A thermodynamics based damage mechanics model for particulate composites

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Abstract

A micro-mechanical damage model is proposed to predict the overall viscoplastic behavior and damage evolution in a particle filled polymer matrix composite. Particulate composite consists of polymer matrix, particle fillers, and an interfacial transition interphase around the filler particles. Yet the composite is treated as a two distinct phase material, namely the matrix and the equivalent particle-interface assembly. The CTE mismatch between the matrix and the filler particles is introduced into the model. A damage evolution function based on irreversible thermodynamics is also introduced into the constitutive model to describe the degradation of the composite. The efficient general return-mapping algorithm is exploited to implement the proposed unified damage coupled viscoplastic model into finite element formulation. Furthermore, the model predictions for uniaxial loading conditions are compared with the experimental data.

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1. Introduction

The application and estimation of the effective mechanical properties of random heterogeneous multiphase materials are of great interest to researchers and engineers in many science and engineering disciplines. There are many different methods and tools that can be used to deliver the macroscopic constitutive response of heterogeneous materials using a local description of the micro-structural behavior (Ju and Lee, 2000, 2001; Ju and Sun, 2001; Ju, 1990a,b; Ju and Tseng, 1997; Ju and Chen, 1994a,b; Wong and Ait-Kadi, 1997; Willis, 1991; Voyiadjis and Thiarajan, 1996; Sun and Ju, 2001; Ravichandran and Liu, 1995; Kwon and Liu, 1998; Ahmed and Jones, 1990; Achenbach and Zhu, 1989. In the development of the homogenization procedures for heterogeneous materials, we have to define both the homogenization step itself (from local variables to overall ones) and the often more complicated localization step (from overall controlled quantities to the corresponding local ones).

The nature of the bond between particles and the matrix material has a significant effect on the mechanical behavior of particulate composites. Most analytical and numerical models assume that the bond between the

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filler and matrix is perfect and can be modeled using the continuity of tractions and displacements across a discrete interface. However, internal defects and imperfect interfaces are well known to exist in composites. The incorporation of such phenomena into the general theory requires modification and relaxation of the continuity of displacements between the constituents (Nie and Basaran, 2005). The imperfect interface bond may be due to the compliant interfacial layer known as interphase or interface damage, which may have been created deliberately by coating the particles. It may also develop during the manufacturing process due to chemical reactions between the contacting particles and the matrix material or due to interface damage from cycle loading. Moreover, the strength of the bond at the interface controls the mechanical response and fatigue life of the composite (Basaran et al., 2006). By controlling the stress–strain response of the interphase, it is possible to control the overall behavior of the composite.

Based on the generalizations of the Eshelby method (Eshelby, 1957), a novel micro-mechanical framework has been proposed by Ju and Chen (1994a,b) to investigate the effective mechanical properties of elastic multiphase composites containing many randomly dispersed ellipsoidal inhomogeneities. Within the context of the representative volume element (REV), three governing micro-mechanical ensemble-volume averaged field equations are presented to relate ensemble-volume averaged stresses, strains, volume fractions, eigenstrains, particle shapes and orientations, as well as elastic properties of constituent phases of linear elastic particulate composites. Then, various micro-mechanical models have been developed based on the ensemble-volume averaged constitutive equations. For example, in Ju and Tseng (1996), a formulation combining a micro-mechanical interaction approach and the continuum plasticity is proposed to predict effective elastoplastic behavior of a two-phase particulate composite containing many randomly dispersed elastic spherical inhomogeneities. Explicit pairwise inter-particle interactions are considered in both the elastic and plastic responses. Furthermore, the ensemble-volume averaging procedure is employed and the formulation is of complete second order.

Constituents of particle filled composites often have very different properties such as the coefficient of thermal expansion (CTE), ductility, and elastic modulus. It is well known that within the particulate composite microstructure there exists a micro-stress associated with the CTE mismatch between the matrix and the filler particles. For a composite made up of lightly cross-linked poly-methyl methacrylate (PMMA) matrix filled with alumina trihydrate (ATH) particles, these micro-stresses can be imaged using the fact that PMMA is optically birefringent under stresses. The micro-stresses imaged using this technique are indeed due to CTE mismatch because they dissipate as the temperature gets close to the \( T_g \) of the PMMA (as shown in Fig. 1). These stresses can be quantified by calculation or by direct measurement and are of the order between 15% and 75% of the tensile strength of the composite. Therefore the thermal stress associated with the CTE mismatch between the matrix and particles is an important factor for the failure of particulate composites subjected to the thermo-mechanical loads. However, the original micro-mechanical models of Ju and Chen (1994a,b), Ju and Tseng (1996), do not account for the effects of the CTE mismatch between the filler particles and the matrix. Lee (1992) analyzed metal matrix composite for strains induced by CTE mismatch. He reported that spherical reinforcement particle to be in hydrostatic stress state and remains in the elastic state. Yet,
Lee (1992) reported that stresses and strains are the largest and plastic deformation occurs in the matrix adjacent to the reinforcement particle. Accordingly, he reported, the reinforcement particle/matrix interface becomes a potential crack initiation site. Taya (1991) reported strengthening mechanisms of metal matrix composites from macro- and nanolevel models. Taya (1991) observed that in both macro- and nanolevel models the residual stresses induced due to CTE mismatch strain, between the filler and the matrix, play an important role in influencing the yield stress of the composite.

