Supplementary information

Models for calculation of X-ray stress factors (XSFs)

X-ray stress factors (XSFs) are linear calibration constants relating the macroscopic stress in a polycrystalline sample (residual or applied) to the lattice strains measured by diffraction in a given direction perpendicular to the diffracting *{hkl}* planes. They describe the average strain response of a set of grains from the gauge volume for which the scattering vector is normal to the *{hkl}* planes. For single-phase polycrystalline material, the relationship between the lattice strains seen by diffraction $< \varepsilon(\phi,\psi)>_{\{hkl\}}$ and macrostress σ_{ij} in the tested volume can be expressed as (after Dölle and Hauk, 1978; Dölle, 1979; Hauk, 1997):

$$< \varepsilon(\phi,\psi)>_{\{hkl\}} = R_{ij}(hkl,\phi,\psi)\,\sigma'_{ij} = F_{ij}(hkl,\phi,\psi)\,\sigma_{ij} \qquad (A1)$$

where σ'_{ij} and σ_{ij} are the macrostress tensors expressed in the **L** and **X** systems[1] (Fig. 1), respectively; $R_{ij}(hkl,\phi,\psi)$ and $F_{ij}(hkl,\phi,\psi)$ are the corresponding XSFs.

The relationship between these two types of stress factors is given by the equation:

$$F_{ij}(hkl,\phi,\psi) = R_{mn}(hkl,\phi,\psi)\gamma_{im}\gamma_{jn} \qquad (A2)$$

where γ_{ij} are elements of the matrix transforming stress from the sample frame (**X**) to the laboratory (**L**) coordinate system (Fig. 1).

It should be mentioned that the $F_{ij}(hkl,\phi,\psi)$ factors are not tensor components, because they relate the stress σ_{ij} expressed in the **X** frame to the elastic strain $< \varepsilon(\phi,\psi)>_{\{hkl\}}$ defined along the L_3 axes of the **L** frame.

Quasi-isotropic polycrystalline material is defined as material characterised by isotropic macroscopic properties despite the anisotropy of particular grains. This condition is fulfilled in the case of random crystallographic texture. For quasi-isotropic materials or materials exhibiting isotropic single-crystal elastic constants, s_1 and s_2 X-ray elastic constants (XECs) are commonly used instead of the more general R_{ij} factors. In this case the following relationships can be expressed as:

$$s_1(hkl) = R_{11}(hkl) = R_{22}(hkl) \quad \text{and} \quad \frac{1}{2}s_2(hkl) = R_{33}(hkl) - R_{11}(hkl) \qquad (A3)$$

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[1] In order to distinguish different reference frames, the prime symbol (') is used to indicate tensors defined with respect to the **L** system, while tensors without the prime symbol are related to the sample system **X**.

Hence, the general Eq. A1 can be written in a more explicit form (cf. for example Noyan and Cohen, 1987 and Hauk, 1997):

$$<\varepsilon(\phi,\psi)>_{\{hkl\}} = \frac{1}{2}s_2\left[(\sigma_{11}-\sigma_{33})cos^2\phi+(\sigma_{22}-\sigma_{33})sin^2\phi+\sigma_{12}\,sin2\phi\right]sin^2\psi$$
$$+s_1\left[\sigma_{11}+\sigma_{22}+\sigma_{33}\right]+\frac{1}{2}s_2\sigma_{33}+\frac{1}{2}s_2\left[(\sigma_{13}\,cos\phi+\sigma_{23}\,sin\phi)\,sin2\psi\right] \tag{A4}$$

where the σ_{ij} values of stress are defined with respect to the X system and $s_1(hkl)$ and $s_2(hkl)$ either depend (in quasi-isotropic materials) or do not depend on the reflection hkl (for isotropic single-crystal elastic constants).

When analysing Eq. A4, it can be concluded that the lattice strains measured for a single hkl reflection linearly depend on $sin^2\psi$ in the case of quasi-isotropic materials when $\sigma_{13}=\sigma_{23}$ = 0. Also, linear $<\varepsilon(\phi,\psi)>_{\{hkl\}}$ vs $sin^2\psi$ plots are obtained, assuming $\sigma_{13}=\sigma_{23}=0$ for multiple hkl reflections, if the single-crystal elastic constants are isotropic.

