting for defocusing analytically, taking into account only change of the test volume by decreasing the depth of the beam penetration.

For a full description of the orientation of the hexagonal crystallite, it is sufficient to know exactly the orientation of the basal normal [0001] and the angle of rotation of the lattice about it, i.e. to assess the crystallographic texture of a polycrystal the construction of two complete DPF (0001) and $\{10\bar{1}0\}$ or $\{11\bar{2}0\}$ is enough. Taking into account the features of the X-ray spectrum of α-Zr the texture of Zr-based alloy products are characterized by DPF (0001) and $\{11\bar{2}0\}$.

Figure 1. The production of composite samples by size 17×17 mm^2 cut out from tube for X-rays study of its texture: *a* – segment of the tube; *b* – *L*-sample; *c* – *T*-sample; *d* – *R*-sample. The test surfaces of composite sample are perpendicular to the corresponding axes of the tube: radial (*R*), tangential (*T*) and longitudinal (*L*).

Figure 2. Change of the ratio of integral texture parameters with decreasing of cross-sectional area of tubular billets ε_Σ and varying of Q-factor at their cold rolling

To calculate the integral parameters of texture, also called Kearns parameters or *f*-parameters, it is necessary to build complete DPF (0001). During recording the one section of the sample, only partial DPF can be constructed, due to the geometry of a standard method of texture registration called "on reflection". This disadvantage can be compensated by different methods of constructing complete DPF from partial DPFs, described in detail in [10, 20]. Two main methods are as follows:

1) mutual combination of three partial DPFs obtained for three mutually perpendicular-sections of the product (see Figure 1);

2) extrapolation of partial DPF on uncharted area of stereographic projection.

We used these methods as the most convenient and reliable. They mutually complement each other. The method of mutual combination cannot be used in case of increased layerwise heterogeneity of the investigated products and in the case of impossibility of preparing samples for all three mutually perpendicular cross-sections (see Figure 1). At the correct using of these methods, the CDPFs constructed with their help coincide. In papers [10, 20] the limits of applicability of the different methods of building CDPF (0001) are considered.

For the purpose of study of structure inhomogeneity of the investigated material the generalized direct pole figures (GDPFs), presenting distributions of the diffraction or substructural parameters of subgrains on stereographic projection depending on orientations of the reflecting surfaces, were build [21-25]. The GDPF construction was carried out by registration of X-ray line profile during measuring of texture for each successive sample position by using a scanning motion of a point detector, or by using a position-sensitive detector. Availability of GDPF allows us to build the distributions of volume fractions of α-Zr grains, differing by values of substructural characteristics.

Investigations were carried out at the industrial or model low-alloyed zirconium-based alloys characterized by a small number of additional phases (β- and intermetallic phases). They include alloys of Zr-Nb, Zr-Fe, Zr-Cr, Zr-Nb-Sn-Fe systems, also alloys additionally doped with oxygen. Paper [20] describes how additional phases influences on features of forming rolling texture.

3. The main stages of the formation of the tubes texture

The final texture of tubes is determined by processes of plastic deformation and heat treatment at successive stages of their production, i.e., during hot and cold deformation and intermediate annealings. Processes of the texture formation at different stages of the manufacturing of tubes are considered in [4, 26-28]. The stress state in sheets under rolling are usually described by the tensor, which is characterize by two components - tension in the rolling direction (RD) and contraction in the normal direction (ND). In the case of pressing, forging or rolling of tube billets along with compression in the radial direction (R) of tube and stretching along its axis (L) the component in the tangential or circumferential direction (T) is added, due to the shape of the product. Therefore, for the description of the stress state in the tube during deformation authors of papers [2, 29] introduced Q-factor ($Q = (\Delta t/t)/(\Delta \overline{D}/\overline{D})$, where t – wall thickness, $\overline{D}$ – average diameter of the tube), allowing to estimate the compressive stress along the R and T-directions. The additional component of the stress in the tangential direction of the tube allows controlling the formation process of its crystallographic texture.