Chatuvedi and Shen (1998) proposed a micro-mechanical characterization for thermal expansion of particle filled plastic encapsulant material where they accounted for different CTE values of the filler and the matrix. Yet their model is based on theory of elasticity therefore viscoplastic damage modeling is not possible. Chen et al. (1982) studied effect of residual thermal stresses due to CTE mismatch on thermal conductivity of W–Cu composites, experimentally. They reported that there are significant thermal stresses at the interface due to CTE mismatch. Liu et al. (2005) conducted finite element simulation of the thermal properties of particulate composites, experimentally. They reported that there are significant thermal stresses at the interface due to CTE mismatch. Liu et al. (2005) conducted finite element simulation of the thermal properties of particulate and continuous network-reinforced metal–matrix composites. They calculated effective CTE using FEM discretization. Agrawal et al. (2003) measured thermal residual stresses in two types of co-continuous composites. They used FEM to simulate the state of stress at the interfaces, where CTE mismatch led to tensile thermal stresses at the metal–ceramic interfaces. Kitazono et al. (2001) proposed micro-mechanics models for CTE-mismatch super-plasticity for metal matrix composites and monolithic poly-crystals. Vo et al. (2001) presented a novel model for predicting the effective CTE of particle filled epoxy-polymer (underfill) composites by considering the effect of an interphase zone surrounding the filler particles in polymer matrix. Xie et al. (2001) measured the CTE values of TiC particulate reinforced ZA43 composites. They reported that thermal stresses developed as a result of CTE mismatch between the reinforcement and the matrix. Elemori et al. (1998) reported observed thermal stresses as a result of CTE mismatch between the reinforcement and the matrix.

It is well known that the interfacial bond properties between the matrix and particles have a significant effect on the behavior of the composite. In order to directly use the micro-mechanical field equations (Mura, 1987) and also to consider the CTE mismatch and interfacial bond properties between the matrix and filler particles, the composite is treated as two distinct phases: namely the bulk matrix and the equivalent particle-interface-region assembly. In the proposed model, the interface region is given a thickness and effective elastic properties of its own. In this paper, we expand Ju and Chen’s micro-mechanical model to incorporate the effects of CTE mismatch between the matrix and the filler particles.

2. Micro-mechanical field equations

In order to obtain effective constitutive equations and properties of random heterogeneous composites, one typically performs the ensemble-volume averaging process within a mesoscopic representative volume element (RVE). The volume-averaged stress tensor is defined as

\[ \bar{\sigma} = \frac{1}{V} \int_V \sigma(x) \, dx = \frac{1}{V} \left[ \int_{V_0} \sigma(x) \, dx + \sum_{q=1}^n \int_{V_q} \sigma(x) \, dx \right] \]

(1)

where \( V \) is the volume of a RVE, \( V_0 \) is the volume of the matrix, \( V_q \) is the volume of the \( q \)-th phase particles, and \( n \) denotes the number of particulate phases of different material properties (excluding the matrix).

Similarly, the volume-averaged strain tensor is defined as follows:

\[ \bar{\varepsilon} = \frac{1}{V} \int_V \varepsilon(x) \, dx = \frac{1}{V} \left[ \int_{V_0} \varepsilon(x) \, dx + \sum_{q=1}^n \int_{V_q} \varepsilon(x) \, dx \right] \equiv \frac{1}{V} \left[ V_0 \bar{\varepsilon}_0 + \sum_{q=1}^n V_q \bar{\varepsilon}_q \right] \]

(2)

According to Eshelby’s equivalence principle, the perturbed strain field \( \varepsilon'(x) \) induced by inhomogeneities can be related to specified eigenstrain \( \varepsilon^*(x) \) by replacing the inhomogeneities with the matrix material. The inhomogeneities may also involve their own eigenstrain caused by phase transformation, precipitation, plastic deformation, or CTE mismatch between different constituents of the composites. However, it is not essential to attribute the eigenstrain to any specific source. Then the stress is the sum of the two parts, one caused by the inhomogeneity, and the other by the eigenstress associated with eigenstrain. For the domain of the \( q \)-th phase...
particles with an elastic stiffness tensor $C_q$, Mura (1987) and Mura and Tanaka (1973) proposed the following stress relation:

$$ C_q \cdot [ \varepsilon^0 + \varepsilon'(\mathbf{x}) - \varepsilon^T_q(\mathbf{x}) ] = C_0 \cdot [ \varepsilon^0 + \varepsilon'(\mathbf{x}) - \varepsilon^T_q(\mathbf{x}) - \varepsilon^*_q(\mathbf{x}) ] $$

