



TRANSFORMATION SHEAR OF PRECIPITATED ZrO₂ PARTICLES IN THE PRESENCE OF MULTI- MODE TWINNING

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Abstract—An approach is proposed for the determination of the transformation shear of a single precipitated ZrO₂ ellipsoidal particle embedded in a three-dimensional infinite elastic medium. Multiple modes twinning inside the monoclinic particle transformed from tetragonal symmetry is considered. Kinematically admissible twinning planes and the corresponding twinning elements are determined according to the continuum theory of dispersive phase transformation. The effect of thermal expansion mismatch between particle and matrix is taken into account. The morphology of the transformed particle is determined by the minimization of the potential energy change of the whole particle–matrix system. Numerical examples are presented to show the dependence of the transformation plasticity on the parameters such as temperature, transformation stresses, particle shape and loading directions.

1. INTRODUCTION

A substantial portion of toughness enhancement in ZrO₂-toughened ceramics comes from the transformation strains due to the martensitic transformation of the precipitated ZrO₂ particles from tetragonal to monoclinic symmetry (Budiansky *et al.*, 1983; Evans and Cannon, 1986; Lambropoulos, 1986; McMeeking and Evans, 1982). When a load is applied, the tetragonal particles around the high stress zones of the macro- and microcracks transform themselves into a monoclinic structure. The strains due to such a structural change and the accompanying structure-invariant sliding and twinning produce compression in the crack wakes, which attempts to close the cracks and hence shield the crack tips from the applied load. The toughness enhancement depends on the sizes of the transformation zones around the cracks as well as on the transformation strains inside the zones. The transformation plasticity of the toughened ceramics has been experimentally shown to be dependent on the particle size, shape, orientation and the precipitating environment. A comprehensive understanding of the mechanical aspects, such as yielding, twinning and staining, of the transformation toughening mechanism is of importance in optimizing the microstructure design of structural ceramics by adequately controlling the above-mentioned geometrical and physical parameters.

In the previous calculations of toughness enhancement (Budiansky *et al.*, 1983; Evans and Cannon, 1986; McMeeking and Evans, 1982), the transformation dilatation $\text{tr } \mathbf{E}$ is assumed to be proportional to the volume fraction ρ of the transformed phase as follows:

$$\text{tr } \mathbf{E} = \rho \text{ tr } \mathbf{E}_B, \quad (1)$$

where ρ is usually determined by the applied stress through transformation criterion and \mathbf{E}_B is the stress-free transformation strain of an individual particle and determined by the lattice parameters before and after the transformation. Such a simple relation exists mainly because the dilatant strain of a transformed particle embedded in a matrix is not influenced by the presence of the lattice-invariant twinning and sliding.

The transformation shear components have been ignored in previous theoretical analyses though there is increasing evidence that the macroscopic transformation shears can be comparable with the dilatation (Chen and Reyes-Morel, 1986). Because of the general occurrence of twinning and other shear accommodation processes, a reasonable estimation

of the macroscopic transformation shears should take into account the twinning morphology inside the transformed monoclinic particles. The Greenwood–Johnson model (Greenwood and Johnson, 1965), which has been extensively used to model the transformation plasticity in steels (Leblond *et al.*, 1986, 1989; Oddy *et al.*, 1990), is employed by Sun *et al.* (1991) for zirconia-toughened ceramics. This model assumes that the strain deviator \mathbf{E}' is parallel to the stress deviator \mathbf{T}'

$$\mathbf{E}' = Kf(\rho)\mathbf{T}', \quad (2)$$

where K is a material constant and $f(\rho)$ an increasing function of the volume fraction ρ . This constitutive relation does not hold for general cases and physical justifications should be given when using this model for zirconia toughened ceramics.

Actually, the transformation strains of ceramic composites toughened with randomly oriented or unidirectionally aligned zirconia particles under a certain load are the volume averages of the transformation deformations of the precipitated zirconia particles. To employ a certain average method for composites such as the self-consistent method and the Mori–Tanaka method, we must have an accurate estimation of the transformation deformation of a single zirconia particle of an arbitrary shape and orientation constrained within an infinite homogeneous elastic matrix. In this paper, we consider the transformation yielding and deformation of a single ellipsoidal ZrO_2 particle embedded in a three-dimensional infinite elastic matrix. Based on the continuum theory of phase transformation, in the transformed $m\text{-ZrO}_2$ particle, there may be four martensitic variants due to twinings on six kinematically admissible twin planes. The twin structure of the transformed particle is such that the particle deformation can be approximately regarded as a homogeneous one over the region occupied by the particle. The total potential energy change ΔG_m due to the transformation in the presence of thermal expansion mismatch between the matrix and the particle is then investigated. The volume fraction of each of the four martensitic variants is such that the whole particle–matrix system assumes a minimum free energy state. This principle incorporated into the end-point thermodynamics of phase transformation is used to determine the yield behavior of zirconia particle with a certain energy barrier. Numerical examples are specially chosen to show the dependence of the transformation plasticity on parameters such as transformation stress, temperature, particle shape and lattice orientation.

2. KINEMATICALLY ADMISSIBLE TWINNING SYSTEMS

One approach to consider the transformation kinematics is to assume that the transformation deformation is piece-wisely linear. The transformed crystal is considered as a polycrystalline edifice, built up of two or more homogeneous portions of the same crystal species in juxtaposition (James, 1981, 1986). The interface between any two homogeneous deformation regions is referred to as a twinned plane and deformation discontinuity occurs across the plane. The possible twin plane and the change in deformation gradients across the twinned plane can be determined from the continuity requirement of the displacement vector and the well-defined twin law. This purely continuum view of elastic and thermoelastic crystals in the prediction of phase changes has been explored by Ericksen (1985), James (1981, 1986) and many others, with emphasis on the relation of convexity conditions on the potential energy density function to the stability of deformations.

In a previous study (Lam and Zhang, 1992), the authors have employed Ericksen's method to investigate the kinematical aspects of transformation twinning in the monoclinic ZrO_2 particles transformed from tetragonal phase. The numerical calculations show that six twin systems may operate in the monoclinic zirconia particles, some of which have been well identified in experimental observations. We have also demonstrated that the fine coherence of the twin interface expects an amplitude vector on the interface which happens to approximately satisfy the invariant plane strain assumption in the phenomenological theory of martensitic transformation. With these surprising results, we have a strong reason to take the continuum theory of dispassive phase transformation as the basis of our

kinematical analysis. In the rest of the section, we will briefly review some concepts in the theory which will be used in the subsequent discussions.

We here assume that the particle of tetragonal crystal is large enough to be taken as a continuum. Let \mathbf{X} be the position vector of a typical material point \mathbb{X} in a single t -ZrO₂ crystal in the reference configuration. A deformation of the crystal is an invertible and continuous function $\mathbf{x} = \mathbf{f}(\mathbf{X}, t)$. If function \mathbf{f} is differentiable at \mathbf{X} , we call

$$\mathbf{F} = \frac{\partial \mathbf{x}}{\partial \mathbf{X}}, \quad (3)$$

the deformation gradient at \mathbf{X} .