The initial stages of the texture formation in sheets, described in detail in [10, 30], are remained in the case of deformation of the tube. Therefore, first of all, we introduce the notation of main texture components observed in the products from zirconium-based alloys:

T0 – basal texture, i.e. when basal axes are parallel to the R(ND)-direction (R – radial direction for tubes, and ND – normal direction for sheets);

T1 – basal axes are deviated from R(ND) to L(RD)-direction on angle ψ=15-20°;

T2 – basal axes are deviated from R(ND) to T(TD)-direction on angle ψ=20-40°;

T3 – basal axes are parallel to the T(TD)-direction;

T4 – basal axes are parallel to the L(RD)-direction.

Texture components T3 and T4 can be detected at the product only in that case, when during technological treatments the PTs $\alpha \leftrightarrow \beta$ occur inside the product material. Texture components T1 and

T0 are observed at the initial stages of plastic deformation and may also appear in the case of high-temperature treatments (thermo-mechanical treatment, involving a PTs). The final texture of tubes is texture T2, the deviation angle of basal axes from radial direction may vary from 20 to 90°, i.e. up to texture T3. Texture component T3 especially prepared at the channel tubes for CANDU-reactor. For this reason, the tubes are manufactured by compression at a temperature within the (α+β)-phase region of the state diagram followed by cold rolling up to 20% of strain in the wall thickness and the long annealing (24 hrs) at temperature 400°C. Such a low degree of cold deformation allows to maintain the texture maximum of DPF (0001) near the T-direction and to eliminate the undesirable maximum in the L-direction.

At the step of hot-pressing or forging the textureless material or material with the texture varying from T2 to T3, as it is observed during manufacturing of CANDU tubes, can be obtained depending on the temperature of its deformation. Common for all textures of the extrusion (the hot deformation) is the location of the basal axes within the zone extending along the T-R-T diameter of the stereographic projection of the sample, which is determined by the symmetry of the used strain schemes. Features of the basal axes distribution within the mentioned above zone depend on the nominal technological parameters and structural characteristics of the material determining the preference activation of slip and twinning or noncrystallographic mechanisms of the grain boundary sliding, and also the dynamic interaction described modes [31].

For the cold rolling of tubes, on the basis of numerous data presented in the form of two-dimensional correlation diagrams [10, 32], we built a three-dimensional graph, which associates the ratio of integral texture parameters f_R/f_T with values of total strain over the cross section of the tubes ε_Σ and Q-factor (Figure 2). According to this diagram, with increasing ε_Σ and Q-factor the ratio f_R/f_T increases. Obtained results help us to systematize manifestations of texture heterogeneity of cladding tubes and to identify the causes of its development.
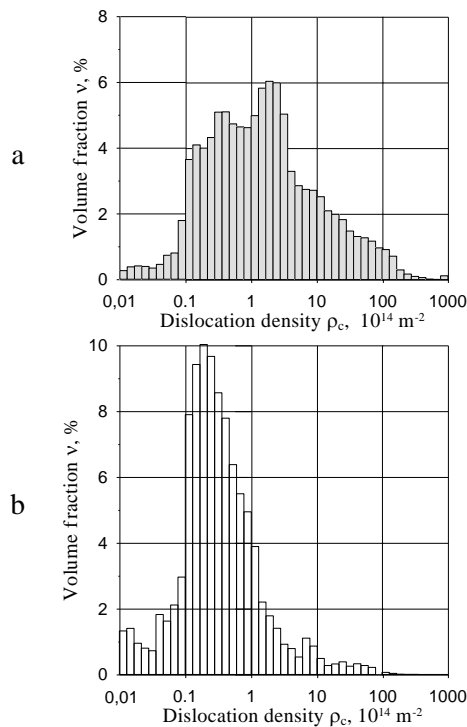
The initial texture of samples is a major factor by means of which the influence of previous treatment is realized on the texture formation under the α-Zr cold rolling. The complex of original orientation predetermines the fraction of grains, in which twinning is activated during the rolling of sample and those that contribute to the formation of the component T1. At the regions where earlier treatment is associated with fragmentation of grains and distortion of the crystal lattice, the activity of twinning and pyramidal slip is reduced. Difficulty of pyramidal slip weakens the components T1 and reduces the stage of its stability. For intermediate degrees of deformation, this corresponds to some deviation of reorientation trajectories of α-Zr grains in the side of the T-direction.

