Optimization of the procedure for determining integral texture parameters of products from zirconium-based alloys using the orientation distribution function

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Abstract. Integral texture Kearns parameters, or $f$-parameters, are the one of the characteristics that is controlled during the tube production of Zr-based alloys now. These parameters define the anisotropy of physical and mechanical properties of the products. $f$-parameters are the sum of effective fractions of basal normals aligned in a selected sample direction. However, so far there is no standardized procedure for their calculation. The widespread availability of X-ray diffractometers, the high statistical significance of the results and the software development for texture analysis causes the optimality of the $f$-parameters determination by complete pole figures (0001) calculated through orientation distribution function (ODF). Accuracy analysis of the ODF extraction by several incomplete pole figures using LaboTex software was carried out in this paper. The comparison of the calculated data was performed with experimental complete pole figures (0001) obtained by "sewing" method. Various texture types, different number and combinations of incomplete pole figures were considered. For this purpose, 6 incomplete pole figures – (0001), {1120}, {1122}, {1012}, {1013}, {1010} – were registered for each of three mutually perpendicular surfaces of the CANDU-tube sample. By comparison of experimental and calculated by ODF complete pole figures as well as $f$-parameters, procedures of their determination were optimized for products from Zr-based alloys.

1. Introduction
At the present time one of the characteristics, controlled during manufacturing of cladding tubes of Zr-1% Nb alloy, are integral texture Kearns parameters, or $f$-parameters, which define anisotropy of different mechanical properties [1]. These parameters are the sum of the effective fractions of basis normals (0001), oriented along any selected direction in the sample. They are determined by complete direct pole figures (PF) (0001) [2]. However there is still no standardized procedure for constructing complete PF. Among the most common methods: extrapolation of the incomplete PF data to an unregistered region; "sewing" of incomplete PF registered for three mutually perpendicular surfaces; the calculation through the grain orientation distribution function (ODF), which in turn can be constructed from several incomplete PF or electron backscattering diffraction data (EBSD) [3-4].

Despite widespread occurrence in recent decades of EBSD method for direct measurements of the volume fractions of grains, having specified orientations, and for ODF calculation, it is very local and therefore cannot be used for the products characterizing. Moreover, the reconstruction error of the grain boundaries by Kikuchi lines when analyzing deformed samples do not allow to study the texture
evolution of cladding tubes at various stages of their thermomechanical treatment, what is necessary for optimization of the production process.

At the same time the majority of material science laboratories, both in research institutions and in the production, have X-ray diffractometers equipped with texture devices, which allow to perform registration of incomplete PFs. In paper [2], the authors had described in detail the limits of applicability of extrapolation methods and "sewing" for the complete PF reconstruction of products from Zr-based alloys. So the extrapolation method can not be applied in the case when basal normals are oriented outside of the experimentally measured PF area. The results of the "sewing" are affected by the layered inhomogeneity of the sample. One more disadvantage of this method is the necessity to prepare three mutually perpendicular surfaces of the product that associated with certain difficulties in the case of thin-walled cladding tubes of small diameter.

In papers [5-6] the attempts were made for the comparison of f-parameters calculated by the complete PFs, which were constructed using mentioned methods. But the authors have not analyzed the features of ODF, reconstructed by different number of incomplete PFs, which could significantly influence on the f-parameters calculation. Moreover, in paper [6] during samples preparation of rod only the section perpendicular to the transverse direction (T) had been strictly kept, while two other cross-sections, perpendicular to the longitudinal (L) and radial (R) directions, were not fixed, so f-parameters for these sections are inaccurate. Furthermore, the authors didn’t take into account during ODF-reconstruction all possible incomplete PFs, which can be obtained for zirconium alloys.

The construction of complete PF through ODF has its limitations and drawbacks, especially as regards the accessibility of required number of incomplete PF for experimental registration. Therefore, in this study in order to optimize the methodology of the f-parameters determining the results of their calculation by complete PF, reconstructed through the ODF using a specific set of initial incomplete PF, were analyzed.