In order to calculate XECs and XSFs for quasi-isotropic and textured materials, it is necessary to use so-called grain-interaction models. A detailed review of these models is beyond the scope of this work and can be found elsewhere (Baczmański et al. 1993; Matthies and Humbert 1995, Welzel 2005). In this appendix, the principles of the classical models (the Reuss, Voigt and Kröner methods) and the free-surface model applicable for the surface layers are presented.

a) The Reuss model

In the approach proposed by Reuss (1929), stress was assumed to be uniform for all polycrystalline grains across the sample. Consequently, the $R_{ij}(hkl,\phi,\psi)$ factors can be calculated as the average value of single-crystal compliances s'^g_{33ij} (Barral et al., 1987; Brakman, 1987):

$$R_{ij}^{(R)}=<s'^g_{33ij}>_{\{hkl\}}=\frac{\displaystyle\sum_{\{hkl\}}\left\{\int_0^{2\pi}s'^g_{33ij}(\boldsymbol{g})\,f(\boldsymbol{g})d\xi\right\}_{(hkl)}}{\displaystyle\sum_{\{hkl\}}\left\{\int_0^{2\pi}f(\boldsymbol{g})d\xi\right\}_{(hkl)}} \tag{A5}$$

where the average is calculated with the texture $f(\boldsymbol{g})$ weight applied for the single-crystal

compliance tensor $s\,'^{\prime g}_{33ij}$ expressed in the $\mathbf{L}$ system. The summation is performed for all symmetrically equivalent planes *{hkl}* and the integration is carried over all crystal orientations $\boldsymbol{g} = \{\varphi_1, \Phi, \varphi_2\}$ representing diffracting grains (i.e. orientations obtained when the lattice is rotated by an angle of $\xi(\boldsymbol{g})$ around a scattering vector normal to the diffracting plane *(hkl)* (Baczmanski et al., 2008)).

b) The Voigt model

The model proposed by Voigt (1928) assumes that uniform grain elastic strain is equal to elastic macrostrain. In this case, the $R_{ij}{}^V$ factors are equal to (Brakman, 1987):

$$R_{ij}^{(V)} = [c'^{g}]^{-1}_{33ij} \qquad \text{where} \qquad [c\,'^{\prime g}_{ijkl}] = \frac{1}{8\pi^2} \int\limits_E c\,'^{\prime g}_{ijkl}(\boldsymbol{g})\, f(\boldsymbol{g})\, d\boldsymbol{g}, \qquad\qquad (A6)$$

while the $[c'^{g}]^{-1}_{33ij}$ symbol is used for the *33ij* component of the inverse of the $[\mathbf{c'^{g}}]$ mean tensor. The average denoted by *[..]* is calculated for stiffness $c\,'^{\prime g}_{ijkl}(\boldsymbol{g})$ over the whole Euler space *E* (Bunge, 1982), and the texture function *f(*$\boldsymbol{g}$*)* is used as a weight. The $[\mathbf{c'^{g}}]$ tensor defined by Eq. A6 is identical to the macroscopic stiffness tensor of the sample, calculated assuming a uniform elastic strain (as in the Voigt model).

c) The Kröner model (self-consistent)

The Kröner model (Kröner, 1961) was used to calculate the XECs of a quasi-isotropic polycrystalline material approaching the grain by a spherical inclusion (Bollenrath et al., 1967) and was applied to determine the XSFs for textured materials (Sprauel et al, 1989, Baczmański et al., 2003). In this approach, the self-consistent model (Lipinski and Berveiller 1989) can be used to calculate the interaction of a polycrystalline grain with a homogeneous continuous matrix. According to this formalism, the elastic strain $\varepsilon\,'^{\prime g}_{nm}$ (or stress $\sigma\,'^{\prime g}_{nm}$) in the grain *g* is related to the elastic macrostrain $\varepsilon\,'_{kl}$ (or macrostress $\sigma\,'_{kl}$), respectively, i.e.