4. Processes of recrystallization and phase transformations inside deformed tube
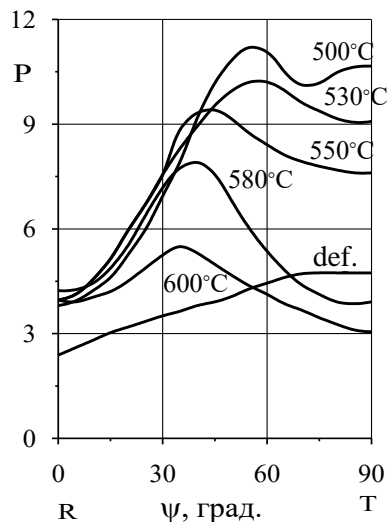
In [33] by means of construction of difference diagrams it was able to demonstrate the preferential growth of the grains, basal axes of which are deviate from its stable positions of the rolling texture. It has been found that the α-Zr matrix regions that were deformed with the prevailing participation of twinning are characterized by a reduced tendency to recrystallization. Based on the established laws of textural changes, recrystallization mechanisms operating in the deformed α-Zr matrix have been revealed. The orientation of recrystallized α-Zr grains corresponds to the sides of deformation texture maxima in which the level of the crystal lattice distortion reaches a maximum and centers of recrystallization arising in these regions can, in the process of their growth, be accompanied by a 30° rotation of the matrix about its basic normal. It has been shown that this rotation of the deformed matrix upon recrystallization annealing of samples only occurs in the case of predominant participation of prismatic slipping in the process of deformation, as well as upon fairly slow heating of samples and in the absence of additional texture components in α-Zr, for example, such as T1+T2.

Similar regularities were obtained for tubes (Figures 3-5). Figures 3-4 show the grain size distribution of the dislocation density in the deformed (Figures 3-*a*, 4) and annealed at 580°C (Figure 3-*b*) tubes from zirconium-based alloys. Strain hardening of α-Zr estimated by the physical broadening of X-ray lines varies in a very wide range and is distributed in such manner that as the distance of the grain orientation from the center of the texture maximum is larger so dispersion and/or
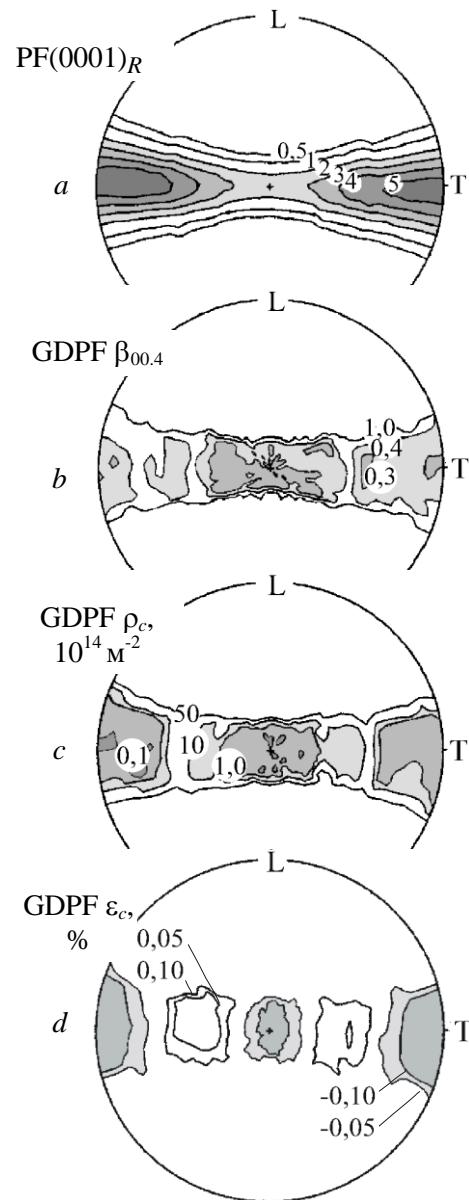
distortion of the crystal lattice is enhanced (Figure 4-*b*). Therefore, the largest crystallites with the least distorted lattice regardless of its type correspond to the central parts of texture maxima, whereas crystallites corresponding to texture minima are the most dispersed and the most distorted lattice.

