

## Inhomogeneity of Phase Transformations $\beta \rightarrow \omega$ and $\beta \rightarrow \alpha$ in the Quenched Cold-Rolled Alloy Zr-20%Nb

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### Abstract.

The inhomogeneous character of phase transformations in the quenched cold-rolled alloy Zr-20%Nb was investigated using new X-ray diffractometric procedures based on a position sensitive detector. It was shown that the distribution of strain hardening in  $\beta$ -grains with different orientations controls the development of phase transformation  $\beta \rightarrow \alpha$  and  $\beta \rightarrow \omega$  in the rolled alloy. Derivative phases inherit the substructure inhomogeneity of the  $\beta$ -phase, i.e. their grains are most dispersive and/or have the most distorted crystalline lattice in texture minima, where both phase transformations develop primarily. To explain the obtained X-ray texture data a mechanism was suggested involving local formation of quasi-amorphous boundary interlayers by cold rolling and their crystallization by annealing.

### 1. INTRODUCTION

New technique of X-ray diffractometry allows to elevate structure investigations of metal materials to an essentially higher level. Among other aspects this is true in regard to study of systematic structure inhomogeneity in deformed materials and resulting inhomogeneous development of all processes induced by various heat treatments, beginning with recovery or primary recrystallization and ending with phase transformation (PT).

According to the widespread point of view a deformed metal polycrystal should be considered as a composite, whose components have different structures and properties. When taking into account the inevitable development of crystallographic deformation texture and usual substructure anisotropy of deformed grains, it becomes evident that an effective criterion for systematization of structure inhomogeneities should be connected with the crystallographic orientation of grains. To confirm this idea experimentally, several methods were proposed using the selective character of X-ray diffractometric measurement [1, 2]. Such selectivity is used consciously by diffractometric texture analysis only, when grains with different orientations are brought into the reflecting position successively.

The geometry of diffractometric texture measurements was put in the basis of procedures elaborated to compare some structure features of grains with different orientations. As applied to a number of rolled metal materials, for each orientation of the normal to the reflecting crystallographic plane  $hkl$  a half-width of the same X-ray line was measured as well as changes of parameters of the registered X-ray line in consequence of different annealings. Measured values were plotted in the stereographic projection and considered as description of the strain hardening distribution in the investigated sample. By analogy with the texture direct pole figure (DPF), this distribution was referred to as the half-width pole figure (WPF). The term strain hardening is taken to mean a joint manifestation of lattice distortion and grain dispersity. Combined analysis of DPF and WPF resulted in revealing of the following principles, typical for rolled textured metals:

- within a sample with developed deformation texture the local strain hardening varies over wide limits depending on grain orientations;
- main texture components differ by their strain hardening;
- as the grain orientation shifts from the central part of a texture maximum to its periphery, strain hardening increases.

To explain the observed character of strain hardening distribution a model was developed originating from mechanisms, responsible for texture formation and maintenance of final stable orientations [2]. Some features of inhomogeneous strain hardening and regularities of its development were considered also with reference to *Zr*-alloys [3].

A question arises of whether there is an inhomogeneity of PT in deformed textured metal materials, corresponding to the initial distribution of strain hardening. It is possible to answer this question convincingly on the condition that an adequate experimental technique is developed to register simultaneously DPF and WPF for both initial and derivative phases. Such a rather complex task was solved by using a position sensitive detector (PSD) and mathematical treatment of the registered spectra, including separation of overlapping X-ray lines as well as determination of their parameters. The execution of multiple similar measurements, involving repeated labourconsuming treatment of primary data, required a sufficiently high automatization level.