$$\varepsilon\,'^{\prime g}_{nm} = A\,'^{\prime g(sc)}_{mnkl}\,\varepsilon\,'_{kl} \quad \text{and} \quad \sigma\,'^{\prime g}_{nm} = B\,'^{\prime g(sc)}_{mnkl}\,\sigma\,'_{kl} \quad , \qquad\qquad (A7)$$

where $\mathbf{A}\,'^{g(sc)}$ and $\mathbf{B}\,'^{g(sc)} = \mathbf{c}\,'^{g}\,\mathbf{A}\,'^{g(sc)}\,\mathbf{S}\,'$ are the strain and stress concentration tensors calculated for purely elastic interaction of grain with the matrix using the self-consistent method, $\mathbf{S}\,'$ is the macroscopic compliance tensor, and $\mathbf{c}\,'^{g}$ is the single-crystal stiffness tensor expressed in the $\mathbf{L}$ coordinates. Assuming the validity of Hooke's law on both the

macro and micro scales, ($\varepsilon'_{ij} = S'_{ijkl}\sigma'_{kl}$ and $\varepsilon'^g_{ij} = s'_{ijkl}\sigma'^g_{kl}$) the above equations can be expressed as:

$$\varepsilon'^g_{ij} = \Lambda'^{g(sc)}_{ijkl}\sigma'_{kl} \tag{A8}$$

where: $\Lambda'^{g(sc)}_{ijkl} = A'^{g(sc)}_{ijmn} S'_{mnkl}$ or $\Lambda'^{g(sc)}_{ijkl} = s'^g_{ijmn} B'^{g(sc)}_{mnkl}$.

Finally, the factors $R_{ij}^{(sc)}$ can be calculated from the following equation (Baczmański et al., 2003) :

$$R_{ij}^{(sc)} = <\Lambda'^{g(sc)}_{33ij}>_{\{hkl\}} = \frac{\sum\limits_{\{hkl\}}\left\{\int\limits_0^{2\pi}\Lambda'^{g(sc)}_{33ij}(\boldsymbol{g}) f(\boldsymbol{g})d\xi\right\}_{(hkl)}}{\sum\limits_{\{hkl\}}\left\{\int\limits_0^{2\pi} f(\boldsymbol{g})d\xi\right\}_{(hkl)}}\ , \tag{A9}$$

where the integration is carried over all $\boldsymbol{g}$ orientations representing reflecting grains, similarly as in Eq. A5.

For an elastically deformed polycrystalline aggregate, the macro-compliance tensor $\mathbf{S}'$ required to calculate the $\boldsymbol{\Lambda}'^{g(sc)}$ tensor can be determined using a self-consistent algorithm (Lipiński & Berveiller, 1989).

d) The free-surface model

Baczmański et al. (1993, 2003, 2004, 2008) developed a methodology involving subsurface grains for which intergranular interaction can be directionally dependent. It was proposed that forces and stress normal to the specimen surface propagate similarly as in the Reuss model, i.e. grains can deform freely, while two-dimensional elastic coupling between grains occurs only in the plane parallel to the sample surface and can be calculated by the self-consistent model. In this approach, the grain stress σ^g_{ij} is related to macrostress by the concentration $\boldsymbol{B}^{g(sc-fs)}$ tensor:

$$\sigma^g_{ij} = B^{g(sc-fs)}_{ijkl}\sigma_{kl}\ , \tag{A10}$$

and the $\mathbf{B}^{g(sc)} = \mathbf{c}^g : \mathbf{A}^{g(sc)} : \mathbf{S}$ tensor is calculated for inclusion in the subsurface volume, which is not completely embedded in the sample. In the free-surface model, the concentration tensor is approximated by (Baczmański et al., 2008):