(3)

where $C_0$ is the stiffness tensor of the matrix and $\varepsilon^0$ is the uniform elastic strain field induced by far-field loads for a homogeneous matrix material only. $\varepsilon^*_q$ is its own eigenstrain associated with the $q$th particle. $\varepsilon^T_q$ is the fictitious equivalent eigenstrain by replacing the $q$th particle with the matrix material. $\varepsilon'(\mathbf{x})$ is the perturbed strain due to distributed eigenstrain $\varepsilon^T_q$ and $\varepsilon^*_q$ associated with all particles in the RVE.

The strain at any point within an RVE is decomposed into two parts, the uniform strain and the perturbed strain due to the distributed eigenstrain. It is emphasized that the eigenstrains $\varepsilon^*$ and $\varepsilon^T$ are non-zero in the particle domain and zero in the matrix domain, respectively. In particular, in accordance with the Eshelby principle, the perturbed strain field induced by all the distributed eigenstrains $\varepsilon^*$ and $\varepsilon^T$ can be expressed as (Mura, 1987)

$$ \varepsilon'(\mathbf{x}) = \int_V \mathbf{G}(\mathbf{x} - \mathbf{x}') \cdot [ \varepsilon^T(\mathbf{x}') + \varepsilon^*(\mathbf{x}') ] d\mathbf{x}' $$

(4)

where $\mathbf{x}, \mathbf{x}' \in V$, and $\mathbf{G}$ is the Green’s function in a linear elastic homogeneous matrix.

From Eqs. (3) and (4), we arrive at

$$ -\mathbf{A}_q \cdot \varepsilon^*_q(\mathbf{x}) = \varepsilon^0 - \varepsilon^T_q(\mathbf{x}) + \int_V \mathbf{G}(\mathbf{x} - \mathbf{x}') \cdot [ \varepsilon^T(\mathbf{x}') + \varepsilon^*(\mathbf{x}') ] d\mathbf{x}', \quad \mathbf{x}' \in V $$

(5)

where $\mathbf{A}_q$ is given by

$$ \mathbf{A}_q = (C_q - C_0)^{-1} \cdot C_0 $$

(6)

Using the renormalization procedure proposed by (Ju and Chen, 1994a), for spherical inclusions, the three basic governing micro-mechanical ensemble-volume averaged field equations can be derived as follows:

$$ \bar{\sigma} = C_0 \cdot \left[ \bar{\varepsilon} - \sum_{q=1}^n \phi_q \left( \bar{\varepsilon}^*_q + \bar{\varepsilon}^T_q \right) \right] $$

(7)

$$ \bar{\varepsilon} = \varepsilon^0 + \sum_{q=1}^n \phi_q \mathbf{S} \cdot \left( \bar{\varepsilon}^T_q + \bar{\varepsilon}^*_q \right) $$

(8)

$$ - (\mathbf{A}_q + \mathbf{S}) \cdot \varepsilon^*_q = \varepsilon^0 - \varepsilon^T + \mathbf{S} \cdot \bar{\varepsilon}^T_q + \bar{\varepsilon}^p_q $$

(9)

where $\mathbf{S}$ is the Eshelby tensor, and $\bar{\varepsilon}^p_q$ represents the inter-particle interaction effects. In order to actually solve Eqs. (7)–(9) and to obtain effective elastic properties of composites, it is essential to express the $q$th-phase average eigenstrain $\bar{\varepsilon}^*_q$ in terms of the average strain $\bar{\varepsilon}$. Namely, one has to solve the integral Eq. (5) exactly for each phase, which requires details of the random microstructure.

3. Effective thermo-mechanical properties of composite sphere assemblage

In many particulate composites, a thin layer of some other elastic phase intervenes between a particle and the matrix. The imperfect interface bond may be due to the very compliant thin interfacial layer that is assumed to have perfect boundary conditions with the matrix and the particle. This defines a three-phase composite that includes particles, thin interphase, and the matrix as shown in Fig. 2. Once the effective mechanical and thermal properties of the inner composite sphere assemblage (CSA), which consists of the particle and interphase layer of thickness $\delta$, are found, the composite models used for the perfect interface composite model can be readily applied to this case of imperfect interface conditions.