Let $\phi(\mathbf{F}, \vartheta)$ be the Gibbs' free energy per unit mass, ϑ the absolute temperature. Let \mathcal{G} be the symmetry group of the crystal at the parent phase. According to the frame-indifference principle and the definition of material symmetry group, it must hold that:

$$\phi(\mathbf{F}, \vartheta) = \phi(\mathbf{QFH}, \vartheta), \quad \forall \mathbf{H} \in \mathcal{G}, \quad \forall \mathbf{Q} \in \mathcal{O}, \quad (4)$$

where \mathcal{O} is the orthogonal transformation group of the three dimensional Euclidean space, and the juxtaposition of two second order tensors denotes tensor multiplication, for instance, $(\mathbf{QF})_{ij} = Q_{ik}F_{kj}$. The restriction (4) implies that the Gibbs' free energy density function $\phi(\mathbf{F}, \vartheta)$ depends on the deformation gradient \mathbf{F} only through the right stretch tensor $\mathbf{U} = (\mathbf{F}^T\mathbf{F})^{1/2}$ and that for any symmetry operation $\mathbf{H} \in \mathcal{G}$ the following holds:

$$\phi(\mathbf{HUH}^T, \vartheta) = \phi(\mathbf{U}, \vartheta). \quad (5)$$

At certain temperatures and stress states, the tetragonal zirconia particle changes its crystal structure into a monoclinic one. We shall therefore presume that the crystal favors a new stable configuration. From the viewpoint of energy, an exchange of stability can occur because the new configuration assumes less free energy in the sense that for all deformations \mathbf{U} in the neighborhood of a stable point \mathbf{U}_0 , we have:

$$\phi(\mathbf{U}, \vartheta_0) > \phi(\mathbf{U}_0, \vartheta_0). \quad (6)$$

Please see James (1986) for more details.

It is obvious that if \mathbf{U}_0 is a minimizer for the free energy density function $\phi(\cdot, \vartheta_0)$, so is $\mathbf{HU}_0\mathbf{H}^T$ for every symmetry operation $\mathbf{H} \in \mathcal{G}$. In general, $\phi(\cdot, \vartheta_0)$ may have up to $v+1$ potential wells with minima at deformations $\mathbf{U}_0, \mathbf{H}_1\mathbf{U}_0\mathbf{H}_1^T, \dots, \mathbf{H}_v\mathbf{U}_0\mathbf{H}_v^T$, in which $\mathbf{H}_1, \dots, \mathbf{H}_v$ is an enumeration of the symmetry point group \mathcal{G} . We call each distinct potential well with minimum of the form $\mathbf{H}_i\mathbf{U}_0\mathbf{H}_i^T$ a variant of the martensite.

A deformation configuration of a crystal is said to be stable if it minimizes the total free energy of the system. A necessary and sufficient condition for stability is that the deformation gradient corresponding to the configuration minimizes the free energy density function $\phi(\mathbf{U}, \vartheta)$ for almost every \mathbf{X} over the crystal under consideration. In other words, for most every point \mathbf{X} , the right stretch tensor \mathbf{U} must take on one of the values

$$\mathbf{U}_0, \mathbf{H}_1\mathbf{U}_0\mathbf{H}_1^T, \dots, \mathbf{H}_v\mathbf{U}_0\mathbf{H}_v^T. \quad (7)$$

A twinned product phase is a polycrystal built up of the same single crystals with different orientations. Each of the single crystal regions undergoes a homogeneous deformation with the right stretch tensor \mathbf{U} being one of the values in eqn (7). The interfaces

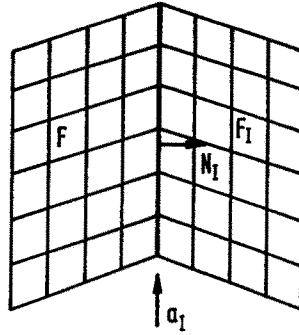


Fig. 1. Twin plane and twinning elements.

between these homogeneous deformation regions are called twin planes (see Fig. 1). Let the deformation gradients be \mathbf{F} and \mathbf{F}_I at two sides of the twin plane \mathcal{S} , respectively, they have polar decompositions:

$$\mathbf{F} = \mathbf{R}\mathbf{U}, \quad \mathbf{F}_I = \tilde{\mathbf{R}}_I = \tilde{\mathbf{R}}_I\mathbf{U}_I. \quad (8)$$

\mathbf{U}_I differs from \mathbf{U} only by a symmetry rotation, as we have given above. That is, there is such a symmetry rotation $\mathbf{H}_I \in \mathcal{G}$ that:

$$\mathbf{U}_I = \mathbf{H}_I^T \mathbf{U} \mathbf{H}_I, \quad (9)$$

or, equivalently:

$$\mathbf{F}_I = \mathbf{R}_I \mathbf{F} \mathbf{H}_I, \quad \mathbf{R}_I = \tilde{\mathbf{R}}_I \mathbf{H}_I^T \mathbf{R}^T \in \mathcal{O}, \quad \mathbf{H}_I \in \mathcal{G}. \quad (10)$$

On the other hand, according to the well-known Hadamard jump condition, continuity requirement of displacement across the plane \mathcal{S} guarantees the existence of an amplitude vector \mathbf{a}_I such that at each point of \mathcal{S} :

$$\mathbf{F}_I = \mathbf{F} + \mathbf{a}_I \otimes \mathbf{N}_I, \quad (11)$$

with \mathbf{N}_I being the unit normal to \mathcal{S} defined with respect to the reference configuration.

Methods are available to determine the possible planes of discontinuity satisfying eqns (10–11) for a transformation whereby the lattices of the parent and product phases are known. Lam and Zhang (1992) have employed Ericksen's method to investigate the kinematical aspects of transformation twinings in the monoclinic ZrO_2 particles transformed from tetragonal structure. For later use, we present here the right-stretch tensors of all the four martensitic variants for the transformation of ZrO_2 particles in Mg-PSZ at room temperature.

$$\mathbf{U}_0 = \begin{vmatrix} 1.0102 & 0 & 0.0824 \\ 0 & 1.0256 & 0 \\ 0.0826 & 0 & 1.0260 \end{vmatrix}, \quad \mathbf{U}_1 = \begin{vmatrix} 1.0102 & 0 & -0.0824 \\ 0 & 1.0256 & 0 \\ -0.0826 & 0 & 1.0260 \end{vmatrix},$$

$$\mathbf{U}_2 = \begin{vmatrix} 1.0256 & 0 & 0 \\ 0 & 1.0102 & 0.0824 \\ 0 & 0.0824 & 1.0260 \end{vmatrix}, \quad \mathbf{U}_3 = \begin{vmatrix} 1.0256 & 0 & 0 \\ 0 & 1.0102 & -0.0824 \\ 0 & -0.0824 & 1.0260 \end{vmatrix}. \quad (12)$$

The lattice parameters for both the parent and product phases are as follows (Kelly and Ball, 1986):

$$\begin{aligned}
 a_t &= 0.50803, & b_t &= 0.50803, & c_t &= 0.51903 \\
 a_m &= 0.51170, & b_m &= 0.51770, & c_m &= 0.53030\beta = 80^\circ 45'.
 \end{aligned}
 \tag{13}$$

3. AVERAGED TRANSFORMATION STRAINS OF THE TWINNED PARTICLE

Let \mathbf{F} be the transformation gradient defined with respect to an orthonormal basis whose directions are consistent with three lattice vectors. Assume that $(\mathbf{F}_1, \mathbf{F}_2, \dots, \mathbf{F}_n)$ are the deformation gradients of all the variants which may be twin-related to \mathbf{F} . That is to say, for each deformation gradient $\mathbf{F}_I \in \{\mathbf{F}_1, \mathbf{F}_2, \dots, \mathbf{F}_n\}$, there exist two orthogonal tensors $\mathbf{R}_I \in \mathcal{O}$, $\mathbf{H}_I \in \mathcal{S}$ and two vectors $(\mathbf{a}_I, \mathbf{N}_I)$ such that eqns (10–11) are satisfied. Now, we will investigate what are the deformation gradients of the variants which are twin-related to \mathbf{F}_I .