$$B^{g(sc-fs)}_{ijkl} = \begin{cases} I_{ijkl} & \text{for } i = 3 \text{ or } j = 3 \Rightarrow \text{as in Reuss model} \\ B^{g(sc)}_{ijkl} & \text{for } i \neq 3 \text{ and } j \neq 3 \Rightarrow \text{as in self} - \text{consistent bulk model} \end{cases}\ , \tag{A11}$$

where $\boldsymbol{I}$ is the rank-four identity tensor and $\boldsymbol{B}^{g(sc)}$ is the concentration tensor (defined in the $\boldsymbol{X}$ coordinate system) calculated for an inclusion completely embedded in the material. In this approach, the components of the grain stress tensor parallel to the sample surface (σ_{ij}^{g} for $i{\neq}3$ and $j{\neq}3$) are calculated in the same way as for an inclusion completely embedded in the material, i.e. the elastic constraints for the grains are determined by the self-consistent model in a plane parallel to the surface. However, the components normal to the sample surface (σ_{ij}^{g} for $i{=}3$ or $j{=}3$) are assumed to be equal to the corresponding components of macrostress (σ_{ij}), i.e. the elastic interaction between the grains is neglected in the direction normal to the surface because the grains can deform freely. The calculation of the XSFs requires the transformation of the stress concentration tensor to the $\boldsymbol{L}$ system (i.e. $B'^{g(sc-fs)}_{ijkl} = \gamma_{im}\gamma_{jn}\gamma_{ko}\gamma_{lp}B^{g(sc-fs)}_{mnop}$); finally, the $R^{(sc-fs)}_{ij}$ factors can be expressed as follows (Baczmański et al., 2008):

$$R^{(sc-fs)}_{ij} = <\Lambda'^{g(sc-fs)}_{33kl}>_{\{hkl\}} = \frac{\sum\limits_{\{hkl\}}\left\{\int\limits_{0}^{2\pi}\Lambda'^{g(sc-fs)}_{33ij}(\boldsymbol{g})f(\boldsymbol{g})d\xi\right\}_{(hkl)}}{\sum\limits_{\{hkl\}}\left\{\int\limits_{0}^{2\pi}f(\boldsymbol{g})d\xi\right\}_{(hkl)}} \quad , \tag{A12}$$

where $\Lambda'^{g(sc-fs)}_{33kl} = s'^{g}_{33mn}B'^{g(sc-fs)}_{mnkl}$.

Finally, it should be underlined that, similarly to the Vook and Witt method (Vook & Witt, 1968; Leeuwen et al., 1999 and Welzel et al., 2003), the concentration tensor $\mathbf{B}^{g(sc-fs)}$ given by Eq. A11 approaches the real state of the sample in which the grains are freely deformed in a direction perpendicular to the surface. Although a stress concentration tensor thus defined does not represent a strict solution to the physical problem, the components of the local stress tensor (σ_{ij}^{g} in the grain g) calculated using $\mathbf{B}^{g(sc-fs)}$ fulfil the necessary conditions:

- diagonal symmetry required for the stress tensor:

$$\sigma_{ij}^{g} = B^{g(sc-fs)}_{ijkl}\sigma_{kl} = B^{g(sc-fs)}_{jikl}\sigma_{kl} = \sigma_{ji}^{g} \quad , \tag{A13}$$

   because the equation $B^{g(sc-fs)}_{ijkl} = B^{g(sc-fs)}_{jikl}$ is fulfilled in both the self-consistent and Reuss models;

- the mean stress tensor weighted by the volume fraction f^{g} and calculated over the total number N_{g} of grains g is equal to the macroscopic stress:

$$\sum_{g=1}^{N_g} f^g \sigma_{ij}^g = \sum_{g=1}^{N_g} f^g (B_{ijkl}^{g(sc-fs)} \sigma_{kl}) = \left(\sum_{g=1}^{N_g} f^g B_{ijkl}^{g(sc-fs)} \right) \sigma_{kl} = I_{ijkl} \sigma_{kl} = \sigma_{ij}, \qquad \text{(A14)}$$

because $\sum_{g=1}^{N_g} f^g B_{ijkl}^{g(sc-fs)} = I_{ijkl}$ in the self-consistent and Reuss models, while I_{ijkl} denotes the identity tensor.