The composite spheres assemblage (CSA) model was first proposed by Kerner (1956) and Van der Poel (1958) as shown in Fig. 3. Smith (1974, 1975), Christensen and Lo (1979) and Hashin and his co-workers (1962, 1963, 1968, 1990, and 1991a,b) eventually improved on the CSA model. The CSA assumes that the particles are spherical and, moreover, that the action on the particle is transmitted through a spherical interphase
shell. The overall macro-behavior is assumed to be isotropic and is thus characterized by two effective moduli: the bulk modulus \( k \) and the shear modulus \( \mu \). In this section, I summarized the theoretical solution for effective thermo-mechanical properties of the CSA consisting of an elastic spherical particle and an elastic interphase layer with a thickness of \( \delta \). In the following formulae, \( k \) represents the bulk modulus, \( \mu \) represents shear modulus, \( \alpha \) represents the coefficient of thermal expansion. The subscripts \( i \), \( f \) and \( m \) refer to the interphase, filler and matrix, respectively.

### 3.1. Effective bulk modulus

The effective bulk modulus \( k_1 \) for the CSA as obtained by Hashin (1962) is given as

\[
k_1 = k_i + \frac{\phi}{\frac{1}{k_i-k_f} + \frac{1}{k_i+k_f}}
\]

where

\[
\phi = \left( \frac{r}{r+\delta} \right)^3
\]

\( r \) is the radius of the filler particle and \( \delta \) is the thickness of interphase.

### 3.2. Effective coefficient of thermal expansion (ECTE)

The effective coefficient of thermal expansion \( \alpha_1 \) for the CSA as obtained by Levin (1967) is given as

\[
\alpha_1 = \alpha_i + \frac{\alpha_i - \alpha_f}{\frac{1}{k_i-k_f} + \frac{1}{k_i+k_f}} \left( \frac{1}{k_i} - \frac{1}{k_i} \right)
\]

### 3.3. Effective shear modulus

Based on the generalized self-consistent scheme (GSCS) model proposed by (Hashin, 1962), Christensen and Lo (1979) have given the condition for determining the effective shear modulus of CSA as follows:
where

\[ A = 8\frac{\mu_1}{\mu_2}(4 - 5\nu_1)\eta_1\phi^{10/3} - 2\{63\frac{\mu_1}{\mu_2}(4 - 5\nu_1)\eta_2 + 2\eta_1\eta_3\phi^{7/3} \\
+ 252\frac{\mu_1}{\mu_2}(4 - 5\nu_1)\eta_3\phi^{5/3} - 50\frac{\mu_1}{\mu_2}(4 - 5\nu_1)\eta_2 + 8\nu_2 \} \eta_2 + 4(7 - 10\nu_1)\eta_2\eta_3 \]

\[ B = -4\frac{\mu_1}{\mu_2}(4 - 5\nu_1)\eta_1\phi^{10/3} - 4\{63\frac{\mu_1}{\mu_2}(4 - 5\nu_1)\eta_2 + 2\eta_1\eta_3\phi^{7/3} \\
- 504\frac{\mu_1}{\mu_2}(4 - 5\nu_1)\eta_3\phi^{5/3} + 150\frac{\mu_1}{\mu_2}(4 - 5\nu_1)\eta_2 + 3(15\nu_1 - 7)\eta_2\eta_3 \]

\[ D = 4\frac{\mu_1}{\mu_2}(4 - 5\nu_1)\eta_1\phi^{10/3} - 2\{63\frac{\mu_1}{\mu_2}(4 - 5\nu_1)\eta_2 + 2\eta_1\eta_3\phi^{7/3} \\
+ 252\frac{\mu_1}{\mu_2}(4 - 5\nu_1)\eta_3\phi^{5/3} + 25\frac{\mu_1}{\mu_2}(4 - 5\nu_1)\eta_2\phi - (7 + 5\nu_1)\eta_2\eta_3 \]

with

\[ \eta_1 = \frac{\mu_1}{\mu_2}(49 - 50\nu_1\nu_1) + 35\frac{\mu_1}{\mu_2}(\nu_2 - 2\nu_1) + 35(2\nu_1 - \nu_1) \]

\[ \eta_2 = 5\nu_1\frac{\mu_1}{\mu_2}(4 - 5\nu_1) + 7\frac{\mu_1}{\mu_2} + 4 \]

\[ \eta_3 = (\frac{\mu_1}{\mu_2})(8 - 10\nu_1) + (7 - 5\nu_1) \]

\( \phi \) is given in Eq. (11).

