Effect of variant strain accommodation on the three-dimensional microstructure formation during martensitic transformation: Application to zirconia

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Abstract—This paper computationally investigates the effect of martensitic variant strain accommodation on the formation of microstructural and topological patterning in zirconia. We used the phase-field technique to capture the temporal and spatial evolution of embryonic formation of the monoclinic phase in tetragonal single crystals. The three-dimensional simulations were able to capture the formation of all the possible monoclinic variants. We used the multivariant single embryo as an initial condition to mitigate the lack of nucleation criteria at the mesoscale. Without a priori constraint, the model can select the transformation path and final microstructure. The phase-field model was benchmarked against experimental studies on surface uplift formation in zirconia reported by Deville et al. (Acta Mater 2004;52:5697, Acta Mater 2004;52:5709). The simulations showed the excellent capabilities of the model in predicting the formation of a surface relief induced by the tetragonal to monoclinic martensitic transformation.

Keywords: Phase-field modeling; Martensitic transformation; Zirconia; Surface relief

1. Introduction

Zirconia-based ceramics are strong, hard, inert and smooth, with low thermal conductivity and good biocompatibility. Such properties make zirconia ceramics an ideal material for a range of applications from thermal barrier coatings (TBCs) to biomedical applications such as femoral implants and dental bridges [3].

Zirconia has three polymorphs: monoclinic, tetragonal and cubic. In pure zirconia, the cubic phase is stable at temperatures higher than 2640 K to the melting point, while the tetragonal phase is stable between 1430 and 2640 K, and monoclinic phase is stable at room temperature up to 1430 K. However, tetragonal zirconia can be stabilized at lower temperatures by suitable addition of alloying elements such as yttrium and cerium [3]. Nonetheless, tetragonal zirconia can still transform to monoclinic phase (stable) under external loadings or due to crack propagation in surrounding regions. In nuclear fuel rod claddings, this tetragonal to monoclinic (T → M) transformation can lead to crack and porosity formation in the thermally growing oxide layer, with deleterious effects on the structural integrity and durability of the zirconium substrate [4].

Stabilizing the tetragonal phase at the room temperature has revolutionized the application of zirconia in industry [5]. This stabilized zirconia is resistant to crack growth, as the stress field at the crack tip stimulates the T → M transformation. This transformation results in a 5% volume expansion, which helps crack closure and toughening [6,7].

T → M transformation in zirconia is one of the most studied phase transformations in ceramics. In the classical literature, the properties and mechanisms of the T → M transformation have been addressed using two different approaches. The first approach relies on the thermodynamics of transformation [7–11], while the second approach captures the crystallography and topology of the growing variants [12–17]. The thermodynamics-based approach provides some information, such as the start and finish temperatures of the transformation, and the crystallographic approach provides information on the directions of the habit and twin planes. However, failure of zirconia influenced by the T → M transformation in, for example, nuclear power plants [4] and biomedical applications [3],

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has raised several questions that cannot be answered by any of the two above-mentioned approaches:

1. What is the transformation path?
2. What is the stress field of the transformation domains?
3. How would the final microstructure change with loading and boundary conditions?
4. How would the T → M transformation evolve in geometrically complicated specimens?

To redress the current gaps, a more reliable model which captures these mechanisms is needed.

Recently, the phase-field method has been used for capturing solid-state phase transformations, including reconstructive and displacive transformations [18,19]. The phase-field approach combines the thermodynamics, kinetics and crystallographic information of a transformation to capture the microstructural developments during the phase transformation [20]. This method has been frequently used in different moving-boundary applications, such as solidification [21–24], solid-state phase transformation [25–28], grain growth [29] and crack growth [30].

For a martensitic transformation (MT), e.g. T → M transformation, various types of phase-field models exist, which mainly differ in terms of order parameters, thermodynamic potentials, model formulations and numerical methods. Recently, Mamivand et al. [31] reviewed and discussed the phase-field models developed to simulate MT. Three different phase-field approaches were recognized for simulating MT. For instance, within the Ginzburg–Landau theory [32], the primary order parameters may be used to describe either some components of the strain tensor or atomic shuffles. In the first approach, the free energy density is a polynomial in terms of strain components [33–38], while in the second approach, the free energy is a Landau polynomial in terms of atomic shuffles plus a linear or quadratic term which couples order parameters and the strain tensor [19,39–46]. A third approach may be worth mentioning here, which uses the same order parameters as in the aforementioned second approach, but it couples the strain tensor components to the order parameter(s) through a 2-3-4 or higher-order polynomial [47–52].

