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Alpha – omega and omega – alpha phase transformations in zirconium under hydrostatic pressure: A 3D mesoscale study

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A B S T R A C T

A three dimensional (3D) elastoplastic phase-field model is developed for modeling the hydrostatic pressure-induced alpha – omega phase transformation and the reverse phase transformation, i.e. omega – alpha, in zirconium (Zr). Plastic deformation and strain hardening of the material are also considered in the model. The microstructure evolution during both phase transformations is studied. The transformation start pressures at different temperatures are predicted and are plotted as a phase diagram. The effect of phase transformations on the mechanical properties of the material is also studied. The input data corresponding to pure Zr are acquired from experimental studies as well as by using the CALPHAD method. Our simulations show that three different omega variants form as laths. On release of pressure, reverse phase transformation initiates at lath boundaries. We observe that both phase transformations are martensitic in nature and also occur at the same pressure, i.e. little hysteresis. The transformation start pressures and the kinetics of the transformation predicted by our model are in good agreement with experimental results.

1. Introduction

Zirconium (Zr) and its alloys are used as structural materials in nuclear reactors and are also used in the chemical industry. These structural components need to withstand a wide range of temperatures and pressures. Thus the components have to be carefully fabricated by a combination of different thermomechanical processes such that a desired microstructure and mechanical properties are obtained. Different phase transformations and consequently different phases can occur in the component during its fabrication and during its service. This can adversely affect the mechanical properties and can lead to failure of the product. Hence, a thorough understanding of microstructural evolution and phase stability of Zr–alloys is essential.

The three phases that mainly form in Zr–alloys are α, β and ω with crystal structures hexagonal close packed (hcp), body centered cubic (bcc) and hexagonal, respectively. In pure Zr, the alpha phase (α) transforms to omega phase (ω) under high static pressure or during shock loading [1,2], whereas at high temperatures the α phase transforms to β phase [3]. The ω phase can also form from beta (β) phase either athermally (by rapid quenching or stress-induced) or isothermally (by holding the material at a constant temperature below 773 K) in Zr–alloys [2,4].

Zirconium, when used as a structural material in a nuclear reactor, can be subjected to high static pressure. This can lead to the formation of brittle ω phase and can affect the mechanical properties of the material. Hence, it is essential to study the co-evolution of microstructure and mechanical properties as well as measure the transformation start pressures during the hydrostatic pressure-induced $\alpha - \omega$ phase transformation in pure Zr using in-situ techniques. However, the difficulties associated with experimental setups in achieving pure hydrostatic conditions can affect the phase transformation kinetics as well as the transformation start pressures, due to the presence of shear stresses in the quasi-hydrostatic environment [5,6].

Although constitutive models have been used to study the relation between phase transformation and plasticity [7–9], the recent advances in modeling the microstructure evolution by using the phase-field method [10,11] are promising and can aid in the study of the $\alpha - \omega$ phase transformation under hydrostatic pressure. Several studies on solid state phase transformations using the phase-field approach have advanced this research area [12–32]. The development of the Ginzburg–Landau model coupled with

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Microelasticity theory, i.e. Phase-field microelasticity theory proposed by Khachaturyan [12], is an important advancement in this field. An alternative approach is developed by using the Ginzburg–Landau formulation in terms of strain order parameters and by enforcing the compatibility condition of strains to ensure that the displacement field is continuous [13,20]. This is in contrast to solving the displacement equations in terms of non-strain order parameters, as proposed by Khachaturyan [12]. Another important advancement is the inclusion of plasticity within the phase-field approach [14,21,24,25,33]. Due to these advances, it is possible to study the interplay between the martensitic microstructure evolution, internal stresses, strains and plastic strains. In our recent work, we have studied the athermal $\beta - \omega$ phase transformation in Zr–Nb alloys [26]. However, the $\alpha - \omega$ phase transformation in pure Zr under hydrostatic pressure has not been studied by using the phase-field approach.

In the present work the hydrostatic pressure-induced alpha (hcp) to omega (hexagonal) diffusionless phase transformation occurring in a single crystal of pure Zr is studied. A three dimensional (3D) phase-field model is developed by including plasticity and strain hardening within the Phase-field microelasticity theory [14,21,25]. The finite element method (FEM) is applied to solve the phase-field as well as the continuum mechanics equations. The effects of application and release of hydrostatic pressure on the microstructure evolution, as well as on the mechanical properties of the material, are studied. The transformation start pressures at different temperatures within the alpha–omega phase field are predicted. The phase diagram predicted by our model is consistent with that obtained from measurements using probes such as third generation light sources [34]. Our results also indicate that at 300 K, both forward and reverse phase transformations occur in a martensitic manner and at the same pressure. On pressure release, reverse phase transformation initiates at lath boundaries. Negligible plastic strain is retained in the material after the reversion of ω to α, which is in agreement with experimental results.