4. Viscoelastic behavior of two-phase composites

Let us consider a perfectly bonded two-phase composite consisting of a viscoplastic matrix (phase 0) with an elastic bulk modulus \( k_0 \) and elastic shear modulus \( \mu_0 \), and randomly dispersed elastic spherical particle-interphase assembly (phase 1) with effective bulk modulus \( k_1 \) and effective shear modulus \( \mu_1 \). The effective elastic moduli of the two-phase composite by neglecting the inter-particle interaction effects has been derived by Ju and Chen (1994a) using the field equations given above without the CTE mismatch between the filler particles and the matrix:

\[ k = k_0 \left( 1 + \frac{3(1 - \nu_0)(k_1 - k_0)\phi}{3(1 - \nu_0)k_0 + (1 - \phi)(1 + \nu_0)(k_1 - k_0)} \right) \]

\[ \mu = \mu_0 \left( 1 + \frac{15(1 - \nu_0)(\mu_1 - \mu_0)\phi}{15(1 - \nu_0)\mu_0 + (1 - \phi)(8 - 10\nu_0)(\mu_1 - \mu_0)} \right) \]

where \( \phi \) is the particle volume fraction, \( \nu_0 \) is the Poisson’s ratio of the matrix.

For simplicity, the Von Mises yield criterion is assumed for the matrix. Accordingly, at any matrix material point, the stress \( \sigma \) and the equivalent plastic strain \( \varepsilon \) must satisfy the following yield criterion:

\[ F(\sigma, \varepsilon) = \sqrt{\sigma : \sigma : \sigma} - \sqrt{3/2}\sigma_y(\varepsilon) \]

where \( \sigma_y(\varepsilon) \) is the isotropic hardening function of the matrix-only material, and \( \sigma_d \) denotes the deviatoric part of the fourth rank identity tensor.

In order to solve for the elastoplastic response exactly, the stress at any local point has to be determined and used to determine the plastic response through the local yield criterion. This approach is generally infeasible due to the complexity of accounting for all possible configurations statistically and micro-structurally. Therefore, a framework in which an ensemble averaged yield criterion is constructed for the entire composite is used instead (Ju and Tseng, 1996). The averaged current stress norm in a matrix point \( x \) can be derived as

\[ \langle H \rangle_m(x) = (\sigma - \sigma^T) : T : (\sigma - \sigma^T) \]

\[ \sigma^T = AC_1 : e^T \]

where \( e^T \) is the uniform eigenstrain associated with CTE mismatch between matrix and particles, and the components of \( T \) are given as

\[ T_{ijkl} = T_1\delta_{ij}\delta_{kl} + T_2(\delta_{ik}\delta_{jl} + \delta_{ij}\delta_{lk}) \]
with

\[ 3\mathbf{T}_1 + 2\mathbf{T}_2 = \frac{200\phi(1 - 2v_0)^2}{(3\alpha + 2\beta)^2(a\phi + 1)^2} \] (26)

\[ \mathbf{T}_2 = \frac{1}{2} + \frac{(23 - 50v_0 + 35v_0^2)\phi}{(b\phi + 1)^2} \] (27)

\[ \alpha = 2(5v_0 - 1) + 10(1 - v_0) \left( \frac{k_0}{k_1 - k_0} - \frac{\mu_0}{\mu_1 - \mu_0} \right) \] (28)

\[ \beta = 2(4 - 5v_0) + 15(1 - v_0) \frac{\mu_0}{\mu_1 - \mu_0} \] (29)

\[ a = \frac{20(1 - 2v_0)}{3\alpha + 2\beta} \] (30)

\[ b = \frac{(7 - 5v_0)}{\beta} \] (31)

In this work it is assumed that viscoplastic yielding and flow occur only in the matrix, and the matrix solely determines the viscoplastic behavior of the composite. The magnitude of the current equivalent stress norm of the matrix can be utilized to determine the possible viscoplastic behavior in the composite. When the ensemble-volume averaged current stress norm in the matrix reaches a certain level, the composite undergoes viscoplastic flow. The effective yield function for the composite in the presence of CTE mismatch induced stresses can be written as

\[ f(\mathbf{\sigma}, \dot{\mathbf{\sigma}}) = \sqrt{(\mathbf{\sigma} - \mathbf{\sigma}^T)} : \mathbf{T} : (\mathbf{\sigma} - \mathbf{\sigma}^T) - \sqrt{\mathbf{T}_1 + 2\mathbf{T}_2} \sigma_y(\bar{\mathbf{e}}) \] (32)

where \( \sigma_y(\bar{\mathbf{e}}) \) is the isotropic hardening function of the composite materials, \( \bar{\mathbf{e}} \) is the equivalent viscoplastic strain that defines isotropic hardening of the yield surface of the composites, and

\[ \dot{\mathbf{e}}^p = \mathbf{T}^{-1} : \dot{\mathbf{e}}^p \] (33)

The factors in the effective yield and effective plastic strain increment equations are chosen so that the effective stress and effective plastic strain increments are equal to the uniaxial stress and uniaxial plastic strain increment in a tensile test. It should be noted that the effective yield function is pressure dependent now (as a result of accounting for CTE mismatch) and not of the Von-Mises type any more. Therefore, the particles have significant effect on the viscoplastic behavior of the matrix.