## 2. MATERIAL AND ITS TREATMENT

For an investigation the alloy *Zr*-20%*Nb* was chosen, having the monotectoid composition and being convenient for modelling of different PT processes. According to the phase diagram *Zr*-*Nb*, the alloy experiences PT  $\alpha \rightleftharpoons \beta$  at the temperature 610°C [4]. The high-temperature  $\beta$ -phase has BCC crystalline lattice and the low-temperature  $\alpha$ -phase - HCP lattice. These phases are connected by the Burgers orientation relationship:

$$(0001)_\alpha \parallel \{011\}_\beta, \quad <1120>_\alpha \parallel <111>_\beta.$$

Besides the stable  $\alpha$ - and  $\beta$ -phases, metastable phases form in quenched *Zr*-alloys. The martensitic  $\alpha'$ -phase has the lattice of the  $\alpha$ -phase, distorted due to an excess on *Nb* atoms, which remained in solution by rapid PT. The transformation  $\alpha' \rightarrow \alpha$  proceeds without noticeable grain re-orientation, causing some small decrease of the ratio  $c/a$  and significant narrowing of X-ray lines. Within the context of this work, differences between  $\alpha'$ - and  $\alpha$ -phases are not of importance, therefore, in what follows, we shall not distinguish these phases.

The metastable  $\omega$ -phase forms either by quenching or by annealing of a quenched alloy. Similar to the  $\alpha$ -phase, the  $\omega$ -phase has a hexagonal lattice; but the ratios  $c/a$  for these phases differ significantly:  $(c/a)_\alpha = 1.593$ , while  $(c/a)_\omega = 0.617$ . PT  $\beta \rightarrow \omega$  obeys the following orientation relationship [5]:

$$(0001)_\omega \parallel \{111\}_\beta, \quad <1120>_\omega \parallel <011>_\beta.$$

The most convenient way to observe PT inhomogeneity

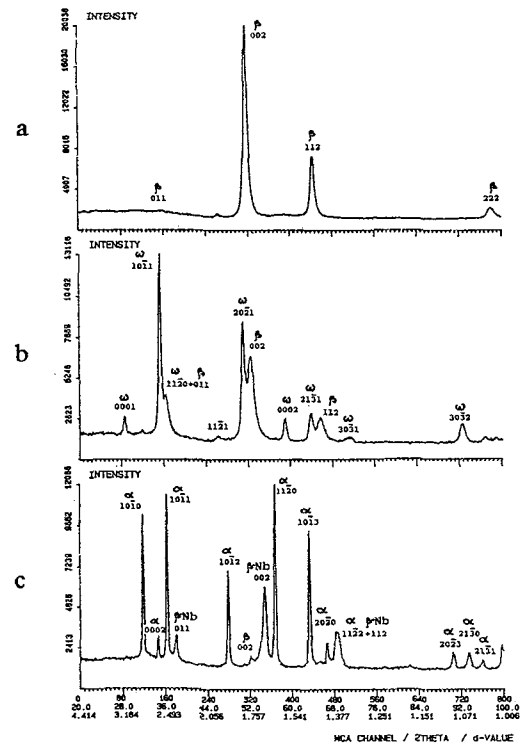


Figure 1: X-ray diffraction spectra for the alloy *Zr*-20%*Nb* in different states: quenching and cold rolling  $\varepsilon=98\%$ (a); annealing at 400°C (b) and 500°C (c) for 10 h.

geneity in the chosen alloy, avoiding X-ray measurements at increased temperatures, consists in the study of PTs  $\beta \rightarrow \omega$  and  $\beta \rightarrow \alpha$  induced by annealing of quenched cold-rolled samples. Quenching of a forged cast from 1000° C was used in order to retain in the alloy the high-temperature  $\beta$ -phase. In order to form in this phase a rolling texture as well as an inhomogeneous distribution of strain hardening the quenched alloy was rolled at 20° C up to a deformation degree of 98%. The obtained 60  $\mu\text{m}$  thick foil was annealed in vacuum at 400° C to produce  $\beta \rightarrow \omega$  PT and at 500° C to produce  $\beta \rightarrow \alpha$  PT. Duration of annealing varied from 0.25 h to 10 h, allowing to study PT kinetics.

On Fig.1 typical X-ray diffraction spectra are shown for the quenched sample in the initial rolled state (a) as well as after annealing at 400° C (b) and 500° C (c). While the spectrum of the rolled sample contains X-ray lines of the  $\beta$ -phase only, the spectra of the annealed samples, along with lines of a residual  $\beta$ -phase and  $\beta - Nb$ , include lines of  $\omega$ - and  $\alpha$ -phases. Most of these lines are overlapping mutually, though, because of the texture, intensities of the lines depend on the sample position and, hence, on the orientation of reflecting grains.

