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Internal stresses in polycrystalline zirconia: Microstructure effects

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ABSTRACT

Zirconium oxide layers are submitted to internal stresses that play a role in damage of the layer. If the first order stresses are usually measured or computed, the second order stresses are also important for damage prediction, but they are difficult to measure. Microstructure simulations are performed with the finite element method to obtain internal stresses at grain scale of zirconia. The influences of the crystallographic and morphological textures are studied for monoclinic and tetragonal zirconia. The results show that crystallographic texture has a strong influence on the second order stresses, but not the morphological texture.

1. Introduction

Internal stresses in oxide layers developed on metallic substrates may induce cracking and spalling of the layer [1], leading to a loss of functional properties. This is the case of the zirconia layers obtained by oxidation of zirconium [2] or deposited by physical or chemical techniques [3]. It is therefore important to account for these internal stresses to predict the durability of the layer/substrate system.

These stresses are generally separated in two types [4]: the stresses developed during the layer growth and the thermal stresses induced by the thermal expansion coefficient mismatch between the layer and the substrate. Internal stresses can be measured by different techniques. They are generally based on strain measurements such as X-ray diffraction measurements [5,6] or curvature measurements [3,7]. The internal stresses are then calculated assuming that the mechanical behavior of the material is elastic linear and, usually, isotropic. The macroscopic stress state (1st order) is then obtained, and a mean value of the stresses at the grain scale (2nd order) can be derived from mean field homogenization models [8].

However, brittle fracture of the oxide layer is sensitive to extreme values arising in the microstructure. Only mechanical computations at the microstructural scale that take into account the full description of the microstructure can provide the intra-granular stress distributions needed to analyze the mechanical failure of the oxide layers [9,10]. Furthermore, crystallographic and morphological textures are usually observed in oxide layers: fiber texture and columnar grains perpendicular to the interface between the layer and the substrate are characteristic features of the microstructure of the layers [1,3,7].

This paper aims at studying the influence of crystallographic and morphological textures on the internal stresses at the grain scale. Finite element analyses of polycrystalline aggregates are performed with Cast3M [11], assuming different crystal orientation distributions and different grain shapes. The monoclinic and tetragonal phases of zirconia are considered: both phases may appear in the same layer [5]. Anisotropic thermo-elasticity is considered for the behavior of each phase.

The thermoelastic properties of monoclinic and tetragonal zirconia are first presented. For each phase, two distributions of crystalline orientations are studied: the isotropic distribution and one ideal fiber texture. Two grain shapes are also considered: equiaxed grains and columnar grains. The finite element meshes of Voronoï polyhedra representing polycrystalline aggregates of zirconia are then presented, together with the loadings and the boundary conditions. The simulations according to the different microstructural parameters investigated, are presented and discussed.

2. Simulation data and procedure

2.1. Thermoelastic properties

Monoclinic zirconia, ZrO_2m, is characterized by the following lattice parameters: $a = 0.508$ nm, $b = 0.521$ nm and $c = 0.531$ nm with an angle $\beta = 99.23°$ defined by $\cos\beta = \vec{e}_a.\vec{e}_c$ [12,13] where

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$\vec{e}_a, \vec{e}_c$ are the unit vectors along the a and c axes, respectively. The crystallographic frame is then $(\vec{e}_a, \vec{e}_b, \vec{e}_c)$. The mechanical behavior of the monoclinic zirconia crystal is anisotropic. It is generally expressed in the crystallophysic frame $(\vec{c}_1, \vec{c}_2, \vec{c}_3)$ defined by Eq. (1). Eq. (2) gives the moduli tensor [12,13] expressed in this frame using the Voigt notation ($C_{44} = C_{1212}$, $C_{66} = C_{2323}$) [11]. The thermal expansion coefficients are $\alpha_{aa} = 7.16 \times 10^{-6}\,K^{-1}$, $\alpha_{bb} = 2.16 \times 10^{-6}\,K^{-1}$ and $\alpha_{cc} = 12.6 \times 10^{-6}\,K^{-1}$ [14].