In order to simulate damage evolution behavior of composite materials, there is a need for introduction of a damage parameter in the above proposed constitutive model. Damage mechanics provides us a basic framework to develop damage evolution models (Lemaitre, 1996). According to the strain equivalence principle, the effective damage coupled yield function for an isotropic damage parameter \( D \) can be written as follows:

\[ f(\mathbf{\sigma}, \bar{\mathbf{e}}) = \sqrt{\left( \mathbf{\sigma} - \mathbf{\sigma}^T \right) : \mathbf{T} : \left( \mathbf{\sigma} - \mathbf{\sigma}^T \right) - \sqrt{\mathbf{T}_1 + 2\mathbf{T}_2} \sigma_y(\bar{\mathbf{e}})} \] (34)

where \( D \) is the damage parameter. It is obvious that the damage increases the equivalent stress norm of the composite, which tends to amplify the viscoplastic flow of the composite. For simplicity, the Perzyna-type viscoplasticity model is employed to characterize the rate dependency (viscosity) behavior of the matrix. It is assumed that viscoplastic flow that takes place in the matrix follows the Perzyna model, where creep strain rate is proportional to over-stress and viscosity coefficient of the material. Therefore, the effective ensemble-volume averaged plastic strain rate for the composite can be expressed as

\[ \dot{\mathbf{e}}^p = \mathbf{\gamma} \mathbf{n} \] (35)
where
\[
n = \frac{\mathbf{T}(\frac{\mathbf{a}}{1 - D} - \mathbf{C}^T)}{\sqrt{\left(\frac{\mathbf{a}}{1 - D} - \mathbf{C}^T\right)^T : \mathbf{T} : \left(\frac{\mathbf{a}}{1 - D} - \mathbf{C}^T\right)}}
\]
and \( \gamma \) denotes the plastic consistency parameter
\[
\gamma = \frac{\langle f \rangle}{\eta} = \frac{\langle f \rangle}{2\mu\tau}
\]
\( \eta \) is viscosity coefficient, \( \tau \) is relaxation time. And the equivalent plastic strain is defined as
\[
\dot{\varepsilon} = \sqrt{\frac{T_1}{1 - D} + 2T_2\frac{\gamma}{1 - D}}
\]

5. Damage evolution function

In solids, the entropy production is a non-negative intrinsic quantity based on irreversible thermodynamics and thus can serve as a basis for the systematic description of the irreversible processes occurring in a solid. Isotropic damage evolution function based on entropy production was proposed and experimentally validated by Basaran and Yan (1998), Basaran and Tang (2002), Tang and Basaran (2003), Basaran et al. (2003), Basaran and Nie (2004) and Gomez and Basaran (2005). The damage evolution function is given by
\[
D = D_c \left[ 1 - e^{-\frac{\Delta s}{\Delta s}} \right] \tag{39}
\]
where \( D_c \) is a proportionally critical damage coefficient, \( R \) is the gas constant, and \( \Delta s \) is the internal entropy production and can be calculated as follows:
\[
\Delta s = \int_{t_0}^{t} \left( \frac{\mathbf{\varepsilon} : \mathbf{\varepsilon}}{T \rho} \right) dt + \int_{t_0}^{t} \left( \frac{k}{T^2 \rho} \left| \text{grad} T \right|^2 \right) dt + \int_{t_0}^{t} r \frac{T}{T} dt \tag{40}
\]
where \( \rho \) is the density, \( T \) is temperature, \( k \) is the thermal conductivity of composite, \( r \) is the distributed internal heat production rate.

6. Computational integration algorithms

In this section, we employ the general return-mapping algorithm to solve for the unified damaged coupled viscoplastic corrector problem proposed above. The general return-mapping algorithm was proposed by Simo and Taylor (1985) for rate independent elastoplasticity and summarized in details by Simo and Hughes (1998). In order to minimize the confusion, the symbol \( \Delta \) is used to denote an increment over a time step, or an increment between successive iterations. For the rate-of-slip \( \gamma \), we adhere to the conventions: \( \Delta \gamma = \gamma \Delta t \) denotes the increment of \( \gamma \) over a time step, and \( \Delta^2 \gamma \) denotes the increment of \( \Delta \gamma \) between iterations.

