Recrystallization of cladding tubes from Zr-based alloys for nuclear reactors

M G Isaenkova1,*, Yu A Perlovich1, V A Fesenko1 and O A Krymskaya1

1National Research Nuclear University MEPhI, Kashirskoe shosse, 31, Moscow, 115409, Russia

*Corresponding author’s e-mail: isamarg@mail.ru

Abstract. This paper is devoted to the regularities of change in the crystallographic texture and the structure of cladding tubes with a total degree of deformation by their cross-section of 70-80% as a result of polygonization or recrystallization during heat treatment in the temperature range from 480 to 600 °C. It is shown, that there are two competing processes of crystal lattice perfection in cladding tubes at annealing temperatures over 540 °C. The oriented growth of grain nuclei, formed at the boundaries of grains and disoriented with respect to the deformed matrix by an angle of 20 - 30° about basal axes, is predominant. At the recrystallization of cladding tubes from E110 alloy, a significant change in their crystallographic texture of cold rolling is observed, i.e. an increase of the integral texture parameter fR increases, and a decrease of fT. The grains, formed at the stage of plastic deformation by twinning, prove to be unstable even at the stage of polygonization, due to the mobility of highly disoriented boundaries under conditions of the anisotropic thermal expansion of neighboring regions.

1. Introduction

Heat treatment is the final stage in the fabrication of cladding tubes from zirconium-based alloys for nuclear reactors. Such treatment is accompanied by partial or complete recrystallization of the alloy, the structure of which is determined by the degree of the preceding deformation, the features of the original texture, the heating rate and the annealing temperature, and also the stressed state at the final stage of deformation [1-3]. The degree of recrystallization of the material is an important factor, which controls the level of operational properties and their anisotropy, because the crystallographic texture is significant changes during its recrystallization [4-7]. In some case at heat treatment of cladding tubes, a substantial reorientation of the basal axes are observed [8-9]. This reorientation is associated with an inhomogeneous distribution of a work hardening in grains of different orientations relative to the unstable position of texture maxima [10-12]. However, the mechanisms of the recrystallization of cladding tubes are very diverse and have not yet been finally established. Bozzollo's paper [13] states that the primary nuclei of new grains grow from perfect regions located within deformed grains, and therefore they are characterized by the same orientation as the deformed matrix. But, during the experiments the samples of the deformed metal are heated with high rate and then they are aged for 15-60 minutes. At high heating rates, the mechanism of the annealing texture formation in the deformed material is fundamentally different from the case of industrial heating of tubes in the furnace. In addition, short-term anneals do not allow to create a structure stable under the conditions of exploitation.

Thus, the aim of this paper is to identify recrystallization mechanisms in rolled cladding tubes by results of texture and structure changes in the course of their industrial annealing. At that, the proposed interpretation of the process is in accordance with the regularities of structure and texture formation by recrystallization described in the literature for alloys based on hexagonal metals such as Ti, Zr and Hf.
2. Studied specimens and used methods

Studied cladding tubes from Zr-1% Nb alloy of two final sizes, described at the table 1, have been annealed during 3 hours at the different temperatures from 480 to 600 °C. The annealing time has been chosen in accordance with the results of [14], showing that textural changes cease, when a duration of annealing exceeds 3 hours.

The outer diameter and wall thickness of the tubes studied, the Q-factor and the degree of deformation, calculated from the cross-sectional area of the tube at the final stage of fabrication are presented in the table 1. Also, the table 1 contents the texture integral f-parameters, the full width on half maximum (FWHM) of X-ray peak (2133), and the degree of recrystallization for some annealing temperatures. Q-factor is equal the ratio of variations of the tube wall thickness to its average diameter under plastic deformation and characterizes the stress state of tube at the process of its deformation.

| Number | D x t, mm | Q | e%, % | T\text{anneal}, °C | f-parameters | FWHM (2133)_r, deg. |
|--------|-----------|---|--------|-------------------|--------------|---------------------|
|        |           |   |        |                   | R | T | L | r        |            |
| 1      | 13.6 x 0.9| 2.5| 76.5   | -                 | 0.469 | 0.465 | 0.066 | 0.04 | 1.704 |
|        |           |   |        | 480               | 0.459 | 0.476 | 0.065 | 0.06 | 0.421 |
|        |           |   |        | 540               | 0.523 | 0.424 | 0.053 | 1.67 | 0.432 |
|        |           |   |        | 600               | 0.517 | 0.431 | 0.053 | 3.83 | 0.355 |
| 2      | 9.13 x 0.6| 1.6| 80.7   | -                 | 0.485 | 0.462 | 0.053 | 0.01 | 1.319 |
|        |           |   |        | 480               | 0.494 | 0.461 | 0.045 | 0.06 | 0.369 |
|        |           |   |        | 540               | 0.558 | 0.399 | 0.043 | 1.85 | 0.313 |
|        |           |   |        | 600               | 0.548 | 0.405 | 0.047 | 3.66 | 0.335 |

The scheme of specimen preparation, used for X-ray study of the structure and crystallographic texture of tubes, is shown in figure 1. The cladding tubes (a) are cut into separate segments (b), which are glued to a flat plate so that a composite sample is obtained. The width of the segment depends on the diameter of the tube. The composite sample is grinded to a flat surface and then is chemically etched on 30 μm for removing the layer of work hardening.