$$\vec{e}_a = \sin\beta\vec{c}_1 + \cos\beta\vec{c}_3,\ \vec{e}_b = \vec{c}_2,\ \vec{e}_c = \vec{c}_3 \tag{1}$$

$$C^c_{IJ}(\mathrm{GPa}) = \begin{pmatrix} 358 & 144 & 67 & 0 & -26 & 0 \\ 144 & 426 & 127 & 0 & 38 & 0 \\ 67 & 127 & 240 & 0 & -23 & 0 \\ 0 & 0 & 0 & 130 & 0 & 39 \\ -26 & 38 & -23 & 0 & 79 & 0 \\ 0 & 0 & 0 & 39 & 0 & 99 \end{pmatrix} \tag{2}$$

For the tetragonal zirconia, ZrO_2t, the moduli tensor is presented in Eq. (3) [12,13].

$$C^c_{IJ}(\mathrm{GPa}) = \begin{pmatrix} 327 & 100 & 62 & 0 & 0 & 0 \\ 100 & 327 & 62 & 0 & 0 & 0 \\ 62 & 62 & 264 & 0 & 0 & 0 \\ 0 & 0 & 0 & 64 & 0 & 0 \\ 0 & 0 & 0 & 0 & 59 & 0 \\ 0 & 0 & 0 & 0 & 0 & 59 \end{pmatrix} \tag{3}$$

Tetragonal zirconia has 2 thermal expansion coefficients: $\alpha_{aa} = 10.8 \times 10^{-6}\,K^{-1}$ and $\alpha_{bb} = 13.7 \times 10^{-6}\,K^{-1}$ [14].

As can be seen, monoclinic zirconia is highly anisotropic with $C_{22}/C_{33} = 1.77$ and $\alpha_{cc}/\alpha_{bb} = 6$, whereas anisotropy of tetragonal zirconia is less important.

In the following, two distributions of crystalline orientations are studied for each phase:

(1) Isotropic distribution: ZrO_2m_iso and ZrO_2t_iso
(2) Ideal fiber texture.

For the monoclinic phase, the considered (001) fiber texture is the one observed for the oxide on nuclear fuel cladding [12]: ZrO_2m_isoT/c3. For tetragonal zirconia, the (100) fiber texture corresponds to zirconia deposited by MOCVD [15]: ZrO_2t_isoT/c1.

Assessments of the moduli can be obtained according to the Voigt and Reuss bounds. In the case of an isotropic distribution of monoclinic zirconia, ZrO_2m_iso, the Voigt bounds of the Young modulus is $E_V = 271$ GPa and the Reuss bounds is $E_R = 226$ GPa. For tetragonal zirconia, ZrO_2t_iso, we get: $E_V = 210$ GPa and $E_R = 192$ GPa. It should be noted that the Voigt and Reuss bounds are similar in the last case.

2.2. Meshes and boundary conditions

The polycrystalline aggregates are modeled by Voronoï polyhedra, even this tessellation of the space probably does not fit exactly the actual topologies of grains [16]. Unit cubes containing two different numbers of polyhedra are designed. They represent polycrystalline aggregates made of 64 or 512 equiaxed grains (Fig. 1). In order to test morphological effects on the internal stress distribution, polycrystalline aggregates with columnar grains are also designed (Fig. 1).

Each polyhedron represents a grain that behaves as a monocrystal characterized by one crystalline orientation. Each crystalline orientation is defined by three Euler angles (φ_1, ϕ, φ_2) [17] that relates the crystallophysic frame $(\vec{c}_1, \vec{c}_2, \vec{c}_3)$ to the global one $(\vec{x}, \vec{y}, \vec{z})$ (Fig. 1). For the isotropic texture, these angles are obtained by a Monte-Carlo process: angles φ_1, φ_2 are uniformly distributed between 0 and 2π, and $\cos\,\phi$ is uniformly distributed between -1 and 1. For the fiber texture, only one angle is needed, the one characterizing the rotation around the fiber axis. It is also obtained by a Monte-Carlo-process.