It is expected as well that the anisotropy of the intergranular interactions also introduces anisotropy of the sample elastic properties. To show this effect, the macroscopic compliance tensor **S** was calculated using $\mathbf{B}^{g(sc-fs)}$ stress concentration for random texture and single-crystal elastic constants of austenite:

$$S_{ijkl} = \sum_{g=1}^{Ng} f^g s_{ijmn}^g B_{mnkl}^{g(sc-fs)} \qquad \text{(A15)}$$

It was found that the in-plane compliances $S_{11} = S_{22}$ (see Table A1, where two-dimensional matrix Voigt notation is used for the compliance tensor) are different from those in the direction normal to the surface S_{33}. Anisotropy is also seen for the shear components, i.e. components $S_{13} = S_{23}$ differ from S_{12}, and $S_{44} = S_{55}$ are not equal to S_{66}. Comparing the macroscopic tensors obtained with different models (Table 1A), it can be stated that the results of the free-surface model fall between those obtained using the Kröner and Reuss models.

Bibliography

Baczmański A., Braham C. & Seiler W. (2003). Phil. Mag. 83, 3225-3246.

Baczmański A., Braham C., Seiler W. & Shiraki N. (2004). Surface and Coating Technology. 182, 43-54.

Baczmański A., Lipinski P., Tidu A., Wierzbanowski K., Pathiraj B. (2008). J.Appl. Cryst. 41, 854-867.

Baczmański A., Wierzbanowski K., Haije W.G., Helmholdt R.B., Ekambaranathan G. & Pathiraj B. (1993). Cryst. Res. Technol. 28, 229-243.

Barral, M., Lebrum, J.L., Sprauel, J.M. & Maeder,G. (1987), Met. Trans. A. 18, 1229-1238.

Bollenrath F., Hauk V. and Muller E.H. (1967). Z. Metallkd. 58, 76-82.

Brakman, C.M. (1987). Phil. Mag. A. 55, 39-58.

Bunge H.J., Texture Analysis in Material Science: Mathematical Methods (1982), edited by: Butterworths & Co., London.

Kröner E. (1961). Acta Metall. 9, 155-161.

Lipinski P. and Berveiller M. (1989). Int. J. of Plasticity. 5, 149-172.

Matthies S. & Humbert M. (1995). J. Appl. Cryst. 28, 254-266.

Reuss A. (1929). Z. Angew. Math. Mech. 9, 49-58.

Sprauel J.M., Francois M. & Barral M. (1989). Proc. of the 2-nd Intern. Conf. on Residual Stresses ICRS2, eds: G.Beck, S.Denis and A.Simon, Elsevier Applied Science.

Van Leeuwen M., Kamminga J.-D. & Mittemeijer E. J. (1999). J. Appl. Phys. 86, 1904-1914.

Voigt W., Lehrbuch der Kristallphysik (1928), edited by: BG Teubner Verlag, Leipzig.

Welzel, U., Leoni, M. and Mittemeijer E.J. (2003). Phil. Mag. 83, 603-630.

Witt F. & Vook, R. W. (1968), J. Appl. Phys. 39, 2773-2776.

Table 1A. Macroscopic compliance tensor (Voigt matrix convention) calculated for an austenitic sample (c_{11}=197 GPa, c_{11}=122 GPa and c_{11}=124 GPa) with random texture orientation using different models

S_{ij} *10^{-3} (GPa^{-1}) Model	$S_{11}=S_{22}$	S_{33}	S_{12}	$S_{13}=S_{23}$	$S_{44}=S_{55}$	S_{66}
Reuss	6.029	6.029	-1.890	-1.890	15.900	15.900
Free surface	5.535	5.717	-1.553	-1.729	14.792	14.200
Kröner	5.101	5.101	-1.426	-1.426	13.104	13.104
Voigt	4.538	4.538	-1.145	-1.145	11.404	11.404