For texture analysis, the studied surface of samples was perpendicular to the radial direction (R) (figure 1-c). In order to estimate the degree of recrystallization using the ratio of intensities of X-ray peaks (1120) and (1010), a cross section perpendicular to the axial direction of the tube (L) was prepared (figure 1-a).

X-ray analysis was carried out using DRON-3 and DRON-3M X-ray diffractometers of Russia manufacture, as well as D8 DISCOVER of Bruker manufacture using filtered copper or chromium radiations. Diffraction spectra of the investigated tubes were used for estimation of the recrystallization degree, parameters and stress state of crystalline structure. The texture direct pole figures (DPFs) were recorded in accordance with the standard method of sample tilting [15-16]. On the basis of the (0002) reflection measurement up to 80° of the sample tilt and using extrapolation to the undetermined region of the stereographic projection, the complete DPF (0001) was constructed and used for calculation of the Kearns integral texture parameters: f_R, f_T and f_L, where T is the tangential direction in tube [17]. Orientation distribution functions (ODF) were calculated by incomplete DPFs (0001), {10\overline{1}0}, {11\overline{2}0} and {10\overline{1}2} by using standard software LABOTEX [18-19].

![Figure 1. Preparation scheme of specimens: a - the initial tube and specimen for study of its L-section; b - the segment of tube; c - the composite R-specimen.](image-url)
The grain size was estimated by the method of X-ray intensity fluctuations in the process of shooting the DPF \{11\overline{2}0\}, described in detail in the work [20]. The degree of recrystallization was estimated from the ratio of the intensities of the X-ray peaks (11\overline{2}0) and (10\overline{1}0), taking into account their ratio in the textureless sample. Lattice parameters were calculated by peak positions of X-ray peaks (21\overline{3}\overline{3}) and (30\overline{3}2).

3. Experimental results

3.1. Structural state of samples

Changes in lattice parameters of the crystal structure as a result of the annealing of the studied tubes are shown in Fig. 2. These plots prove, that already at a temperature of 480 °C the crystalline structure undergoes recovery to the equilibrium state of \(\alpha\)-Zr, owing to the anomalously high self-diffusion coefficient of Zr [21]. The parameters of the structure are practically preserved at all other annealing temperatures (figure 2).

The increase of the annealing temperature causes a perfection of the crystalline structure of the deformed grains, about which we are judging by the FWHM of the X-ray lines (see table 1, figure 3). It is known [1-3, 9, 13], that as a result of recrystallization of Zr-based alloys, a crystal lattice of recrystallized \(\alpha\)-Zr grains rotates about the basal normal with respect to the deformed matrix. By the study of considered processes, X-ray peaks (11\overline{2}0) and (10\overline{1}0) were recorded for the cross section perpendicular to the tube axis L and the ratio of their intensities \(r=I(11\overline{2}0)/I(10\overline{1}0)\). The last parameter allows to estimate the rotation of the hexagonal lattice about the basal direction. Table 1 contains the values \(r\) for some annealing temperatures, and figure 4 shows the change in the recrystallization degree \(r\) depending on the annealing temperature of the investigated cladding tubes.

![Figure 2](image1.png)  
**Figure 2.** Changes of parameters of \(\alpha\)-Zr crystal structure by increase of the annealing temperature.

![Figure 3](image2.png)  
**Figure 3.** Changes of X-ray peaks’ FWHMs by increase of annealing temperature (by an example of tube with external diameter 13.6 mm)

Figure 5 illustrates the dependence of the grain size on the degree of recrystallization of a zirconium alloy. From the given graphs it follows, when an annealing temperature is higher than 540 °C and \(r > 2\), recrystallization by rotation of the grains lattice about basal normals of the deformed matrix by 30° is observed and a stable grain size is varied in a narrow range from 2.5 to 3.5 μm.

3.2. Crystallographic texture of annealed tubes

A typical changes in the ODF section with an Euler angle \(\varphi_1 = 0\) at an increasing of the annealing temperature of cold-rolled tubes, having an external diameter of 13.6 mm, are shown in figure 6. Figure 7 illustrates the dependence of the change in the ratio of f-parameters on the annealing temperature of the studied tubes, differing by the technological characteristics of their deformation at the final stage.
Figure 4. Rise of recrystallization degree $r$ by increase of annealing temperature of studied tubes

Figure 5. Correlation of grain size $d$ from recrystallization degree $r$ of studied tubes

Figure 6. Change in ODF-section for $\Phi_1=0$ by increase of annealing temperature of cladding tubes:
(a) rolled tube; (b) 480 °C – 3 h; (c) 540 °C – 3 h; (d) 600 °C – 3 h

4. Discussion of obtained results

The following discussion of X-ray data, presented in the given paler, is based on analysis of revealed regularities of structure and texture formation in zirconium alloys by recrystallization. The first of all, it is the improvement of the crystalline structure of the deformed alloys, when the parameters of $\alpha$-Zr unit cell attain the level, typical for their stable state. At that, the parameter $a$ decreases, while the parameter $c$ increases (figure 2), as well as the sharpening of X-ray peaks, coming down due to active diffusion processes, the drop of dislocation density, and, finally, the growth of grains with new orientations (figure 3).