2. Elastoplastic phase-field model

Martensitic transformation can be modeled using the Cahn–Allen equation [35], also known as the Time-Dependent Ginzburg–Landau kinetic equation [12]. The microstructure evolution can be simulated by predicting the time-dependent variation of the phase-field variable η_p, which in turn is related to the minimization of the Gibbs energy G of the system with respect to the phase-field variable as:

$$\frac{\partial \eta_p}{\partial t} = -\sum_{q=1}^{q=v} L_{pq} \frac{\delta G}{\delta \eta_q} \tag{1}$$

where $\delta G/\delta \eta_q$ is a variational derivative that serves as a driving force for the formation of the product phase denoted by the phase-field variable η_q, which is dependent on the position vector $\mathbf{r}$. v is the total number of crystallographic orientations of the product phase and L_{pq} is a kinetic parameter.

The phase transformation of alpha (α) (hexagonal close packed, hcp) to omega (ω) (hexagonal structure) causes asymmetry in the crystal structure and thereby gives rise to three possible crystallographic orientations (variants) of ω. Thus, in order to represent the three possible variants, three phase-field variables, i.e. η_1, η_2, η_3, need to be considered in the model. Hence three phase-field equations, i.e. for $p = 1,2,3$ and $v = 3$ in Eq. (1), need to be solved at each time step. Each of the above three variants is governed by a stress-free transformation strain tensor $\varepsilon_{ij}^{0}(p)$.

From a thermodynamic point of view, the Gibbs energy of a system undergoing a pressure-induced phase transformation consists of the following parts:

$$G = \int_V \left(G_v^{chem} + G_v^{grad} + G_v^{el} + G_v^{appl} \right) \mathrm{d}V \tag{2}$$

G_v^{chem} corresponds to the chemical part of the Gibbs energy density of an unstressed system at the temperature under consideration. G_v^{grad} is the extra Gibbs energy density caused by the interfaces. G_v^{el} is the elastic strain energy density, generated due to the structural phase transformation of $\alpha - \omega$. G_v^{appl} is the extra Gibbs energy density caused by the externally applied load.

2.1. *Chemical energy, G_v^{chem}*

The chemical part of the Gibbs energy density G_v^{chem}, expressed as a Landau-type polynomial [14,17], is given by:

$$G_v^{chem}(\eta_1, \eta_2, \eta_3) = \frac{1}{V_m}\left[\frac{1}{2}A\left(\eta_1^2 + \eta_2^2 + \eta_3^2\right) - \frac{1}{3}B\left(\eta_1^3 + \eta_2^3 + \eta_3^3\right) + \frac{1}{4}C\left(\eta_1^2 + \eta_2^2 + \eta_3^2\right)^2\right] \tag{3}$$

By considering the driving force ΔG_m, i.e. the difference in the Gibbs energies of alpha and omega phases, and the Gibbs energy barrier ΔG^* terms in the above equation, the coefficients are modified [14] as: $A = 32\Delta G^*$, $B = 3A - 12\Delta G_m$ and $C = 2A - 12\Delta G_m$. Here $\Delta G^* = V_m \beta / 2\delta^2$, where V_m is molar volume, δ is thickness of the interface and β relates to interfacial energy and is discussed in the following section.

2.2. *Gradient energy, G_v^{grad}*

The gradient energy density term, G_v^{grad} can be expressed as [14,17]:

$$G_v^{grad} = \frac{1}{2}\sum_{p=1}^{p=v} \beta_{ij}(p) \frac{\partial \eta_p}{\partial r_i}\frac{\partial \eta_p}{\partial r_j} \tag{4}$$

where $\mathbf{r(x,y,z)}$ is the position vector expressed in cartesian coordinates, β_{ij} is the gradient coefficient matrix expressed in terms of the interfacial energy γ, molar volume V_m and Gibbs energy barrier ΔG^*. In the present work isotropic interfacial properties are considered, i.e. β_{ij} is considered as a diagonal tensor with all the elements equal to β [14], given as:

$$\beta = \frac{9\gamma^2 V_m}{16\Delta G^*} \tag{5}$$

2.3. *Elastic energy, G_v^{el}*

In order to calculate the elastic strain energy density G_v^{el}, the elastic stress needs to be calculated by using the Microelasticity theory [12], which is briefly explained below. The martensitic transformation gives rise to deformation of the crystalline lattice and thereby induces stress-free transformation strains $\varepsilon_{ij}^{0}(\mathbf{r})$ into the material. The surrounding parent matrix exerts strain $\varepsilon_{ij}(\mathbf{r})$ on the product phase to resist the stress-free transformation strains and thereby induces elastic strain $\varepsilon_{ij}^{el}(\mathbf{r})$, which in turn gives rise to elastic stress $\sigma_{ij}(\mathbf{r})$, in the material. When the elastic stress exceeds the yield limit of the material, plastic deformation initiates and