In fact, if we replace $(\mathbf{R}_I, \mathbf{H}_I, \mathbf{a}_I, \mathbf{N}_I)$ by $(\mathbf{R}_I^*, \mathbf{H}_I^*, \mathbf{a}_I^*, \mathbf{N}_I^*)$ and \mathbf{F} by \mathbf{F}_I in eqns (10–12), respectively, we will easily find that the new equations have exactly n solutions $(\mathbf{R}_I^* \mathbf{F}_I, \mathbf{H}_I^*, \dots, \mathbf{R}_I^* \mathbf{F}_I \mathbf{H}_I^*)$. This implies that there may be a variant whose deformation gradient is $\mathbf{R}_I^* \mathbf{F}_I \mathbf{H}_I^*$ with respect to the given coordinate system. Similarly, we can obtain a variant with deformation gradient:

$$\mathbf{F}_{IJ\dots K} = \mathbf{R}_I \mathbf{R}_J \dots \mathbf{R}_K \mathbf{F} \mathbf{H}_K \dots \mathbf{H}_J \mathbf{H}_I,
 \tag{14}$$

where I, J, K can be arbitrary integers among $[1, 2, \dots, n]$.

Now, we may have a clear and complete picture of the twinned crystal. It is built up of the homogeneous deformation regions $\mathbf{F}_{IJ\dots K}$ which interface with each other only on a finite number of well-defined twin planes (see Fig. 2). As we mentioned before, we are mainly interested in the macroscopic transformation strains of the twinned particle since these strains are essential for toughness calculations. Theoretically, there are two approaches to averaging the deformations of the variants involved. One is based on the assumption that the averaged deformation gradient \mathbf{F}_p of the particle is the volume average of the deformation gradient variants, i.e.

$$\mathbf{F}_p = \frac{1}{V} \sum_{I,J,\dots,K} V_{IJ\dots K} \mathbf{F}_{IJ\dots K},
 \tag{15}$$

where $V_{IJ\dots K}$ is the volume of the region with transformation deformation gradient $\mathbf{F}_{IJ\dots K}$ as defined by eqn (14), V the total volume of the particle considered. The strain tensor \mathbf{E}_p of the particle is then determined by the formula:

$$\begin{aligned}
 \mathbf{E}_p &= \frac{1}{2}(\mathbf{F}_p^T \mathbf{F}_p - \mathbf{I}) \\
 &= \frac{1}{2} \left(\frac{1}{V^2} \sum_{I,J,\dots,K} \sum_{\alpha\beta,\dots,\gamma} V_{IJ\dots K} V_{\alpha\beta,\dots,\gamma} \mathbf{F}_{IJ\dots K}^T \mathbf{F}_{\alpha\beta,\dots,\gamma} - \mathbf{I} \right).
 \end{aligned}
 \tag{16}$$

This averaging approach cannot be used directly for the calculations of transformation

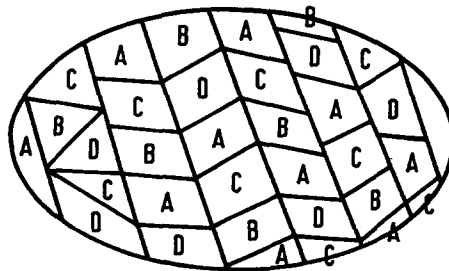


Fig. 2. A schematic description of the twin structure of the transformed zirconia particle. A, B, C, D denote four possible martensitic variants.

shear in the case of $t-m$ Zirconia transformation since the deformation gradient variants given by eqn (14) are too many to be manageable. In this paper, we shall employ another approach which has been widely used in composite mechanics where the macroscopic strains of multi-phase material are considered as the volume average of the strains of the phases involved. For a given deformation gradient variant $\mathbf{F}_{IJ\dots K}$, the Green strain tensor $\mathbf{E}_{IJ\dots K}$ is defined as:

$$\mathbf{E}_{IJ\dots K} = \frac{1}{2}(\mathbf{F}_{IJ\dots K}^T \mathbf{F}_{IJ\dots K} - \mathbf{I}) = \frac{1}{2}(\mathbf{U}_{IJ\dots K}^2 - \mathbf{I}). \quad (17)$$

As we have already pointed out in the last section, the right stretch tensor $\mathbf{U}_{IJ\dots K}$ must take on one of the values \mathbf{U}_i 's in (7). The average transformation strains are therefore expressed as follows:

$$\mathbf{E} = \frac{1}{V} \sum_{IJ\dots K} V_{IJ\dots K} \mathbf{E}_{IJ\dots K} = \sum_{i=0}^v \lambda_i \mathbf{E}_i, \quad (18)$$

where λ_i is the total volume fraction of the variants $\mathbf{F}_{IJ\dots K}$ whose right stretch tensors are \mathbf{U}_i and

$$\mathbf{E}_i = \frac{1}{2}(\mathbf{U}_i^2 - \mathbf{I}). \quad (19)$$

Since the dilatations of all the martensitic variants \mathbf{U}_i are the same, obviously, we have:

$$\text{tr } \mathbf{E}_i = \text{tr } \mathbf{E}_\sigma, \quad i \neq \sigma \quad \text{and} \quad \text{tr } \mathbf{E} = \sum_{i=0}^v \lambda_i \text{tr } \mathbf{E}_i = \text{tr } \mathbf{E}_0. \quad (20)$$

This leads to our previous conclusion that the presence of twinning does not alter the transformation dilatation of the particle.

For deviatoric transformation strain, it has been shown that some combinations of martensitic variants can essentially reduce the shear deformation from about 16% to 1% (Lam and Zhang, 1992). The amount of reduction is dependent on the volume fraction λ_i of each of the deformation variants operating in the particle. This should be determined from the volume fractions $\lambda_0, \dots, \lambda_3$ in eqn (18) based on some physical principle which will be elaborated in the next section.

4. MINIMUM POTENTIAL ENERGY PRINCIPLE

Another issue to be clarified in order to use the eqn (18) is to determine what force controls the selection of twinning mode and the amount of each of the martensitic variants. Jaswon and Dove (1956) assume that the operative twinning mode always involves the smallest possible homogeneous shear which can twin the lattice. In this paper, it is postulated that the minimization of the potential energy is the principal factor in dictating the morphology and crystallography of a martensitic transformation. This postulate can be deduced from the principle of minimum Gibbs' free energy for stress-induced transformation by ignoring the dependence of the interfacial energy change on the combination of the martensitic variants. It was also employed by Shibata and Ono (1975, 1977) to calculate the habit planes and the martensite shape for b.c.c. to h.c.p. martensitic transformation. The results correlate well with experimental observations.