Secondly, it is the weakening of the texture maximum in the ODF-section (compare figure 6-a and 6-b), corresponding to the orientation of basal normals along the radial direction. Grains with basal normals, oriented near the radial direction are localized within twinning regions of the deformed matrix [1, 3], which were formed during the final deformation of a tube with a high $Q$-factor. Analysis of texture formation at successive stages of tube manufacturing [22] allows us to state that before the last rolling the basal normals of grains, appeared near the radial direction were oriented near the tangential direction. Therefore, the misorientation of the twinned grains relative to neighboring regions of the deformed matrix can reach $85^\circ$ and cause significant stresses during heating of tubes for heat treatment. Part of twins can decrease in size as a result of the arising high stresses caused by the anisotropy of the thermal variation of the linear grain sizes along the $a$ and $c$ directions. In this case, the twinned volume can return to its initial orientation, which we observed in figures 6-a and 6-b.
Namely, as a result of annealing at temperature of 480 °C, i.e. when polygonization takes place, there is an increase in the pole density of the basal normals near the tangential direction and its decrease near radial direction.

In the case of intensive growth of new grains misoriented relative to the deformed matrix by rotation around the basal normals, a sharp changes in the ODF-section are observed (compare figures 6-a and 6-c or 6-d). At the same time, a sharp drop in the broadening of the X-ray line (1120) characterizing the grains of the new orientation is noticeable, while improvement of grains with the former orientation occurs at a lower rate of perfection (figure 3). It is likely that this is the reason for the subsequent absorption of distorted regions, in which only the process of structure polygonization occurs, by more perfect recrystallized grains.

It was shown in [13] that in the case of short-term annealings, conducted to establish mechanisms of recrystallization in Zr and Ti, at the initial stage, perfect regions formed in the deformed matrix, they maintain their orientations by means of the migration of defects to sinks or of polygonization. However, with an increase in the annealing time, a rotation of the crystalline lattice in the growing grains about basal normals is observed.

According to thermodynamic concepts at the initial moment of heat treatment in the deformed material, the formation of grains of different orientations and by different mechanisms must take place, being determined by local conditions of diffusion processes in the deformed material. The presence of dislocations of various types, their orientations, microstresses that result from the anisotropy of the expansion of grains with different orientations in the heating process, predetermine the orientation of the primary nuclei of the future grains. Nuclei of new grains can appear both in the body of deformed grains and on their boundaries, characterized by greater stored energy. This point of view is shared by the authors of the theories of oriented nucleation and oriented growth of recrystallized grains [4, 22].

Evidence of the formation of new grains, disoriented with respect to the deformed matrix at an angle of 20-30° about basal normals, precisely in the boundary of deformed grains is the results of comparison of recrystallization in two-phase titanium alloys and in zirconium alloys with a two-component texture, where similar grains are absent [3, 23].

At the heat treatment of deformed materials, all possible thermal mechanisms operate simultaneously in the workpiece being processed: recovery and polygonization inside grains as well as the formation of new perfect grains at the boundaries of the initially deformed grains. The presence of two competing processes is observed throughout the investigated temperature range, but at 480 °C the nuclei of new grains, formed at the boundaries, can not grow because of the lack of speed for movement of the 30°-boundaries.

In connection with the above facts, the rate of heating of the sample has fundamental importance for activating of one or another mechanism of recrystallization. As it was shown in paper [1], in the case of accelerated heating (100°/s) of deformed samples from a Zr-based alloys, the growth of nuclei disoriented with respect to the deformed matrix by rotation about basal normals is suppressed.

5. Conclusions

1. In competing processes of crystal lattice perfection in cladding tubes at annealing temperatures over 540 °C, the oriented growth of grain nuclei, formed at grain boundaries and disoriented with respect to the deformed matrix by an angle of 20-30° about basal normals, is predominant.
2. After recrystallization of cladding tubes from E110 alloy, a significant change in their crystallographic texture of cold rolling was observed, consisting in the increase of integral texture parameter $f_k$ and the decrease of texture parameter $f_r$.

3. The grains, formed at the stage of plastic deformation by twinning, prove to be unstable even at the stage of polygonization due to the mobility of highly disoriented boundaries under the conditions of anisotropic thermal expansion of neighboring regions.

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