We recently developed a two-dimensional (2-D) phase-field model for T → M transformation in both single-crystal and polycrystal zirconia [53,54]. The model was envisioned based on the well-known approach of Khachaturyan, Chen and Wang [25,40,45]. The model was able to capture some important features observed or measured in zirconia, such as twin morphology, transformation toughening, shape memory effect and pseudoelasticity.

In this paper, we present a three dimensional (3-D) phase-field model for T → M transformation in zirconia which is anisotropic and elastically inhomogeneous. The 3-D formulation enables us to capture all the possible monoclinic variants. Therefore, we can acquire more realistic microstructural patterns from the simulations. The paper is organized as follows. Section 2 describes the nature of the T → M transformation, including the thermodynamic and crystallography aspects of the transformation; Section 3 presents the process of developing the governing equations of the phase-field model for the T → M transformation; Section 4 includes model parameters; and Section 5 presents and discusses the simulation results for monoclinic embryo evolution and compares these to the experimental results.

2. The nature of the T → M transformation

2.1. Thermodynamics

Solid-state phase transformations can be reconstructive (diffusional) or displacive (diffusionless). In reconstructive transformations, long-range diffusion is required for the growth of the new phases. The main characteristic feature of reconstructive transformations is the necessity of an atomic bond breaking in the parent phase, and new atomic bond reconstruction in the product phase. However in displacive transformations, atoms move only short distances in order to join the new phases. T → M transformation takes place by a displacive mechanism [55]. An important type of displacive phase transformation, which is very common in both metals and ceramics, is martensitic transformation. In martensitic transformation atoms have to move in a coordinated manner, so there is a shape change in the crystal which is associated with transformation strains. The nature of the displacive T → M transformation has led it to be classified as a martensitic transformation, which was first suggested by Wolten [56].

Wang et al. [57] calculated the equilibrium temperature for the T → M phase transformation for pure zirconia and adopted it to assess the Gibbs free energy of zirconia in different phases. According to Ref. [57], the equilibrium temperature is a temperature at which the Gibbs free energy of both tetragonal and monoclinic phases are the same; this temperature for T → M is 1367 ± 5 K, and the Gibbs free energies for monoclinic and tetragonal zirconia are:

$$G^M_{ZrO_2} = -1126163.5 + 424.8908T - 69.38751T \ln T$$
$$- 0.0037588T^2 + 683000T^{-1}, \tag{1}$$

$$G^T_{ZrO_2} = 5468 - 4T + GZrO_2M, \tag{2}$$

where the Gibbs free energies are in J mol^{-1}, and the temperature (T) is in Kelvin.

2.2. Crystallography

The T → M transformation has three correspondences: *A*, *B* and *C* (correspondence determines which atom of the parent phase becomes which atom of the product phase). These are named based on which monoclinic axis is derived from the unique tetragonal *c* axis (c_t). The tetragonal *c* axis can become the *a*, *b* or *c* axis in the monoclinic product phase (a_m, b_m or c_m). When c_t becomes a_m (b_m or c_m) the correspondence is *A* (*B* or *C*). This notation system was introduced by Kriven et al. [58].

Each correspondence has two variants (variants are crystallographically equivalent, but rotated with respect to each other). For example, in correspondence *C* (Fig. 1), the c_t axis becomes the c_m axis, but each of the two other tetragonal axes, which are crystallographically equivalent, has a chance to become a_m or b_m axis. To distinguish between these correspondence variants, Hayakawa et al. [59–61] presented another notation system. They denoted the tetragonal axes by a_t, b_t and c_t (even though the a_t and b_t are crystallographically equivalent), and used a three-letter notation for different monoclinic correspondences and variants. In this notation, the first, second, and third letters indicate which axes would derive from