We assume that a tetragonal zirconia particle of single crystal is embedded in a three-dimensional infinite medium with remotely applied load which produces a homogeneous stress \mathbf{T} field in the matrix in the absence of the particle. The particle is assumed to be bonded to the matrix perfectly in the sense that there are no microcracks developed along the matrix-particle interface. Although the high internal stresses due to the thermal expansion mismatch will induce microcracks around the particle for such ceramics as $\text{Al}_2\text{O}_3\text{-ZrO}_2$ systems, the effects of the microcracks can be approximately modelled by

replacing the matrix with a damaged one with reduced stiffness (Garvie and Swain, 1985). For ZrO₂-toughened ceramics, the ordinary plastic strains due to crystallographic sliding are negligible both inside the matrix and inside the particles because both the matrix and the particles are brittle. We further assume that both the matrix and the particle are elastically isotropic and have the same elasticity tensor C .

As a result of thermal expansion mismatch with the matrix, residual stresses will arise inside the particle during cooling from the fabrication temperature. To take into account the effect of residual stresses, we assume that there exists an eigenstrain :

$$\mathbf{E}^r = \mathbf{E}^{ri} - \mathbf{E}^m, \quad (21)$$

inside the particle before any external force is loaded, where \mathbf{E}^{ri} and \mathbf{E}^m are the thermal expansions of the particle and the matrix, respectively. According to Eshelby's results (Eshelby, 1957), the residual stress tensor \mathbf{T}^r inside the particle due to \mathbf{E}^r is :

$$\mathbf{T}^r = \mathbf{S} \cdot (\mathbf{C} - \mathbf{I}_4) \cdot \mathbf{E}^r, \quad (22)$$

where \mathbf{S} is the so-called Eshelby tensor which depends on the Poisson's ratio and the geometric parameters of the particle, and \mathbf{I}_4 is the fourth-order identity tensor. The contracted products $\mathbf{C} \cdot \mathbf{S}$ and $\mathbf{S} \cdot \mathbf{E}$ denote $C_{ijkl}S_{klmn}$ and $S_{ijkl}E_{kl}$, respectively.

When the applied load \mathbf{T} increases to a critical level, the stress-induced transformation occurs, accompanied with a transformation eigenstrain \mathbf{E} . According to the classical Eshelby solution, the stress \mathbf{T}^p inside the particle after the transformation is as follows :

$$\mathbf{T}^p = \mathbf{C} \cdot (\mathbf{S} - \mathbf{I}_4) \cdot \mathbf{E} + \mathbf{T} + \mathbf{T}^r. \quad (23)$$

The change in Gibbs' free energy due to the transformation is expressed as :

$$\Delta G = \Delta G_m + \Delta G_{ch} + \Delta G_s, \quad (24)$$

where ΔG_m is the net change in the potential energy of the whole matrix-inclusion system which is given as follows (Mura, 1987)

$$\Delta G_m/V = -\frac{1}{2}(\mathbf{T}^p - \mathbf{T}) \cdot (\mathbf{E} + \mathbf{E}^r) + \frac{1}{2}\mathbf{T}^r \cdot \mathbf{E}^r - \mathbf{T} \cdot \mathbf{E}, \quad (25)$$

with $\mathbf{T} \cdot \mathbf{E}$ defined by $T_{ij}E_{ij}$, ΔG_s is the surface energy change both in the interface between the particle and the matrix and in the interfaces between the twinned variants and ΔG_{ch} is the change in chemical energy which, when martensite is the low temperature phase, is given approximately by :

$$\Delta G_{ch}/V = f_s(\vartheta_s - \vartheta), \quad (26)$$

with f_s being a negative coefficient depending on the alloying content, ϑ_s the stress-free equilibrium transformation temperature and ϑ the temperature at which the stress-induced transformation deformation is considered (Garvie and Swain, 1985). The expression for the interfacial energy change is quite complicated. Although a complete consideration for it requires a full knowledge of the twin structure of the transformed particle, it can be reasonably approximated as a function of the number n of the twins inside the particle as follows (Garvie and Swain, 1985) :

$$\Delta G_s/V = \ell_1 \chi^{-1} (1 + \ell_2 \eta(n)), \quad \eta(n) = \frac{(n-1)(n+1)}{n}, \quad \ell_1, \ell_2 > 0, \quad (27)$$

where ℓ_1 and ℓ_2 are two material constants and χ is the characteristic radius of the particle. Taking into account the fact that the chemical energy change ΔG_{ch} is independent of the

twinning morphology inside the transformed particle, the requirement that the twinning inside the particle minimizes the free energy change ΔG is equivalent to the requirement that the twinning minimizes the potential change ΔG_m .

Substituting eqn (23) into eqn (25), we obtain the following

$$\Delta G_m/V = -\frac{1}{2}(\mathbf{C} \cdot (\mathbf{S} - \mathbf{I}) \cdot \mathbf{E} + \mathbf{C} \cdot (\mathbf{S} - \mathbf{I}) \cdot \mathbf{E}' + ((\mathbf{S} - \mathbf{I}) \cdot \mathbf{C}) \cdot \mathbf{E}') \cdot \mathbf{E} - \mathbf{T} \cdot \mathbf{E}. \quad (28)$$

The dependence of the potential change ΔG_m on the volume fraction λ_i of the deformation variant \mathbf{U}_i can be clearly seen by taking into account eqn (18). Although the volume fractions $(\lambda_0, \lambda_1, \lambda_2, \lambda_3)$ minimize the energy change ΔG_m , the stability point obtained from the requirement:

$$\frac{1}{V} \frac{\partial \Delta G_m}{\partial \lambda_i} = 0, \quad i = 0, 1, 2, 3, \quad (29)$$

may be beyond the bound $0 \leq \lambda_i \leq 1$. Therefore, the volume fractions $(\lambda_0, \lambda_1, \lambda_2, \lambda_3)$ should be determined by minimizing the goal function (28) under the following restrictions

$$0 \leq \lambda_i \leq 1, \quad \lambda_0 + \lambda_1 \lambda_2 + \lambda_3 = 1.0. \quad (30)$$

The quadratic term $-1/2 (\mathbf{C} \cdot (\mathbf{S} - \mathbf{I}) \cdot \mathbf{E}) \cdot \mathbf{E}$ must be positive definite with respect to \mathbf{E} since it denotes the potential change of the whole particle and matrix system due to eigenstrain \mathbf{E} . This leads to the conclusion that the goal function (28) is a positive definite quadratic function in the volume fractions $(\lambda_0, \lambda_1, \lambda_2, \lambda_3)$. There exists a *unique* solution for the quadratic programming problem since the constrain (30) determines a nonempty feasible domain.

5. TRANSFORMATION YIELD CRITERIA

In order to evaluate the potential energy change, one has to know the yield stress at which the transformation takes place. According to Lambropoulos (1986), a necessary condition for the transformation to take place is that the change in Gibbs' free energy of the whole matrix-particle system is nonpositive, i.e.

$$\Delta G = \Delta G_{cr} \leq 0. \quad (31)$$

where ΔG_{cr} is the critical value that ΔG has to attain for the transformation to be thermodynamically favorable.