Figure 3. Distributions of volume fractions $\nu(\rho_c)$ for α-Zr grains of Zr-1%Nb tube 9 mm in diameter:
a – deformed state, ε=80%;
b – annealed state, 480°C / 3 h.

Figure 5. Change of distribution of intensity of X-ray reflectivity (0004) in the R-T-section of tube with increasing of annealing temperature of deformed tube. The annealing temperature is indicated near the corresponding curve.

Figure 4. Complete GDPF for α-Zr of rolled Zr-1%Nb tube 20 mm in diameter, ε=65%: *a* - CDPF (0001); *b* - GDPF β_{0004} in deg.; *c* - GDPF ρ_c в 10^{14} m^{-2}; *d* - GDPF ε_c in percent

State of the material is characterized by a wide range of substructure conditions and must be described by the distribution of substructure parameters. Full distributions of c- and a-dislocation density in the grains of α-Zr plotted for tubes from Zr alloys indicate that the dislocation density depends on the orientations of the grains and varies within a few orders of magnitude, from $\sim 10^{12}$ up to 10^{16} m^{-2} for the rolled tube and up to 10^{15} m^{-2} in the same tube after annealing at 480°C.

Grains with basal axes deviated from the texture maxima are characterized by higher values of the X-ray line broadening $\beta_{(0004)}$ and of the density of c-dislocations ρ_c (Figures 4-b and c). According to a comparison of GDPF ρ_c and GDPF ε_c (Figures 4-c and d), the elastic strain of the crystal lattice in the tube is distributed in strict accordance with dislocation density: compressive elastic strain is characteristic for regions with the lowest density of dislocations and stretching ones is localized in grains with higher dislocation density.

In [34] it was shown that the reorientation themselves basal axes are observed at the recrystallization of α-Zr in the tubes (annealing temperature 580°C) (Figure 5), resulting in that rolling texture with predominant component T3 becomes texture with predominant components T2 (30-50°). The observed growth of grains, the basal axes of which are deviated from the radial direction at 30-50°, is fully explained by the maximum dislocation density in these grains (Figure 5-c).
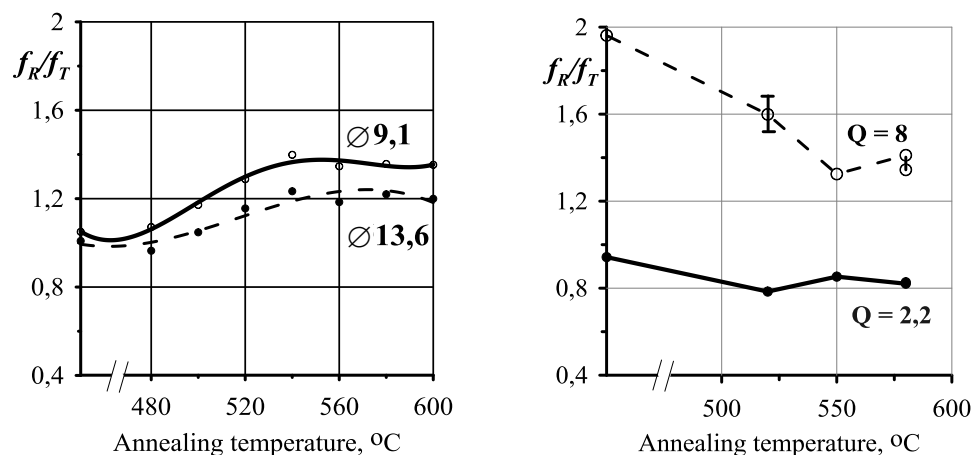
The most widespread opinion, concerning recrystallization of α-Zr, consists in assertion that the main mode of its lattice reorientation comes to 30°-rotation about basal axes [5, 35]. But, the real situation is more complicated; in particular, under recrystallization of α-Zr its basal axes rotate as well [7]. Recrystallization of α-Zr, connected with the rotation of its crystalline lattice by 30° about basal axes, takes place only in the case, when the prismatic slip participated in previous plastic deformation. The greatest number of recrystallization nuclei forms in α-Zr grains, plastically deformed by operation of different slip systems, as in crystallites, corresponding to stable orientations of the rolling texture. The increase of the heating rate to reach the annealing temperature and the presence of additional texture components, such as T1 + T2 [10, 30], also prevent the rotations of the prismatic axes.

