Influence of substructure inhomogeneity on the anisotropy of physical properties of textured materials

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Abstract. Calculation of the physical properties anisotropy is based on single crystal properties and information about material texture, i.e. orientation distribution function (ODF). At the same time, the grains of different orientations are known to have different substructure parameters including the lattice distortion and microstrains. Determination of these parameters is possible with the X-ray method of Generalized Pole Figures (GPF), which consists of registration of the whole X-ray reflection profile under the pole figure recording. As a result, one can obtain the FWHMs of X-ray line profiles and microstrains distributed on the stereographic projection. Despite the observable equilibrium of tensile and compressive microstrains of differently oriented grains within the studied material volume, they can affect the product properties anisotropy. In this study the calculation method for elastic properties and thermal expansion coefficients taking into account substructure inhomogeneity is offered on the basis of elastic energy minimization. Its efficiency is demonstrated for the samples of zirconium alloys.

1. Introduction
One of the main motives for crystallographic texture analysis is its effect on the anisotropy of physical and mechanical properties of materials. The anisotropy calculation of both elastic properties and linear thermal expansion coefficients (LTEC) are essentially based on models that average the single crystals properties with a weight function which is the orientation distribution function (ODF). However, a textured polycrystalline material that has undergone a certain thermo-mechanical treatment is characterized by some range of structural parameters and presence of residual elastic microstrains even in the recrystallized condition [1-3]. Therefore, in addition to the crystallites orientation, the properties are influenced by the substructural inhomogeneity of grains of different orientations.

In this paper we propose an approach for calculation of elastic moduli anisotropy as well as LTECs taking microstrains inhomogeneity into account. The algorithm was tested on a sample from a channel tube of Zr-2.5%Nb alloy, with the main $\alpha$-phase having a hexagonal closed-packed (HCP) structure.

2. Principles of proposed approach
The calculation of the anisotropy of elastic properties and LTEC of textured materials is based on averaging the properties of single crystals with the ODF as a weight function ($f(g)$). Moreover, the assumption of deformations (Voigt model) or stresses (Reuss model) homogeneity in a polycrystal is used for elastic moduli anisotropy estimation [4, 5]. So averaging is implemented either for components of stiffness matrix $[C_{ijkl}]$ or for compliance coefficients $[S_{ijkl}]$. However, already Hill showed that true values of the elastic coefficients are between the values determined by Voigt or Reuss.
Another so-called self-consistent approach for averaging properties developed by Lebenson and Tome [5] consists in considering individual grains as inclusions embedded in a homogeneous medium with averaged elastic properties of the entire polycrystal. In this case, it is assumed that the stresses and total strains (elastic and thermal) are constant in each grain, and the entire polycrystal experiences uniform stresses. This results in the proximity of the Hill model.

The presence of substructure inhomogeneity, i.e. microstresses, and consequently microstrains variation, results in violation of both the Voigt and Reuss models. Moreover, as noted in [4], relationship between different properties should be taken into account to prevent significant errors in such calculations. For example, LTECs define the response of the material on such an external impact as temperature, which results in the development of elastic strains, which can cause stresses between adjacent grains. However the described calculations ignore the combined effect of temperature and stresses, whereas it is necessary to additionally consider the elastic moduli when averaging the LTEC’s anisotropy.

2.1. Elastic moduli anisotropy

Since a polycrystalline material is characterized by a range of elastic microstresses of differently oriented grains, it can be represented as a composite of stretched and compressed “springs”, which are individual grains (figure 1). Despite the stiffness of an individual grain remains constant, due to the difference in their orientation and interaction with neighbours, the stiffness of the entire polycrystal is not equal to the mathematical averaged elastic constants. Thus, when calculating the elastic properties, it is necessary to take into account the stored elastic energy of individual grains, which is equal to [5, 6]:

\[ W_c = \frac{1}{2} \{\sigma_c\} \{\varepsilon_c\} = \frac{1}{2} C_{ijkl} \varepsilon_{ij} \varepsilon_{kl}. \]  