2. Experimental data
In the absence of layered inhomogeneity of products, as the more accurate method for the complete PF reconstruction can be considered "sewing" because in this case the construction is implemented entirely on the basic of experimental data. Therefore, the error analysis of the ODF calculation and f-parameters determination was conducted by complete PFs obtained by "sewing". For this purpose, for three mutually perpendicular surfaces (longitudinal (L), transverse (T), radial (R)) of the CANDU-tube sample on DRON-3 diffractometer with automatic texture device by using CrKα radiation by incomplete PFs were recorded: (0001), {1120}, {1122}, {1012}, {1013}, {1010} – which available in the spectrum of α-Zr, having a HCP structure. The selection of the sample is determined by its significant difference of the texture obtained for its different surfaces (Fig. 1). This makes it possible to analyze by this example the influence of number of used incomplete PFs, as well as their combinations, on the accuracy of ODF reconstruction. Such an analysis is especially topical for cases of the (0001) PF reconstruction when significant (70-90°) deviation of basis normals from the studied direction occurs (Fig. 1-b), or the presence of additional phase in the material prevents the correct PF (0001) registration.

2. Accuracy analysis of the complete PF reconstruction
The problem of the four-dimensional ODF reconstruction by several incomplete PFs, which are its three-dimensional projections, belongs to the ill-posed class. The uniqueness of its solutions by using any of proposed methods is still questionable. Despite the huge amount of works on accuracy of ODF reconstruction, one of their main drawbacks consists in the determination of error only within the experimentally measured PFs area, which is limited due to geometric features of the X-ray registration with angle 70° when use the "reflection" tilt method.
The quality of the reconstruction is determined by so-called RP-factor \[7\], which is the average deviation of the pole densities \(P_i\) in each point \(i\) of measured region of the stereographic projection by all used PFs:

\[
RP_{(hkl)} = \frac{1}{I} \sum_{i=1}^{I} \left| \frac{P_{\text{exp}}(i) - P_{\text{calc}}(i)}{P_{\text{exp}}(i)} \right| \cdot 100\% ; \quad RP = \frac{1}{N} \sum_{n=1}^{N} RP_{(hkl)}
\]

where

- \(RP_{(hkl)}\) – relative error for each used PF \(\{hkl\}\);
- \(\{P_{\text{exp}}\}(i)\) – experimentally measured \(P\) in the \(i\)-th point of the stereographic projection;
- \(\{P_{\text{calc}}\}(i)\) – calculated \(P\) in the \(i\)-th point of the stereographic projection;
- \(I\) – number of experimentally measured points of the stereographic projection;
- \(N\) – number of incomplete PFs used in the ODF calculation.

The low value of RP-factor does not always indicate the high accuracy of the ODF reconstruction, because the main deviations are concerned with the unmeasured region of PF. Thus, analysis of reconstruction errors is an additional problem of quantitative texture analysis, which solution demands the experience and conception of the types of textures.

In the present work for the ODF reconstruction the commercial program LaboTex, based on the discrete method of ADC (Arbitrarily Defined Cells), was used \[8, 9\]. At that, the analysis of the reconstruction quality was carried out by both RP-factor, used in LaboTex, and the average deviation of calculated \(f\)-parameters from the experimental ones:

\[
\Delta f = \frac{1}{3} \left( |f_R^{\text{exp}} - f_R^{\text{calc}}| + |f_T^{\text{exp}} - f_T^{\text{calc}}| + |f_L^{\text{exp}} - f_L^{\text{calc}}| \right)
\]

The quality of reconstruction was also evaluated visually by the appearance of complete PF (0001).

Many experimental PFs are not always available for X-ray registration, as in case of the \(\beta\)-phase presence in the sample, whose lines may overlap the reflections of \(\alpha\)-Zr. When using the most common X-ray radiation, having wavelength less than CrK\(_\alpha\), the reflection (0002) is overlapped by the line (10\(\bar{1}\)1), and (0004) has a rather low intensity, so there is no possibility to get the basis PF (0001).
Contrariwise for some specific types of textures only PF (0001) has sufficient intensity in the measured region (T-sample), while the others PFs are rather weak within tilt angle of 70°. Therefore, for minimization the number of incomplete PFs, required for the ODF calculation, and optimization of their combination, the influence of total weighting factor $W$ [7] of experimental PFs, used in the concrete calculation, was studied. $W$ is one of the accuracy characteristics of experimental PFs:

$$W = \sum_{n=1}^{N} \sum_{i=1}^{I} \langle P_{\text{exp}} \rangle_{f}^{2} d\omega_{i},$$

where $d\omega_{i}$ is the area of $i$-th element of the unit sphere limited by the tilt angles ($\psi_{i} - \frac{\Delta\psi}{2}$); ($\psi_{i} + \frac{\Delta\psi}{2}$), and rotation angles ($\varphi_{i} - \frac{\Delta\varphi}{2}$); ($\varphi_{i} + \frac{\Delta\varphi}{2}$), ($\Delta\psi$, $\Delta\varphi$ – variation step of the corresponding angles during PF registration).