### 3. MAIN FEATURES OF THE EXPERIMENTAL PROCEDURE

The used PSD, being in a fixed position, registers simultaneously a region of the spectrum with an angular width 9°. For the treatment of the measured spectra the following computer programs were applied:

- approximation of a joint profile of overlapping lines with a combination of Gauss and Cauchy functions for cases, when the number of these lines is up to 6 [6];
- calculation of the true physical half-width for all registered lines taking into account their geometrical widening measured by comparison with an annealed textureless sample;
- constructing of WPFs along with DPFs for all X-ray reflections within the registered region of the spectra, when texture measurements are realized by use of PSD technique.

The procedure of mutual division of DPFs was also used. Its results were mapped on the stereographic projection, allowing to compare PT development in grains with different orientations. Thus, in consequence of phase decomposition the general level of DPFs constructed in units of the measured intensity, becomes lower, but this lowering varies from one point to another in accord with PT inhomogeneity. Therefore, distributions obtained by mutual division of DPFs were considered as diagrams of PT inhomogeneity (DIPT).

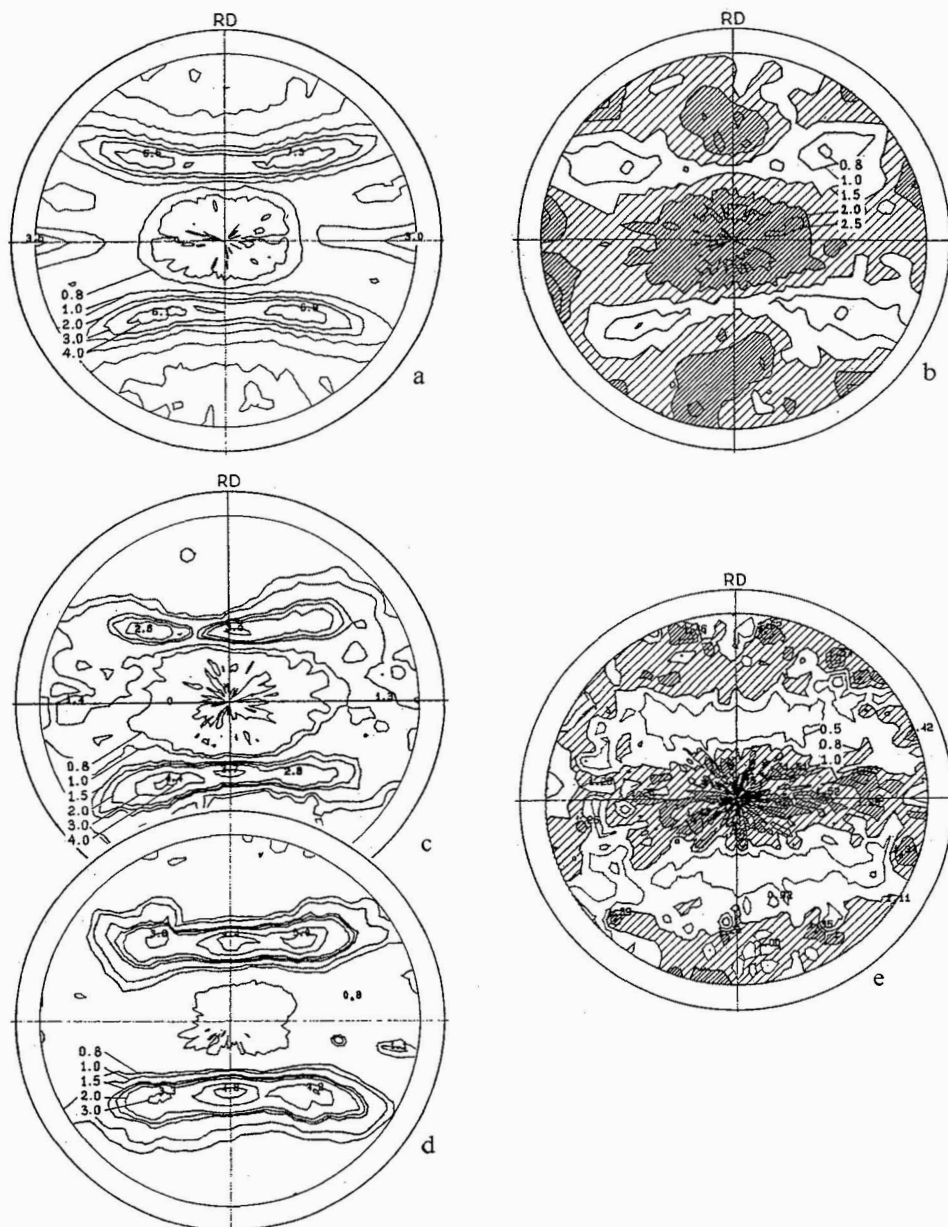
In principle, the obtained data allow to construct the orientation distribution function (ODF) for the present phases. However, the main subject of interest in this work is the influence of strain hardening on PT features, while the measured parameter of strain hardening is referred in the strict sense, to the orientation of the crystallographic normal instead of the grain orientation. Therefore, at the first stage of data analysis, it may be more correct to restrict the consideration to the level of DPF and WPF.