(1)

Values of the crystallite’s elastic microstrains \( \{\varepsilon_c\} \) can be obtained from the Generalized Pole Figures \( \Delta d/\Delta d \) (GPF \( \Delta d/d \)), calculated from the Bragg’s angle of the X-ray reflection profile [1-3]. Then it is possible to find the tensor of elastic stresses in the crystallite, for which the expression in matrix form is [4, 5]:

\[ \{\sigma_e\} = \{C_e\} \{\varepsilon_e\}. \]  

(2)

Averaging tensors of elastic strains and stresses over a polycrystalline aggregate gives [4]:

\[ \{\sigma_s\} = \{f(\varepsilon_c) \cdot f(g) \} dg; \quad \{\varepsilon_s\} = \{f(\varepsilon_c) \cdot f(g) \} dg. \]  

(3)

Now the calculation of the elastic properties of a polycrystal (sample) incorporating the stored elastic energy of individual grains, due to the presence of elastic tension or compression, can be carried out on the basis of the principle of minimizing its elastic energy:

\[ W_s = \frac{1}{2} \{C_s \cdot \varepsilon_s\} \cdot \varepsilon_s \rightarrow min. \]  

(4)

This function is linear with respect to the coefficients \( C_s \) and for their estimation one can use the standard simplex minimization algorithm with constraints of \( \{\sigma_s\} = \{C_s\} \{\varepsilon_s\} \) and the additional condition of \( C_s \) non-negativity. As textured polycrystalline materials no longer possesses the symmetry of its constituent single crystals, we suppose orthotropy of the material properties, i.e. orthorhombic sample symmetry typical for rolling products, which reduce the number of \( C_s \) coefficients to 9. Since the solution of linear programming problems is rather sensitive to the initial approximation, we used the values determined by the Hill model.

Solving linear programming problems of this type is possible by special functions which are implemented in almost any math software. We used the DLPRS-subroutine of IMSL libraries for FORTRAN.
2.2. LTECs anisotropy
The substructural inhomogeneity in textured polycrystalline materials results in the occurrence of initial tensile/compressive microstrains in grains, so during their interaction the deformation character of whole polycrystalline aggregate changes. This affects the LTECs anisotropy as well as the elastic properties.

The LTECs are related to the elastic coefficients through the Grüneisen parameters (γ) \[6\]. Therefore it is possible to calculate the LTECs of a polycrystal using averaging of these parameters for single crystals together with the elastic constants \( [C_e] \) calculated as described above.

LTECs (\( \alpha \)) are related to the elastic coefficients through the relations:

\[
\alpha_{ij} = \frac{C_{ij}}{\nu} \sum_{j=1}^{6} C_{ij} \gamma_j ,
\]

which for the HCP single crystals with lattice parameters \( a \) and \( c \) are described by a 2\(^{nd}\)-rank tensor of a diagonal form in the principal axes:

\[
\begin{bmatrix}
\alpha_a & 0 & 0 \\
0 & \alpha_a & 0 \\
0 & 0 & \alpha_c
\end{bmatrix}.
\]

At near room temperatures, the main contribution to the LTECs value is from the crystalline lattice, so the Grüneisen parameters are also isotropic in the basal plane:

\[
\begin{align*}
\gamma_1 &= \frac{dV}{V} \left[ C_{111} + C_{122} \alpha_a + C_{133} \alpha_c \right], \\
\gamma_2 &= \frac{dV}{V} \left[ 2C_{133} \alpha_a + C_{333} \alpha_c \right],
\end{align*}
\]

where \( c_e \) – heat capacity at constant strain; \( dV \) – single crystallite volume; \( \alpha_c \) and \( \alpha_a \) for Zr-alloys equal to \( 7.4 \times 10^{-6} \) and \( 4.9 \times 10^{-6} \), respectively \[7\].