3. Results

Different combinations of incomplete PFs used for the ODF calculation, are shown in Table. 1 together with total weighting factors, $RP$-values and $f$-parameters deviations $\Delta f$. Combinations of PFs were chosen in a way to maximize the occupation of elementary stereographic triangle for hexagonal lattice. Fig. 2 (a, b) show reconstructed complete PF (0001) for the cases of minimum and maximum $\Delta f$ and $RP$, which are marked in bold in the table 1.

| Table 1. The results of the $f$-parameters determination |
|---------------------------------------------------------|
| Calculation variant | R-sample | T-sample | L-sample |
| $\langle$sewing$\rangle$ | $f_{R}$ | $f_{T}$ | $f_{L}$ | $f_{R}$ | $f_{T}$ | $f_{L}$ | $f_{R}$ | $f_{T}$ | $f_{L}$ |
| (0001) (1120) (1010) (1013) (1122) | 67.1 | 45.1 | 0.013 | 64.9 | 44.9 | 0.022 | 78.7 | 53.2 | 0.024 |
| (0001) (1120) (1010) (1012) (1122) | 67.5 | 55.9 | 0.014 | 64.5 | 43.0 | 0.021 | 77.5 | 59.1 | 0.021 |
| (0001) (1120) (1012) (1013) | 44.1 | 51.7 | 0.018 | 44.2 | 34.5 | 0.022 | 46.1 | 57.7 | 0.026 |
| (0001) (1120) (1013) (1122) | 46.8 | 48.5 | 0.015 | 46.6 | 38.1 | 0.022 | 47.1 | 45.3 | 0.009 |
| (0001) (1120) (1013) (1010) | 57.8 | 38.0 | 0.015 | 56.2 | 42.3 | 0.024 | 68.5 | 48.1 | 0.011 |
| (0001) (1120) (1010) (11.2) | 60.9 | 46.5 | 0.013 | 58.2 | 45.2 | 0.025 | 68.3 | 51.4 | 0.027 |
| (1010) (1120) (1013) (1122) | 54.3 | 45.1 | 0.023 | 48.8 | 45.5 | 0.085 | 69.5 | 63.0 | 0.028 |
| (1010) (1120) (1013) (1012) | 51.6 | 48.1 | 0.014 | 46.4 | 41.7 | 0.090 | 68.5 | 64.6 | 0.026 |
| (0001) (1120) (1013) | 37.5 | 39.2 | 0.019 | 37.9 | 33.5 | 0.027 | 36.9 | 55.9 | 0.013 |
| (0001) (1120) (1012) | 37.9 | 63.4 | 0.021 | 37.5 | 28.6 | 0.032 | 35.7 | 45.9 | 0.028 |
| (0001) (1120) (1122) | 40.6 | 58.4 | 0.017 | 39.9 | 30.7 | 0.029 | 36.7 | 41.7 | 0.020 |
| (0001) (1120) (1010) | 51.6 | 37.7 | 0.015 | 49.5 | 39.8 | 0.028 | 58.1 | 44.3 | 0.031 |
| (1010) (1120) (1122) | 48.1 | 50.6 | 0.024 | 42.1 | 51.6 | 0.075 | 59.1 | 64.8 | 0.032 |
| (1010) (1120) (1012) | 45.4 | 59.0 | 0.025 | 39.7 | 47.2 | 0.071 | 58.1 | 71.9 | 0.031 |
| (1120) (1122) (1012) | 34.4 | 56.4 | 0.017 | 30.1 | 33.4 | 0.033 | 36.7 | 72.9 | 0.023 |
| (0001) (1120) (1122) | 42.4 | 43.4 | 0.023 | 43.1 | 35.1 | 0.030 | 51.0 | 46.4 | 0.028 |
| (0001) (1120) | 31.3 | 47.6 | 0.023 | 31.2 | 20.0 | 0.041 | 26.5 | 24.5 | 0.034 |
| (1010) (1122) | 29.6 | 50.2 | 0.037 | 27.0 | 46.3 | 0.079 | 41.8 | 66.5 | 0.051 |
| (1120) (1012) | 25.1 | 61.1 | 0.030 | 21.4 | 35.0 | 0.068 | 26.5 | 76.4 | 0.038 |
| (0001) (1010) | 33.1 | 28.6 | 0.036 | 34.4 | 26.1 | 0.032 | 40.8 | 34.8 | 0.044 |