### 4. RESULTS

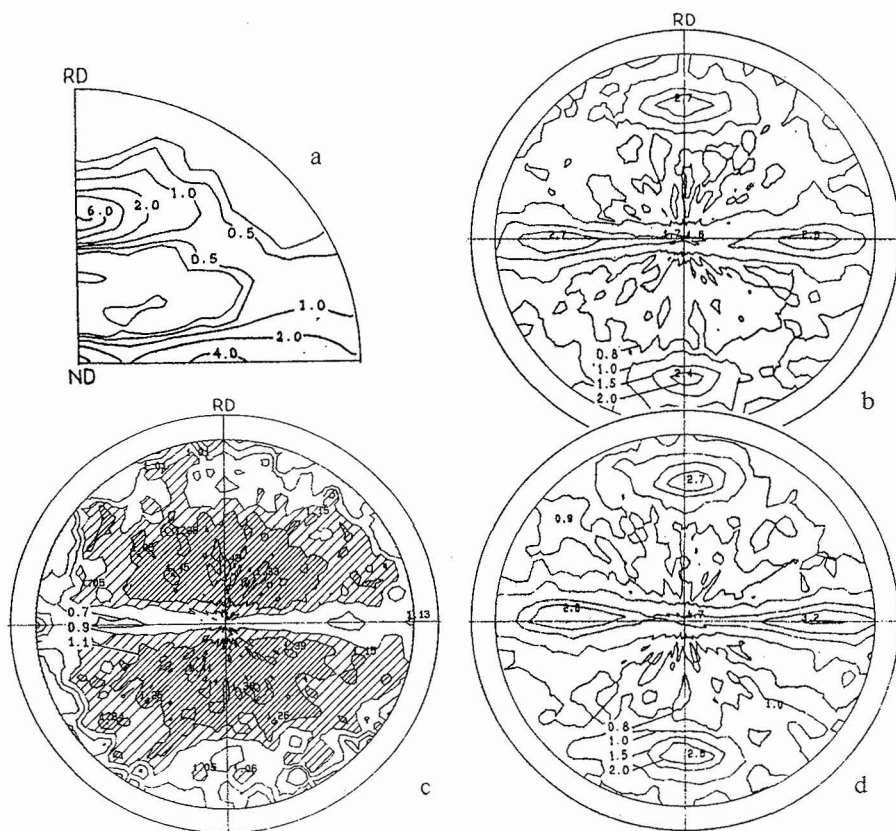
Some typical results are shown on Fig.2 and 3, allowing to carry out a number of informative comparisons of DPFs and WPFs for the initial  $\beta$ -phase and the derivative  $\alpha$ - and  $\omega$ -phases. Presented DPFs and WPFs are partial, having an angular radius of 70°.