Let \( \mathbf{C} \) be the elastic stiffness matrix of the composite, then Hooke’s law yields
\[
\mathbf{\sigma}_{n+1} = (1 - D)\mathbf{C} : (\mathbf{\varepsilon}_{n+1} - \mathbf{\varepsilon}_{n+1}^{ep})
\]
Since \( \mathbf{\varepsilon}_{n+1}^{ep} \) is fixed during the return-mapping stage, it follows that
\[
\Delta \mathbf{\varepsilon}^{ep} = -\frac{1}{1 - D} \mathbf{C}^{-1} \Delta \mathbf{\sigma}
\]
From Eqs. (39) and (40), we have
\[
\Delta D = -\frac{1}{1 - D} \frac{D_cm_s}{T \rho R} \exp \left( -\frac{m_s}{R} \Delta s \right) \mathbf{C}^{-1} \Delta \mathbf{\sigma}
\]
From Eqs. (35) and (38) we also have

\[\varepsilon_{n+1}^p = \varepsilon_n^p + \frac{1}{1 - D} \Delta \gamma n\]

\[\alpha_{n+1} = \alpha_n + \frac{1}{1 - D} \sqrt{T_1 + 2T_2} \Delta \gamma\]

(44) (45)

Let us define the residual functions as follows:

\[R_f = \frac{2 \mu \Delta \gamma}{\Delta t} - \langle f \rangle\]

\[R_c = -\varepsilon_{n+1}^p + \varepsilon_n^p + \frac{1}{1 - D} \Delta \gamma n\]

\[R_s = -\varepsilon_{n+1}^p + \varepsilon_n^p + \frac{1}{1 - D} \sqrt{T_1 + 2T_2} \Delta \gamma\]

(46) (47) (48)

If we linearize these residual functions and solve them, we obtain

\[\Delta^2 \gamma_{n+1} = \frac{-(1 - D)(R_f + \sqrt{T_1 + 2T_2}KR_s) - n': E : R_c}{(T_1 + 2T_2)K + \frac{2(1 - D)\mu}{\Delta t} + \frac{1}{1 - D} n': E : n}\]

\[\Delta \bar{\sigma}_{n+1} = -E \left( R_c + \frac{\Delta^2 \gamma_{n+1}}{1 - D} n \right)\]

\[\Delta \bar{\varepsilon}_{n+1}^p = -\frac{1}{1 - D} C^{-1} \Delta \bar{\sigma}_{n+1}\]

\[\Delta^2 s_{n+1} = \frac{D_{\alpha m_s}}{T \rho R} \exp \left( -\frac{m_s}{R} \Delta s_{n+1} \right) \left( \sigma_{n+1} \Delta \bar{\varepsilon}_{n+1} \right)\]

\[\Delta \bar{z}_{n+1} = R_s + \frac{1}{1 - D} \sqrt{T_1 + 2T_2} \Delta^2 \gamma_{n+1} + \frac{1}{(1 - D)^2} \sqrt{T_1 + 2T_2} \Delta \gamma_{n+1} \Delta D_{n+1}\]

(49) (50) (51) (52) (53) (54)

where

\[E^{-1} = \frac{1}{1 - D} \left[ C^{-1} + \frac{\Delta \gamma N}{1 - D} - \frac{D_{\alpha m_s} \Delta \gamma}{(1 - D)^2 T \rho R} \exp \left( -\frac{m_s}{R} \Delta s \right) \left( n + \frac{1}{1 - D} N \sigma \right) \otimes (\sigma C^{-1}) \right]\]

\[N = \frac{T \otimes n - \nabla}{\sqrt{(\nabla^2 - \sigma^2) : T : (\sigma^2 - \sigma) \otimes \sigma}}\]

\[n' = n - \frac{D_{\alpha m_s}}{(1 - D)^2 T \rho R} \exp \left( -\frac{m_s}{R} \Delta s \right) [n\sigma - (T_1 + 2T_2)K \Delta \gamma] (\sigma C^{-1})\]

(55)

where

\[K = \frac{\varepsilon_\sigma}{\varepsilon_s}\]

is the isotropic hardening modulus.

We then update the viscoplastic strain, consistency parameter, stress, damage parameter and entropy production and effective equivalent plastic strain at each iteration and iterate until the convergence tolerances are achieved. An important advantage of this algorithm lies in the fact that it can be exactly linearized in closed form. This leads to the notion of consistent elastoplastic tangent moduli, which is derived for the composite as

\[C^* = E^*\]