Define the applied stress as $\mathbf{T} = \sigma \mathbf{T}_0$, where σ is a scalar loading parameter characterizing the loading level and is always taken as positive, \mathbf{T}_0 is a second order symmetric tensor characterizing the loading direction and complexity. Taking into account eqns (23) and (24) and the fact that the transformation with a combination $(\lambda_0, \lambda_1, \lambda_2, \lambda_3)$ of the martensitic variants will first occur if the combination requires a lower positive parameter σ , the critical loading parameter σ_{cr} can therefore be determined by:

$$\sigma_{cr} = \min \left\{ \frac{\Delta G_c/V - 0.5(\mathbf{C} \cdot (\mathbf{S} - \mathbf{I}) \cdot (\mathbf{E} + \mathbf{E}') + (\mathbf{S} - \mathbf{I}) \cdot \mathbf{C}) \cdot \mathbf{E}'}{\mathbf{T}_0 \cdot \mathbf{E}} \right\}, \quad (32)$$

where $\Delta G_c = \Delta G_{ch} + \Delta G_s - \Delta G_{cr}$ and the minimizer is sought among those volume fractions which satisfy restriction (30).

Similar formula for \mathbf{E}' being zero was also given by Zhang *et al.* (1993). Transformation yielding has been investigated in detail for the cases of uniaxial loadings. It has been numerically shown that the theory expects a strongly anisotropic yield surface under uniaxial loading and that uniaxial compressive loading cannot induce the $t \rightarrow m$ phase transformation at all at some loading directions.

It can be shown that under the restrictions (30) and $\mathbf{T}_0 \cdot \mathbf{E} > 0$, the minimizer of goal function (32) is also a minimizer of the potential energy change (28). In fact, let the minimizer of the goal function (32) be $(\lambda_0, \lambda_1, \lambda_2, \lambda_3)$. For any other combination $(\hat{\lambda}_0, \hat{\lambda}_1, \hat{\lambda}_2, \hat{\lambda}_3)$ of the martensitic variants, satisfying $\mathbf{T}_0 \cdot \hat{\mathbf{E}} > 0$, we always have:

$$\sigma_{cr} \leq \frac{\Delta G_c/V - 0.5(\mathbf{C} \cdot (\mathbf{S} - \mathbf{I}) \cdot (\hat{\mathbf{E}} + \mathbf{E}') + (\mathbf{S} - \mathbf{I}) \cdot \mathbf{C}) \cdot \mathbf{E}' \cdot \hat{\mathbf{E}}}{\mathbf{T}_0 \cdot \hat{\mathbf{E}}}, \quad (33)$$

where $\hat{\mathbf{E}}$ is the average Green strain corresponding to the martensite combination $(\hat{\lambda}_0, \hat{\lambda}_1, \hat{\lambda}_2, \hat{\lambda}_3)$. Some simple manipulations of the above inequality lead to the conclusion that at the stress level σ_{cr} , the martensite combination $(\lambda_0, \lambda_1, \lambda_2, \lambda_3)$ does minimize the free energy change,

$$\Delta G_{cr}/V \leq -\frac{1}{2}(\mathbf{C} \cdot (\mathbf{S} - \mathbf{I}_4) \cdot (\hat{\mathbf{E}} + \mathbf{E}') + (\mathbf{S} - \mathbf{I}_4) \cdot \mathbf{C}) \cdot \mathbf{E}' \cdot \hat{\mathbf{E}} - \sigma_{cr} \mathbf{T}_0 \cdot \hat{\mathbf{E}} + \Delta G_{ch}/V + \Delta G_s/V, \quad (34)$$

with $\Delta G_{ch}/V$ independent of the martensite combination $(\hat{\lambda}_0, \hat{\lambda}_1, \hat{\lambda}_2, \hat{\lambda}_3)$. The equality holds when $(\hat{\lambda}_0, \hat{\lambda}_1, \hat{\lambda}_2, \hat{\lambda}_3)$ is equal to $(\lambda_0, \lambda_1, \lambda_2, \lambda_3)$.

Transformation criterion (32) seems to be theoretically most reasonable. Before it can be employed for the determination of the yield behavior of ceramic composites, a bridge should be built between the single particle model discussed here and the ceramic composites toughened with randomly oriented or unidirectionally aligned tetragonal zirconia particles. Appropriate approach should be searched to average the transformation behavior of a single particle with respect to particle location, particle orientation and particle size and shape. Once this has been done, the dependence of the material parameter ΔG_c on particle size, alloying content, temperature and other geometric and physical parameters can be quantitatively identified by experiments on ceramics with specially designed microstructures. We will discuss these issues in detail in a separate paper. Here we will numerically show that criterion (32) could be justified qualitatively as well as quantitatively by our knowledge about the transformation behavior or composite ceramics already recorded in the literature.

6. NUMERICAL ANALYSIS

Here, we will investigate the $t \rightarrow m$ transformation of spheroidal, oblate spheroidal and prolate spheroidal inclusions under hydrostatic and uniaxial loadings. The aspect ratio τ is defined as $\tau = c/b = c/a$ for prolate spheroid and as $\tau = c/a = b/a$ for oblate spheroid, where $a \geq b \geq c$ are the three principal radii of the ellipsoidal particle. The complexity tensor \mathbf{T}_0 is \mathbf{I} for hydrostatic compression and

$$\mathbf{T}_0 = \begin{bmatrix} n_1 n_1 & n_1 n_2 & n_1 n_3 \\ n_2 n_1 & n_2 n_2 & n_2 n_3 \\ n_3 n_1 & n_3 n_2 & n_3 n_3 \end{bmatrix}, \quad (35)$$

for uniaxial tensile loading, where:

$$\mathbf{n} = \{n_1, n_2, n_3\} = \{\cos \beta \cos \alpha, \cos \beta \sin \alpha, \sin \beta\}, \quad (36)$$

is the loading direction relative to the principal axes of the inclusion.

The general solution for the components of the Eshelby tensor \mathbf{S} has been described in terms of elliptic integral (Eshelby, 1957). Closed solutions can be found for the three special inclusion shapes to be considered: spherical, prolate spheroidal and oblate spheroidal (Mura, 1987). The elasticity tensors \mathbf{C} for both the particle and the matrix are assumed to be isotropic, which can be expressed as:

$$C_{ijkl} = G \left(\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} + \frac{2\mu}{1-2\mu} \delta_{ij} \delta_{kl} \right), \quad (37)$$

where G is the shear modulus, μ the Poisson's ratio and δ_{ij} the Kronecker delta. $G = 90$ GPa and $\mu = 0.3$ (Chen and Reyes-Morel, 1986) will be used for all the following calculations.