The findings suggest about realization in the material of two fundamentally different mechanisms of recrystallization. Firstly, in the absence of 30 ° rotation of prismatic axes, i.e., at the maintaining of the deformation texture, recrystallization nuclei are formed in the body of the deformed grains with orientation preserved strained matrix. Secondly, the rotation of prismatic axes means nucleation of perfect structure at the junction of neighboring grains, which are then completely absorbed deformed matrix, due to the increased rate of migration of the borders with misorientation angle of 30 ° [35]. Tendency of α-Zr grains to recrystallization, involving a rotation of the crystal lattice around the basal normal depends on the orientation of this normal.

At the final stage of manufacturing of cladding tubes of the finishing size the annealing allows to change the value of the f_R-parameter at 0.05-0.10 by varying the pole density of the basal normal near the R-direction and shifting of texture maxima (Figure 7). Besides, it is possible both increase and decrease f_R-parameter determining by the initial texture of the cold rolling and used at the same time the value of Q-factor [32].

PTs α→β in cold-rolling tubes from zirconium-based alloy also develop non-uniformly, including the mutual absorption of grains with different orientations, as evidenced by the results described in [36]. The grains with orientation deviated from the maximum of initial rolling texture primarily undergo to PT α→β. The essential shift of the texture maxima relatively to their initial position is observed. According to findings, these orientations belong to the areas of the higher strain hardening. A feature of texture changes in the tubes at PFs α→β→α is to strengthen the textural component with the tangential orientation of the basal normal.

At the heating of cold-worked tubes above the temperature of PTs, competition between recrystallization and α→β PT arises in the case of sufficiently high strain hardening. As a result, the heterogeneous structure consisting of grains, which have undergone α→β PT from the deformed and recrystallized states, organizes in materials. The extremely heterogeneous structures could be observed in the weld zone obtained by arc welding of cold rolled sheets [36].

Figure 7. Change of the ratio of integral texture parameters during recrystallization of tubes of different diameters (*a*) and deformed with using different values of Q-factor

5. Conclusions

Creation of a complex of new methods of X-ray studies of reactor zirconium-based alloys, taking into account the existence in them developed crystallographic texture and the associated substructure inhomogeneity, as well as the development of methods of their most complete description of them by distributions of diffraction and substructural parameters allowed to systematize new experimental results:

1) regularities of formation of texture and substructure inhomogeneity in the deformation of zirconium alloys in a wide temperature range, the corresponding β-, (α + β) - and α-region of the phase diagram were identified, and at the same time acting deformation mechanisms were refined;

2) regularities of texture changes during recrystallization and phase transformations in tubes from zirconium alloy with a real polycomponent texture were established;

3) effect of different parameters of technological process on the texture formation was identified.

Acknowledgements

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