Since the calculation is carried out for normal conditions and constant temperature, the lattice volume can be considered as constant, i.e. \( \oint dV \cdot dg = V \). So the components of the Grüneisen parameters tensor of a single crystal can be averaged for calculating the polycrystalline Grüneisen parameters \( \gamma_s \):

\[
[\gamma_{sij}] = \oint [\gamma_{ij}] \cdot f(g) dg,
\]

and then determine the LTECs of polycrystal by formula (5) using tensor \( [C_e] \), calculated earlier. The value of \( \frac{c_e}{\nu} \) will cancel out.

3. Samples and research methods
The proposed algorithm was tested on samples from a channel pressure tube \( \varnothing 113.2 \times 103.6 \) mm made of the Zr-2.5% Nb alloy after 20% of cold rolling. The samples are characterized by a sharp texture with a predominant arrangement of basal normals in the tangential direction (T). Figure 2 shows typical pole figures (PF), GPFs (\( \alpha_{250} \)) and distributions of grain volume fractions (VF) by microstrains along various crystallographic directions.

The obtained data confirm the principal laws of substructural inhomogeneity in textured materials formulated in [1-3]: counterbalancing of elastic microstrains within individual texture maxima, which is seen in the GPF (\( \alpha_{250} \)) for the left and right texture components. At the same time the microstrains equilibrium for prismatic directions, \( <11\overline{2}0> \) and \( <10\overline{1}0> \), apparently associated with macroscopic stress-strain state determined by rolling, under which the sample is compressed along the normal direction (R) and elongated along the rolling direction (L). After unloading the inhomogeneity of the stress-strain state in different tube’ layers results in tension along R. However, the balancing of microstrains along basal normals occurs at the expense of grains’ interaction within texture maxima having different origins, i.e. formation due to activation of different deformation systems.
Figure 2. PFs and GPFs \( \frac{\Delta d}{d} \) of the channel tube sample of the Zr-
2.5\%Nb alloy and the distributions of grain volume fractions \( (VF) \) by microstrains.

The crystallographic directions \(<0001>, <11\overline{2}0\text{> and }<10\overline{1}0\text{> represent the basis of the hexagonal lattice; therefore, the set of obtained GPFs \( \frac{\Delta d}{d} \) can be converted into the elastic microstrain tensor of individual crystallites \( \{\varepsilon_c\} \) by assigning proper Euler angles. Assuming the absence of shear components:

\[
\{\varepsilon_c\} = \begin{bmatrix}
\varepsilon_x & 0 & 0 \\
0 & \varepsilon_y & 0 \\
0 & 0 & \varepsilon_z
\end{bmatrix} = \begin{bmatrix}
(\Delta d / d)_{\overline{1}0\overline{1}0} & 0 & 0 \\
0 & (\Delta d / d)_{1\overline{1}20} & 0 \\
0 & 0 & (\Delta d / d)_{0001}
\end{bmatrix} .
\] (9)

Since the construction of the full function of microstrains, similarly to an ODF, is a rather difficult task, in this work extrapolation was carried out to the unregistered regions of Euler angles using available GPFs. Thus, each set of angles \( (\phi_1, \Phi, \phi_2) \) is associated with an ODF value and three components of the tensor \( \{\varepsilon_c\} \).

Figure 3. Typical indentation
loading diagrams \( (a) \) and
dilatometric curves \( (b) \) of the
channel tube obtained for different sample surfaces.

The elastic moduli for different sample’s directions \( (R, T, \text{ and } L) \) were measured by the instrumented indentation method with registration of loading diagrams \[8-10\] using indentometer DNT1/5. Ten measurements were carried out for each surface under a load of 1000 \( \mu N \) with a Berkovich pyramid. Examples of loading diagrams for various sample surfaces are shown in figure 3-a. Based on the obtained data the mean value and dispersion were estimated.
The statistical error of the hardness determining does not exceed 5.5%, and for elastic modulus – 7.8%. The experimental LTECs were estimated by the slope of the temperature $\Delta l/l_0$ dependences for the three directions, obtained using a NETZSCH DIL 402 dilatometer on cubic samples with dimensions of $4 \times 4 \times 4$ mm (figure 3-b).