a) In accordance with the above-presented orientation relationship, PDF  $(0001)_\alpha$  should be compared with DPF  $\{011\}_\beta$  and DPF  $(0001)_\omega$  - with DPF  $\{111\}_\beta$ . While DPF  $\{011\}_\beta$  (Fig.2-a) could be constructed experimentally from the reflections (011) or (022), DPF  $\{111\}_\beta$  (Fig.3-a) was calculated theoretically using the ODF constructed on the basis of the registered  $\beta$ -reflections by DPF  $\{001\}$ ,  $\{011\}$  and  $\{112\}$  [7]. It can be seen by superposition of DPFs  $\{011\}_\beta$  with  $(0001)_\alpha$  (Fig.2-a, c) and  $\{111\}_\beta$  with  $(0001)_\omega$  (Fig.3-a, b) that the principal positions of the texture maxima coincide though some details of their configuration alter in the course of PT. Hence, formal application of the known orientation relationship is insufficient for a full reproduction of the real texture in the derivative phases.

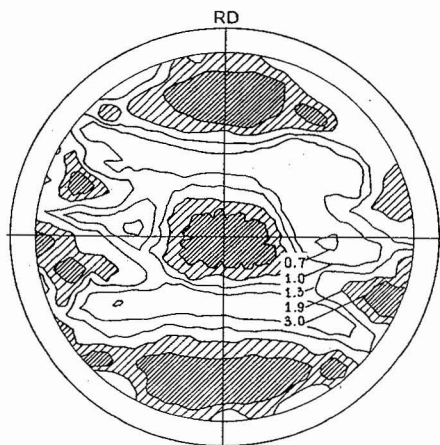
b) Comparison of DPFs  $(0001)_\alpha$  and  $(0001)_\omega$  for samples annealed during 1 h and 10 h at 500° C and



**Figure 2:** PT  $\beta \rightarrow \alpha$ : (a) DPF  $\{011\}_\beta$ , quenching+cold rolling; (b) WPF  $\{011\}_\beta$ , quenching+cold rolling; (c) DPF  $(0001)_\alpha$ , annealing 500°C, 1 h; (d) DPF  $(0001)_\alpha$ , annealing 500°C, 10 h; (e) WPF  $(0002)_\alpha$ , annealing 500°C, 1 h.



**Figure 3:** PT  $\beta \rightarrow \omega$ : (a) calculated DPF  $\{111\}_{\beta}$ , quenching + cold rolling; (b) DPF  $(0001)_{\omega}$ , annealing  $400^{\circ}\text{C}$ , 1 h; (c) WPF  $(0002)_{\omega}$ , annealing  $400^{\circ}\text{C}$ , 1 h; (d) DPF  $(0001)_{\omega}$ , annealing  $400^{\circ}\text{C}$ , 10 h.



**Figure 4:** Ingomogeneity of PT  $\beta \rightarrow \alpha$ :  

$$\text{DIPT}\{011\}_{\beta} = \frac{\text{DPF}\{011\}_{\text{rolling}}}{\text{DPF}\{011\}_{500^{\circ}\text{C}-0.5\text{h}}}$$

400°C respectively (Fig.2-c, d and Fig.3-b, d) gives an indication of the kinetics of PTs  $\beta \rightarrow \alpha$  and  $\beta \rightarrow \omega$ . If PT would be homogeneous, all normalized DPFs for the same phase would be identical while unnormalized DPFs would differ by some coefficient depending on the attained PT stage. Since on Fig.2, 3 normalized DPFs are presented, their mutual differences in the case of  $\beta \rightarrow \alpha$  PT testify about its inhomogeneous character, while similarity of DPFs  $(0001)_\omega$  in the case  $\beta \rightarrow \omega$  PT indicates that PT development is insensitive to the increase of annealing duration from 1 h to 10 h.

c) An evident correlation exists between positions of texture maxima on DPFs and half-width minima on the corresponding WPFs for all three phases (compare Fig.2-a and -b, Fig.2-c and -e, Fig.3-b and -c). At the same time all maxima of X-ray line half-width are localized in minima of pole density. By any means this rule can not be connected with an increase of instrumental errors as the registered intensity decreases. This is obvious from the gradual build-up of registered half-width when passing from the central part of a texture maximum to its periphery. Equally the smooth character of several main contours on WPFs testified that the obtained distribution of half-width represent the real physical regularity rather than random errors of measurements.