6.1. Residual stresses and strains

Let the rectangular cartesian coordinate system $\mathfrak{R}_{\text{incl}}$ be taken whose axes coincide with the principal axes of the ellipsoidal inclusion. Assume that the thermal expansion of both the tetragonal zirconia particle and the matrix is isotropic with thermal expansion coefficients $\alpha_t = 15.2 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$ and $\alpha_m = 8.6 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$ (Garvie and Swain, 1985), respectively. The eigenstrain \mathbf{E}^r due to the thermal expansion mismatch is $\mathbf{E}^r = 6.6 \times 10^{-6} \Delta\vartheta \mathbf{I}$, where $\Delta\vartheta = \vartheta_f - \vartheta$ and ϑ_f is the fabrication temperature. The following are the residual stresses according to eqn (22) for spherical, prolate spheroidal ($\tau = 0.1$) and oblate spheroidal particles ($\tau = 0.1$), respectively:

$$\mathbf{T}_{\text{sph}} = -1.47 \times 10^6 \Delta\vartheta \mathbf{I}, \quad (38)$$

$$\mathbf{T}_{\text{pro}} = -1.00 \times 10^6 \Delta\vartheta \text{diag} \{2.06, 2.06, 0.31\}, \quad (39)$$

$$\mathbf{T}_{\text{obl}} = -1.00 \times 10^6 \Delta\vartheta \text{diag} \{2.16, 1.13, 1.13\}, \quad (40)$$

where $\text{diag} \{k, l, n\}$ denotes diagonal matrix with diagonal elements k, l, n . It can be seen that the hydrostatic tension due to thermal expansion mismatch is as high as 1.47 GPa when the difference $\Delta\vartheta$ between the fabrication temperature ϑ_f and the working temperature ϑ is 1000°C . This residual stress has to be considered for the transformation yield and deformation of zirconia particles.

6.2. Transformation yielding of $t\text{-ZrO}_2$ particle under hydrostatic and uniaxial loadings

The transformation yielding of $t\text{-ZrO}_2$ particle under uniaxial loading in the absence of residual stress has been numerically investigated by Zhang *et al.* (1993). Here, we will look at how the residual thermal stresses effect the phase transformation of spherical $t\text{-ZrO}_2$ particle under hydrostatic and uniaxial loading.

Figure 3 consists of plots of uniaxial compressive yield stress Y_c versus hydrostatic pressure p for spherical particle. For the three loading directions considered, the uniaxial compressive yield stress Y_c is approximately linearly dependent on the hydrostatic pressure p for p being larger than 0.5 GPa. The linear coefficient α_c is between 2.1 to 2.3. The

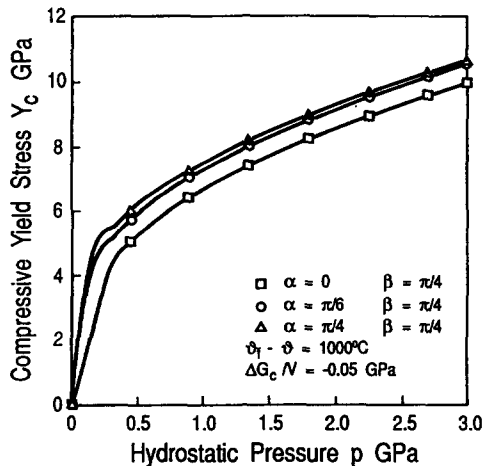


Fig. 3. Uniaxial compressive yield stress Y_c vs hydrostatic pressure p for spherical particle.

compressive yield stress at zero pressure is determined by critical energy parameter $\Delta G_{cr}/V$ and the chemical energy change $\Delta G_{ch}/V$. As we have pointed out in a previous paper (Zhang *et al.*, 1993), hydrostatic pressure and uniaxial compressive loading in some lattice direction cannot induce the $t \rightarrow m$ phase transformation because such compressive loading can never lower the free energy of the whole system. The hydrostatic pressure p always attempts to restrain from taking place the phase transformation which is accompanied with dilatational deformation. Its presence therefore enhances the yield stress.

Chen and Reyes-Morel (1986) proposed the following mixed yield criterion for composite ceramics :

$$T_e/T_{ec} + T_m/T_{mc} = 1, \quad (41)$$

where

$$T_e = (\frac{3}{2}\mathbf{T}' \cdot \mathbf{T}')^{1/2}, \quad T_m = \frac{1}{3}\mathbf{I} \cdot \mathbf{T}, \quad \mathbf{T}' = \mathbf{T} - T_m \mathbf{I}. \quad (42)$$

T_{ec} and T_{mc} are, respectively, the characteristic equivalent stress and the characteristic mean stress. According to eqn (41), under hydrostatic compression p , the uniaxial compressive Y_c is related to p by the linear relation :

$$Y_c = Y_c^0 + \alpha_c p, \quad (43)$$

where Y_c^0 is the compressive yield stress at zero pressure and α_c is a constant. It is found experimentally that α_c approaches 2 except at very small strains and later at very large strains (Chen and Reyes-Morel, 1986). The theoretical results for a single particle presented in Fig. 2 agree very well with these experimental results.

From eqn (5.2), the yield stress σ_{cr} depends on temperature ϑ through the residual thermal strain \mathbf{E}' and the chemical energy change $\Delta G_{ch}/V$. The former leads to a hydrostatic tension on the particle and therefore assists the phase transformation to take place; large thermal coefficients mismatch and large temperature difference will substantially reduce both the uniaxial tensile yield stress and the uniaxial compressive yield stress. The latter is a linear function of the temperature difference ($\vartheta_s - \vartheta$) as in eqn (26). The lower is the temperature ϑ , the larger is the chemical energy change $\Delta G_{ch}/V$ available for overcoming the energy barrier $\Delta G_{cr}/V$ and the lower stress σ_{cr} is required to assist the supposed phase transformation. Tensile yield stress σ_{cr} for spherical particle is plotted in Fig. 4 vs temperature ϑ for five different energy barriers in the absence of global hydrostatic compression, where $f_s = -0.235 \text{ MPa}^\circ\text{C}^{-1}$, $\vartheta_s = 1000^\circ\text{C}$, $\vartheta_f = 1200^\circ\text{C}$. The tensile yield stress for particles with given energy barrier is found to decrease linearly with decreasing temperature. This agrees well with the experimental results for ceramic composites on the comprehensive

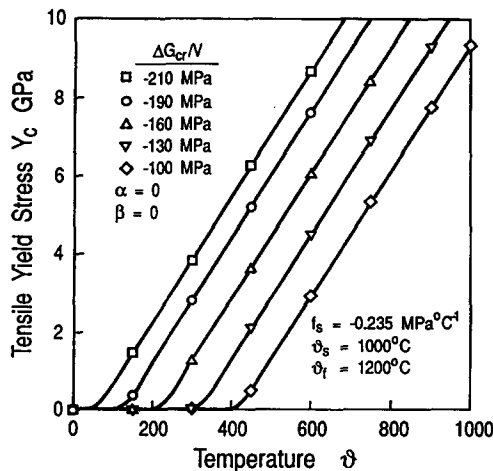


Fig. 4. Uniaxial tensile yield stress σ_{cr} vs temperature ϑ for spherical particle.

effect of temperature on transformation zone depth, a linearly decreasing function of the yield stress, by Swain *et al.* (1983) and Becher *et al.* (1987). It can also be seen that when temperature decreases, particles with a certain energy barrier may transform from tetragonal to monoclinic symmetry even without the assistance of external load.