As it can be seen from Fig. 2, assessment of the quality at the ODF extraction by widely used $RP$-factor often leads to false results, what can be determined by visual comparison of reconstructed PFs. For example, in the R-sample at minimum $RP$ at the overstep border to the unmeasured area of the stereographic projection a discontinuous variation of the pole density is observed, as well as sharp deviation of the maximum towards the L-direction. Such a PF view is mistaken. However, the experimentally measured value of the error is low. In the case of maximum $RP$ the complete PF is quite smooth, its view is very similar to obtained by "sewing" and the $f$-parameters deviation is much lower.
For T-sample the presence of a weak maximum at PF \{11\bar{2}0\} in the T-direction (Fig. 1-b) in many calculation variants results in the uncertainty of the orientation of basis normals - the appearance of an additional weak maximum in the L-direction. Since most of the additional PFs is quite diffused, the only ODF-extraction with involved PF \{10\bar{1}0\} gives the result without this maximum. However, PF \{10\bar{1}0\} is located at low Bragg angles and therefore its measurement using radiations with less than the CrK_{\alpha} wavelength is associated with loss of accuracy due to the increasing of the defocusing coefficients and the overlapping with the reflection (0002). Furthermore, for this type of texture all calculation variants without PF (0001) result in inaccurate ODF reconstruction. This is due to the fact that the location of the basis normals in the center of the stereographic projection is associated with the arrangement of the maxima in other PFs to the periphery of the stereographic projection. So they do not contribute any significant information about the grains orientation.

The disagreement to the obtained by "sewing" PF can be seen also for the L-sample at both the minimum and maximum \(R_P\) factor. The wider maximum are observed here, and in some cases an additional maximum in the R-direction, which is associated with the ambiguity in case of absence of PF (0001) in the calculation procedure.

The analysis of the PF with the lowest \(\Delta f\) testifies about their maximum similarity with obtained by "sewing" (Fig. 1). Therefore, based on the analysis of table 1 the optimal calculating variant should be selected by the minimum \(f\)-parameters deviation. Fig. 3 summarizes the results of table 1, which analysis shows that an increasing of total weight of the PFs used for ODF extraction does not result in an reducing of \(R_P\)-factor, and in some cases (T-sample) it even slightly increases. I.e. the increasing of total "precision" of used in calculating PFs has no effect on this characteristic, what once again confirms its failure in the assessment of the reconstruction quality.

The deviation \(f\)-parameters (Fig. 3-b) tends to decrease with increasing of \(W\), but with some plateau in the range \(W= 40-60\), which can be considered as optimal values for ODF calculation, because within this range there are calculation variants, having \(\Delta f\approx0.02\), what defined by authors of [2] as the minimum error in the \(f\)-parameters determining. According to Fig. 3-b specified range corresponds to

**Figure 2.** Reconstructed complete PFs (0001) for cases with the minimum and maximum \(\Delta f\) (a) and \(R_P\)-factor (b), as well as for the optimal combination of incomplete PFs (c)
3-4 used PFs. Due to the necessity of their minimization, one should focus on measuring of 3 incomplete PFs.

![Figure 3](image-url)

**Figure 3.** The dependences of $RP$-factor ($a$), $\Delta f$ ($b$) and number of used incomplete PFs $N$ ($c$) on their total weighting factor $W$

The minimum error in the $f$-parameter determining for all possible combinations observed for R-sample, in spite of the fact that the basis normals is significantly tilted from the normal to the studied surface for this type of texture. The minimum $\Delta f$-values for all samples are marked in Table 1 with a red underline, herewith for the T- and L-sample it is the same combination of PFs: (0001) \{1120\} \{10\13\}. Since in the R-sample for this combination $\Delta f$ is slightly higher, then it can be considered as optimal for ODF-extraction.

However, in case of unavailability of one of mentioned PF any of combinations with $\Delta f$ value below 0.03 can be selected. This is particularly actual when there is no opportunity to get basis PF, for example, in case of Zr-alloys with the presence of $\beta$-phase. At that, the main problem occurs during ODF extraction for T-sample, for which even the increase of $N$ does not lead to an accurate $f$-parameters calculation without PF (0001). The most precise option is a combination of \{11\20\} \{11\22\} \{10\12\}, or it may be preferred to calculate the $f$-parameters by extrapolation method in this case [2].

4. **Conclusion**

Influence of the number of incomplete PFs and their various combinations for different texture types of cladding tubes from Zr-based alloy on quality of ODF extraction was demonstrated. The error analysis of $f$-parameters calculation by complete PF (0001), reconstructed through the ODF, with the experimental data obtained by "sewing" method was implemented. The procedure of $f$-parameters calculation for products from Zr-based alloys was optimized.

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