On Fig.4 DIPT  $\{011\}_\beta$  is shown, obtained by mutual division of DPFs  $\{011\}_\beta$  for the quenched cold-rolled sample and for the same sample annealed at 500°C during 0.5 h. Maximal values of the ratio on DIPT, testifying about most intensive PT development, are registered in minima of the corresponding DPF. At the same time in some places within the maxima of DPF the ratio takes on values below 1. This fact is connected with the definite increase of registered intensity due to recovery, which was not compensated by an intensity decrease due to PT. In any case, such values of the ratio mark grain orientations appropriated to weakened development of PT.

## 5. DISCUSSION

### 5.1 Inheriting of substructure inhomogeneity

Increased grain dispersion and distortion of crystalline lattice at slopes and periphery of texture maxima, responsible for the registered widening of X-ray lines, seem to be a typical feature of any textured material regardless of texture formation mechanisms. In the deformed alloy with a rolling texture a substructure inhomogeneity of the initial  $\beta$ -phase is caused by different local conditions for the action of plastic deformation mechanisms. In the same alloy in the course of PT the texture of the derivative  $\alpha$ - or  $\omega$ -phase forms from the rolling texture according to the known orientation relationship, while the observed substructure inhomogeneity of these phases is inherited from the  $\beta$ -phase. It appears natural that the more disperse are initial  $\beta$ -grains, the more dispersive are grains of the derivative phases.

In spite of such inheriting, the half-width distribution within the texture minima on WPF  $\{011\}_\beta$  differs by its more regular character from the distributions in texture minima on WPFs  $(0001)_\alpha$  and  $(0001)_\omega$ . This fact indicates that by PT the dependence of substructure inhomogeneity on grain orientation in the derivative phases becomes somewhat weaker, at least in regions with most disperse grains and distorted lattice. Orientation dependences realize more rigidly in dynamic conditions under rolling than in a spontaneous process activated thermally. The investigated volume is sufficiently large to ignore local variations of the strain hardening distribution in the quenched cold-rolled sample, but in cases of the derivative phases this volume is sufficiently small for noticeable violating of the continuous orientation dependence when the number of reflecting grains is statistically insignificant. Therefore, by consideration of WPFs their common main feature, i.e. coincidence of WPF minima with DPF maxima, should be set apart from secondary peculiarities, being sometimes accidental and unreproducible.

### 5.2 Analysis of PT kinetics

Judging from DPFs  $(0001)_\alpha$  on Fig.2-c, d, during the first hour of annealing at 500°C, participation of  $\beta$ -grains with periphery orientations in PT  $\beta \rightarrow \alpha$  was relatively higher than during the following nine hours. This is evident from comparison of the contours, corresponding to the unit pole density on both DPFs. Relative PT rates at central and periphery regions of the texture maxima change as annealing duration increases. This result is supported by DIPT  $\{011\}_\beta$  on Fig.4, characterizing inhomogeneous PT

development at its initial stage. Thus, an obvious correlation exists primarily between strain hardening of the  $\beta$ -phase and its tendency to PT  $\beta \rightarrow \alpha$  at  $500^\circ\text{C}$ . But with time the initial mismatch of tendencies to PT  $\beta \rightarrow \alpha$  in regions with different orientations becomes reverse. As a result, the final texture of the  $\alpha$ -phase appears to have the same perfection as the original rolling texture of the  $\beta$ -phase.

As to PT  $\beta \rightarrow \omega$ , presented data do not reveal the analogous difference of tendencies to PT in central and periphery regions of the texture maxima. The most probable reason for this fact consists in the characteristic kinetics of PT  $\beta \rightarrow \omega$  [4], according to which the  $\omega$ -phase forms at definite short-term stages only. Evidently, the stage of inhomogeneous PT  $\beta \rightarrow \omega$  has significantly shorter duration than 1 h. Hence, in order to observe the manifestations of PT  $\beta \rightarrow \omega$  inhomogeneity, it is necessary to use shorter annealings at  $400^\circ\text{C}$ , than those at  $500^\circ\text{C}$  used to produce PT  $\beta \rightarrow \alpha$ .