The dependence of transformation yielding on loading direction under uniaxial loading is found to be similar to that in the absence of thermal expansion mismatch (Zhang *et al.*, 1993). In Fig. 5, the uniaxial tensile yield load is depicted against loading angle α with fixed $\beta = 0, \pi/6, \pi/4, \pi/3$ for spherical particle. Higher uniaxial tension along the A - B plane of the tetragonal lattice is needed for the transformation to take place than along the C axis. Maximum yield load of uniaxial tension usually occurs at $\alpha = \pi/4, \beta = 0$, while minimum tensile yield load occurs at $\alpha = 0, \beta = \pi/2$.

6.3. Transformation shear in the presence of multiply modes twinning

The maximum engineering shear strain defined by :

$$\gamma_m = E_3 - E_1, \quad (44)$$

is used to measure the shear of the transformation plastic deformation, where E_1, E_2 and E_3 are the principal tensions of \mathbf{E} in increasing order.

In Fig. 6, the shear deformation γ_m is depicted against the loading angle α with β fixed at $0, \pi/6, \pi/4, \pi/3$. For spherical inclusion [Fig. 6(a)], the shear strains depend approximately sinusoidally on the loading parameter $\alpha/2$ for $\beta = 0$, and become less sensitive to α for increasing β . For prolate inclusion with aspect ratio $\tau = 0.1$ [Fig. 6(b)], the shear strains are less sensitive to angle α than to angle β . Since the inclusion is geometrically symmetric with respect to any α rotation but not to β rotation, the dependence of the shear strains on β are credited to the shape-effect of transformation plasticity. For oblate inclusion with aspect ratio $\tau = 0.1$ [Fig. 6(c)], the shear strains are found to be intermediate, less than those for prolate inclusions with the same aspect ratio and usually larger than those for spherical inclusions. The shear deformation γ_m is at minimum 0.9% for oblate inclusion and at maximum 3.7% for prolate inclusion, for the physical and geometric parameters used. Similar numerical results can have also been obtained using a model of single twinning mode (Zhang and Lam, 1993). An important observation from the numerical calculations should be noted that the twinning modes for oblate inclusion investigated here are always $(\zeta, \zeta, \xi, \xi), \zeta + \xi = 0.5$ for loading direction $\beta = 0$ and $(\lambda, 1 - \lambda, 0, 0)$ or $(0, 0, \lambda, 1 - \lambda)$ for loading directions with larger β . Single mode twinning prevails for the latter. Multiple modes twinning usually happens in spherical and prolate inclusions.

To examine the dependence of the maximum shear deformation γ_m on the critical energy parameter $\Delta G_c/V$, in Fig. 7, the shear deformation γ_m is plotted against the yield parameter G_c/V : (a) for spherical particle, (b) for prolate spheroidal particle and (c) for oblate spheroidal particle. For all the three particle shapes considered, the transformation shear γ_m is found to increase with energy barrier $\Delta G_c/V$ for uniaxial loads when $\beta \neq 0$. The transformation shear γ_m is found to be independent of the energy barrier $\Delta G_c/V$ when the uniaxial loading is applied along the lattice plane $\beta = 0$. For a given temperature \mathcal{D} , the larger the critical energy parameter $\Delta G_c/V$, the larger the uniaxial tensile yield stress. Therefore, the particle transformation shear will increase with increasing yielding load. Moreover, the difference of the maximum transformation shear strain γ_m for uniaxial yield loading on the lattice A - B plane and for uniaxial yield loading off the plane will increase with the critical energy parameter $\Delta G_c/V$. Comparing Figs 7(a) and 7(c) with Fig. 7(b), we can see that the transformation shear γ_m due to uniaxial loading off the lattice A - B plane ($\beta \neq 0$) for prolate spheroidal particles will increase with the critical energy parameter $\Delta G_c/V$, or say, the yield load, much faster than those for spherical particles and for oblate spheroidal particles.

7. CONCLUSIONS AND REMARKS

The transformation twinning of a single ellipsoidal zirconia particle embedded in a three-dimensional infinite matrix is considered in order to model the transformation shears

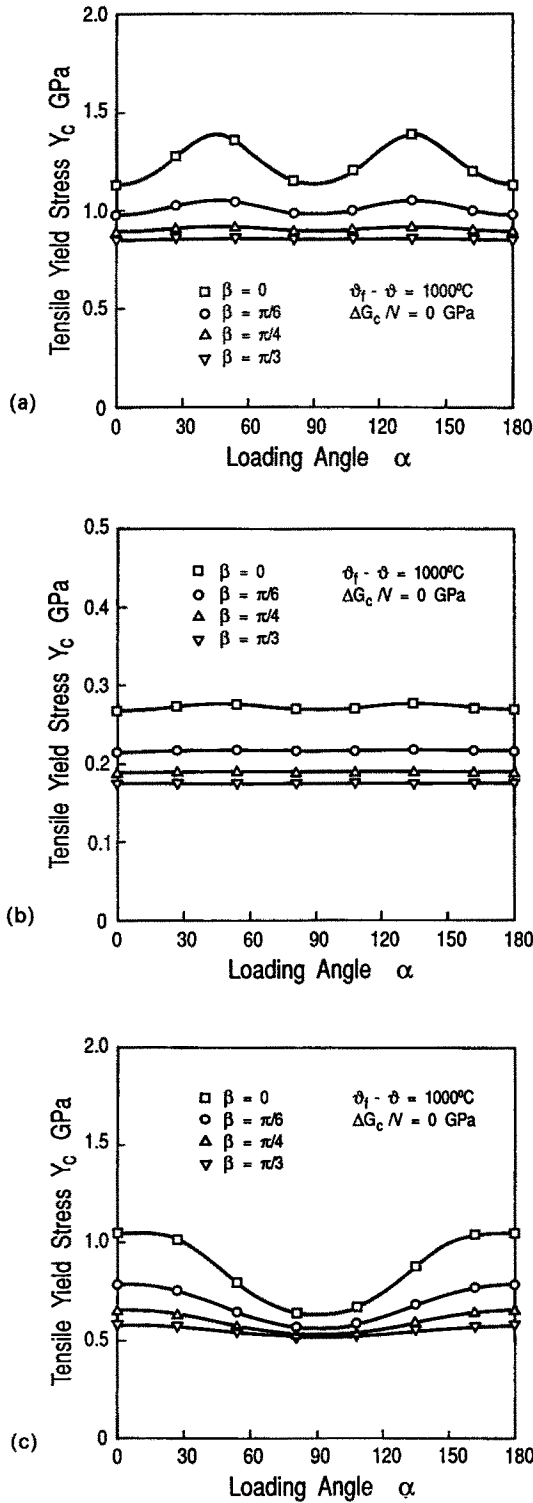


Fig. 5. Uniaxial tensile yield stress σ_{cr} vs loading angle α : (a) for spherical particle, (b) for prolate spheroidal particle and (c) for oblate spheroidal particle.

in zirconia-toughened ceramics. A model of multiple twinning systems has been proposed for the determination of the shear deformation of the zirconia particle transforming from tetragonal to monoclinic phase, in the presence of thermal expansion mismatch between particle and matrix. It has been established that the particle–matrix system assumes a state of minimum potential energy change after the transformation. The transformation yield