The aggregates are meshed with about 100 linear tetrahedrons per grain. To study the internal stress distributions for any loading, 6 elementary loadings are defined: 3 extensions in each global direction, x, y and z, and 3 glides along the three planes associated to the global coordinate frame. The loadings are prescribed using kinematic uniform boundary conditions. The displacement vector $\vec{u}$ is imposed at each node of the mesh boundary according to the following relation: $\vec{u} = \varepsilon_0 \tilde{\varepsilon}^* \cdot \vec{x}$, where $\varepsilon_0, \tilde{\varepsilon}^*, \vec{x}$ are respectively the strain magnitude, a unit strain tensor and the node location, $\vec{x} = x\vec{e}_x + y\vec{e}_y + z\vec{e}_z$. For example, the unit strain tensor for an extension in the x-direction is given by: $\tilde{\varepsilon}^* = \vec{e}_x \otimes \vec{e}_x$. It should be noted that computations performed with static uniform boundary conditions demonstrated that the boundary conditions do not modify the conclusions of the study.

The internal stresses stemming from a uniform temperature loading are also computed. In order to avoid any rigid body motion, the following boundary conditions are used at the vertices of the unit cube: $\vec{u}(0,0,0) = \vec{0}$, $u_z(1,0,0) = u_x(1,0,0) = 0$, and $u_z(0,1,0) = 0$. A standard temperature variation equal to $-100\,^\circ$C is prescribed, since the material parameters are supposed to be temperature independent.

3. Results

3.1. Internal stress distributions

The distribution density of each stress component is obtained considering the volume fraction of the mesh submitted to a given stress value. It was verified that the mean value of shear stresses is equal to zero for extension loadings, as for the normal stresses perpendicular to the direction of extension. In the same way, we verified that the mean value of the normal stresses is equal to zero for gliding. Several meshes of polycrystalline aggregates with the same number of grains were generated. And for each mesh, several distributions of crystal orientations were investigated. The results show that the aggregates with 512 grains can be considered as representative volume elements since the stress distributions do not depend on the aggregates. For aggregates of 64 grains, the variation of the mean stress is about 1% between the different aggregates.

As it can be seen in Fig. 2, the stress field is heterogeneous. The mean value is higher for the isotropic monoclinic zirconia (ZrO_2m_iso) than for the isotropic tetragonal zirconia (ZrO_2t_iso). Indeed, the mean value is proportional to the elastic moduli of the equivalent homogeneous material. For the extension in the x-direction, the mean value is equal to $C^x_{11}\,\varepsilon_0$ (where subscript 11 refers to the global coordinate x, and superscript x refers to global frame). With kinematic uniform boundary conditions, the Young modulus is equal to 254 GPa ($C^x_{11} = 320$ GPa) for ZrO_2m_iso, whereas it is equal to 203 GPa ($C^x_{11} = 256$ GPa) for ZrO_2t_iso. These assessments of the Young modulus are lower than the Voigt bounds, as expected. The dispersion of the stresses is high for ZrO_2m_iso, with a stress level that can be up to 30% higher or lower than the mean value. This variation reaches 25% for ZrO2t_iso. This result is induced by the anisotropy of the monoclinic phase that is higher than for the tetragonal phase.

Stress values vary from grain to grain. The component σ_{xx} was averaged over each grain g, to obtain its mean value $\langle\sigma_{xx}\rangle_g$. These values are plotted in Fig. 3 versus C^{xg}_{11}, the component of the moduli tensor expressed in the global frame for the grain g (Fig. 3). The dispersion of the C^{xg}_{11} values gives rise to the higher crystalline