### 5.3. Arguments in favour of quasi-amorphous interlayers

It should be noted, that in the studied case besides the above-considered PTs  $\beta \rightarrow \alpha$  and  $\beta \rightarrow \omega$ , some indirect evidences of an additional PT are present. This is crystallization of the quasi-amorphous phase, formed locally by cold rolling of the quenched alloy. There is a number of arguments in favour of such phenomenon.

a) The theory of texture formation predicts different modes of lattice reorientation in inner and boundary parts of grains in the course of deformation [1, 2]. While inner parts of  $\beta$ -grains deform in accord with rigid crystallographic regularities of texture formation, near grain boundaries more complex conditions of plastic flow retard attainment and maintenance of stable orientations. As a result, fragmentation of substructure attains its maximal degree within boundary zones, least fragments lose their ability to deform further by means of symmetric crystallographic slip and their orientations change accidentally. These boundary zones correspond to minima and periphery regions of texture maxima on DPF.

b) Development of a rolling texture ends usually by deformation degrees of 70 – 80%. Maintenance of the stable texture in the course of further rolling is possible due to two main mechanisms: crystallographic glide in several symmetric systems and intergranular slip by flat boundaries in the rolling plane. While the first mechanism should cause strain hardening, fragmentation and inevitable scattering of the texture, the latter mechanism is free from similar consequences, but needs quasi-liquid boundary interlayer. In this work the foil of the quenched alloy was obtained by cold rolling up to 98% without intermediate annealings, and though its X-ray lines are extremely wide, the rolling texture remains quite perfect. Therefore, an active participation of intergranular slip in the deformation process seems to be very probable in this case.

c) Measurements of X-ray line half-width for grains with different orientations give results covering a very wide interval of values. In some points within the maxima of WPFs, i.e. within minima of DPF, the true physical widening of X-ray line, calculated by the standard procedure [8], corresponds to particle sizes of 60 – 70 Å and even less. In many cases the width of X-ray line was too large to be measured with a satisfactory precision. These experimental observations testify that locally a grain dispersity lies outside the sensitivity of X-ray methods and can be extrapolated correctly to a quasi-amorphous state. The volume fraction of material in such state is relatively small and it is invisible practically for direct observation, giving some background component within all regions of DPF. But the contribution of this component to the maxima of DPF is negligible, whereas it is responsible for a significant part of registered intensity in the minima of DPF.

d) A decisive point in support of quasi-amorphous interlayers can be obtained by detailed analysis of intensity changes within the minima of DPF in consequence of low-temperature annealings. Recovery of the quenched cold-rolled sample at  $150^\circ\text{C}$  results in an increase of the registered intensity within the texture minima by a factor of 1.3 – 2.4. For a Zr-alloy this temperature is too low to induce some grain reorientation or PT, therefore, observed changes of DPF could be connected only with annealing of point defects. But such a sharp growth of diffracted intensity indicates that the original content of point defects in corresponding  $\beta$ -grains was considerably higher than the crystalline lattice would accommodate without disintegration.

## 6. CONCLUSIONS

1. The use of diffractometric PSD technique and separation of overlapping X-ray lines allow to widen significantly the possibilities of X-ray texture analysis as applied to study PT inhomogeneity.
2. In the quenched cold-rolled alloy  $Zr-20\%Nb$ , both  $\beta \rightarrow \alpha$  and  $\beta \rightarrow \omega$  PTs develop first of all in texture minima of the initial  $\beta$ -phase.
3. The distribution of strain hardening in  $\beta$ -grains with different orientations controls the development of PTs in the rolled alloy.
4. Both  $\alpha$ - and  $\omega$ -phases inherit the substructure inhomogeneity of the initial  $\beta$ -phase, i.e. in texture minima their grains are most disperse and/or have the most distorted crystalline lattice, while at texture maxima lattices of  $\alpha$ - and  $\omega$ -phases are relatively perfect.
5. On the basis of the obtained data a mechanism was suggested involving the formation of quasi-amorphous interlayers within boundary zones under cold rolling and their crystallization at the initial stage of annealing.

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