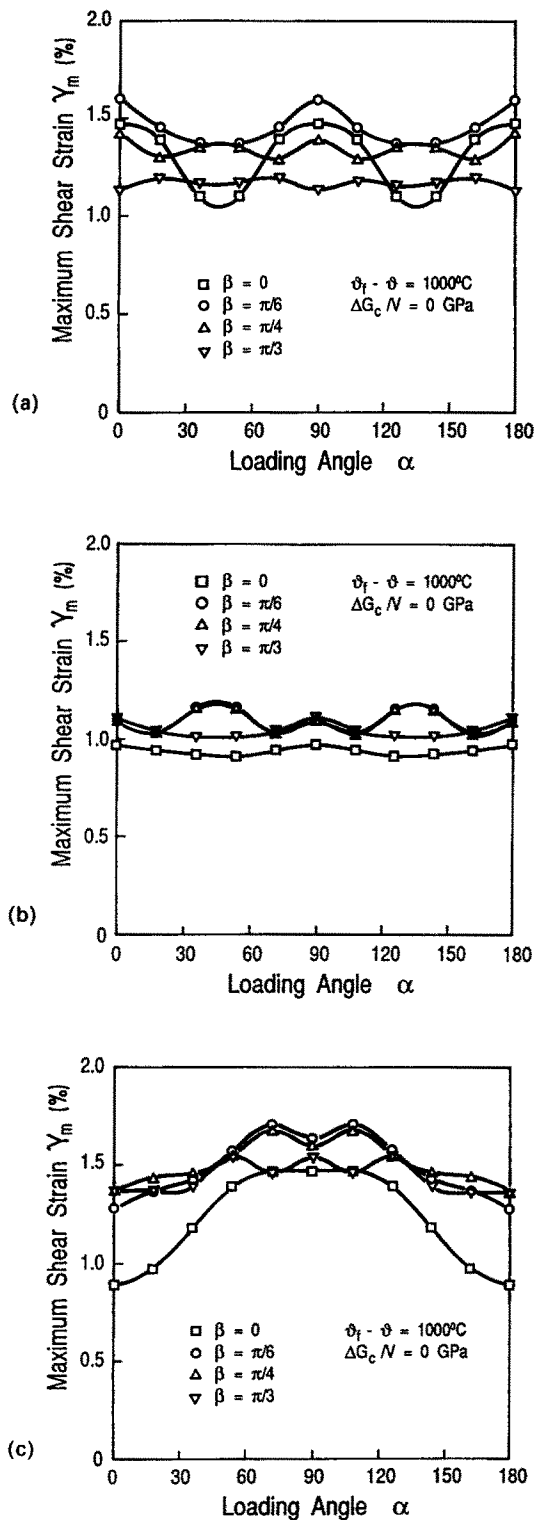


Fig. 6. Transformation shear deformation γ_m vs loading angle α : (a) for spherical particle, (b) for prolate spheroidal particle and (c) for oblate spheroidal particle.

behavior under various loading conditions is determined by incorporating the minimum free energy principle with the end-point thermodynamics of phase transformation. The macroscopic particle deformation which is assumed to be homogeneous inside the particle is uniquely determined by the volume fractions of the twinned martensitic variants.

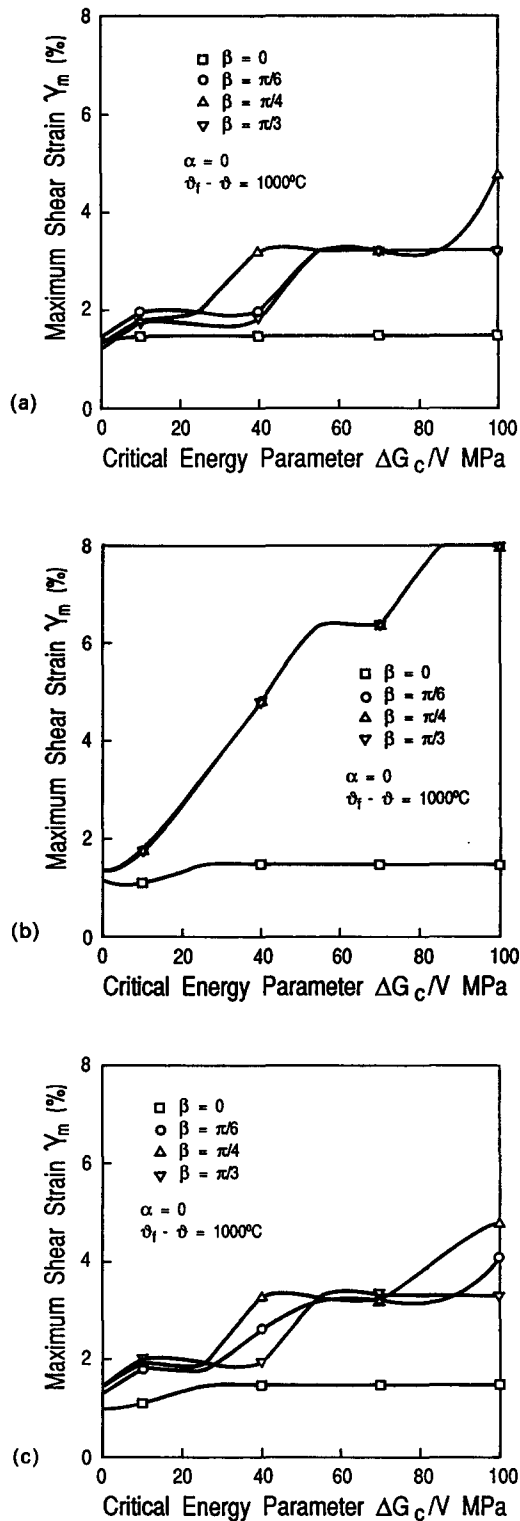


Fig. 7. Transformation shear deformation γ_m vs energy barrier $\Delta G_c/V$: (a) for spherical particle, (b) for prolate spheroidal particle and (c) for oblate spheroidal particle.

Numerical results according to this model agree very well with experiments. The uniaxial compressive and tensile yield stresses depend linearly on the hydrostatic pressure with a linear coefficient between 2.1 and 2.3. Yield stresses are shown to be linear dependent on temperature ϑ . For particles transformed under uniaxial tensile stress around 0.2 to 1.5 GPa, the transformation twinning will substantially reduce the maximum particle shear

strain γ_m from 16% to between 0.9% and 4.0%. The transformation twinning is shown to be dependent on yield stress, particle shape, working temperature, stress-free transformation temperature ϑ_s , fabrication temperature ϑ_f , thermal expansion mismatch and the thermodynamics coefficient f_s .

It seems very difficult, if not impossible, to relate experimentally the transformation particle shear strain to the applied load for the case of a single particle. We are unable to find such experimental results in the literature. To assess the validity of the theory developed, some average method such as Mori–Tanaka method or self-consistent method should be used to build a bridge between the idealized single particle model and the composite ceramics toughened with randomly distributed and orientated or unidirectionally aligned zirconia particles. Once this has been done, the experimental results on composite ceramics about the effects on transformation yielding and transformation shear of various geometrical and physical parameters can be used to compare with the theoretical expectations based on the single particle model. We will investigate this issue in a separate paper (Lam and Zhang, 1993).

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