4. Results

Table 1 and figure 4 show the results of the measurements and calculation of the elastic modulus ($E$) according to both Hill's averaging and the proposed algorithm. In the calculation ODFs obtained from three perpendicular surfaces were used [11]. In addition, computational methods make it possible to determine the elastic modulus not for individual directions, but to construct its distribution over the entire stereographic projection. In this case the elastic moduli for an arbitrary direction $y$ in orthotropic sample is determined by the compliances tensor $\mathbf{S} = [C_s]^{-1}$:

$$\frac{1}{E_y} = y^1_1 \cdot S_{11} + y^2_2 \cdot S_{22} + y^3_3 \cdot S_{33} + 2 \cdot y^1_1 \cdot y^2_2 \cdot (S_{12} + 2S_{66}) + 2 \cdot y^1_1 \cdot y^3_3 \cdot (S_{13} + 2S_{33}) + 2 \cdot y^2_2 \cdot y^3_3 \cdot (S_{23} + 2S_{44}),$$

(10)

where $y_1, y_2, y_3$ are the direction cosines of the studied direction.

The obtained data testifies that calculation of the elastic moduli without taking into account the microstrains inhomogeneity leads to higher deviations from the values obtained by instrumental indentation, the value of anisotropy is significantly lower than the experimental one. In the case of calculations by the proposed algorithm, the anisotropy coefficient is much closer to the experimental one.

**Table 1.** Comparison of the elastic moduli for different directions of the chosen sample obtained by various methods.

| parameter | indentation | Hill model | proposed algorithm |
| --- | --- | --- | --- |
| | | ODF R | ODF T | ODF L | ODF R | ODF T | ODF L |
| $E_R$, GPa | 101.2±6.6 | 97 | 98 | 98 | 99 | 105 | 100 |
| $E_T$, GPa | 113.0±7.8 | 102 | 103 | 103 | 110 | 115 | 114 |
| $E_L$, GPa | 93.3±5.4 | 98 | 98 | 96 | 96 | 97 | 96 |
| $\frac{E_{max} - E_{min}}{E_{max}}$, % | 17.4 | 4.9 | 4.85 | 4.85 | 12.7 | 15.7 | 15.8 |

Table 2 show the results of experimental and calculated LTECs of the studied sample. The obtained data again show that the calculation of the LTECs without taking into account elastic constants gives a lower anisotropy compared to the experimental value, while calculation by the proposed algorithm reveal a large value of anisotropy, which approaches to the experimental data.

**Figure 4.** Dependences of elastic moduli in different sample sections.
Table 2. Comparison of LTECs for different directions of the studied sample obtained by various methods.

| parameter | LTEC \( \times 10^{-6} \), \( \frac{1}{\alpha} \) |
|-----------|----------------------------------|
| \( \alpha_R \) | dilatometry | Bunge formulation | proposed algorithm |
| \( \alpha_T \) | 6.48 | 5.84 | 6.16 |
| \( \alpha_L \) | 7.62 | 6.31 | 6.60 |
| \( \frac{a_{\text{max}} - a_{\text{min}}}{a_{\text{max}}} \) \( \% \) | 5.0 | 5.05 | 4.80 |
| \( \frac{a_{\text{max}}}{a_{\text{max}}} \) | 25.2 | 19.9 | 27.2 |

5. Conclusions
This work proposes an approach for the calculation of the anisotropy of elastic moduli and LTECs with incorporation of substructural inhomogeneity expressed in the presence of tensile/compressive microstrains in grains of different orientations.

Algorithms and programs for calculating the anisotropy of elastic properties and LTECs, taking into account the substructure inhomogeneity, have been developed, the effectiveness of which is demonstrated by the example of a textured zirconium sample. The possibility of calculating the anisotropy of properties is shown in the presence of information about the texture of the material, measured from any of its surfaces, which significantly reduces the laboriousness of preparing samples for various tests. This is especially urgent for thin-walled zirconium alloy cladding tubes.

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