\[\frac{E^* W_n \otimes n' E^*}{2\mu (1 - D)^2 m_s + (T_1 + 2T_2)(1 - D)K + n' : E^* W : n - \frac{D_{\alpha m_s}}{(1 - D)^2 T \rho R} \exp \left( -\frac{m_s}{R} \Delta s \right) [n\sigma - (T_1 + 2T_2)K \Delta \gamma] \sigma W_n}\]
where
\[
\mathbf{n}^* = \mathbf{n} + \frac{D_\alpha m_s}{(1 - D)T^3 \rho R} \exp \left( -\frac{m_s}{R} \Delta s_{n+1} \right) \mathbf{n} \sigma - (T_1 + 2T_2)K\Delta \gamma_{n+1}\right)(\sigma : \mathbf{W})
\]
\[
E^s^{-1} = \frac{1}{1 - D} \left[ C^{-1} + \frac{\Delta \gamma_{n+1}}{1 - D} \mathbf{W} \right]^{-1}
\]
\[
W^{-1} = 1 - \frac{D_\alpha m_s \Delta \gamma_{n+1}}{(1 - D)T^3 \rho R} \exp \left( -\frac{m_s}{R} \Delta s_{n+1} \right) \left( \mathbf{n} + \frac{1}{1 - D} \mathbf{N} \sigma \right) \otimes \sigma
\]

7. Numerical simulations and experimental comparison

The proposed unified damage coupled micro-mechanical model is implemented into ABAQUS through user defined subroutine UMAT. The influences of interfacial bond and CTE mismatch (between the matrix and the filler particles) are studied under uniaxial loading conditions. The finite element simulation results are compared with the corresponding strain-controlled monotonic tensile tests on particulate composite prepared using lightly cross-linked PMMA filled with ATH particles. Details of this testing procedure is given in Basaran et al. (submitted for publication).

The geometry of the dog boned shape specimen prepared according to ASTM D 638-98 used in the uniaxial tests is shown in Fig. 4. Owing to the symmetry, only one half of the gauge length was used for finite element simulation.

The Ramberg–Osgood plastic model was used to model the monotonic tensile plastic response, which is reasonably accurate for tensile behavior:
\[
\bar{\sigma}_y = \bar{\sigma}_0 + K\varepsilon^n
\]

where the parameters are determined from the experimental data as
\[
\bar{\sigma}_0 = -0.1791T + 19.6, \quad T \leq 100 \degree C
\]
\[
K = -8.5881 \times 10^{-4}T^3 + 9.1312 \times 10^{-2}T^2 - 4.8155T + 424.9, \quad T \leq 100 \degree C
\]
\[
n = 4.098 \times 10^{-7}T^3 - 4.476 \times 10^{-3}T^2 + 2.33 \times 10^{-3}T + 0.3542, \quad T \leq 100 \degree C
\]

Viscosity relaxation time \(\tau\) and the coefficient of thermal expansion (CTE) of the matrix are determined from the test data. They are as follows:
\[
\tau = \begin{cases} 
1.12406 \times 10^{-6}T^3 - 1.67823 \times 10^{-4}T^2 + 7.91134 \times 10^{-3}T + 7.35 \times 10^{-3}, \quad T \leq 90 \degree C \\
-2.6348 \times 10^{-6}T^4 - 1.08452 \times 10^{-3}T^3 - 1.64629 \times 10^{-1}T^2 + 10.9673T - 271.11, \quad T \geq 90 \degree C
\end{cases}
\]
\[
\text{Cte} = \begin{cases} 
3.035 \times 10^{-7}T + 2.347 \times 10^{-5}, \quad T \leq 90 \degree C \\
1.0992 \times 10^{-6}T - 5.012 \times 10^{-5}, \quad T \geq 90 \degree C
\end{cases}
\]

The material parameters needed for the proposed unified damage coupled viscoplastic micro-mechanics model are listed as follows:

- the average specific mass of composite \(m_s = 85 \text{ g/mole};\)
- density of composite \(\rho = 1750 \text{ kg/m}^3;\)

![Fig. 4. Uniaxial tensile test specimen geometry (mm).](image-url)
gas constant $R = 8.3145 \text{ J/mole/K}$;
volume fraction of filler particle $\phi = 0.48$;
Poisson’s ratio of filler particle $\nu_p = 0.24$;
elastic modulus of filler particle $E_p = 70,000 \text{ MPa}$;
coefficient of thermal expansion of filler particle $Cte = 1.47 \times 10^{-6}$;
Poisson ratio of the matrix PMMA $\nu_m = 0.31$;
elastic modulus of the matrix PMMA $E_m = -23.36T + 4123.8 \text{ MPa}$.

Fig. 5. Comparison of stress–strain relationship among damage coupled plastic model Ramberg–Osgood plasticity model and experiment data at 24 °C.

Fig. 6. Comparison of stress–strain relationship among damage coupled plastic model Ramberg–Osgood plasticity model and experiment data at 75 °C.
For the interface region between the particle and the matrix, Young’s modulus and interface thickness are adjustable parameters according to the new proposed model. We assume that the Poisson’s ratio and CTE for the interface are the same as the matrix PMMA.

The stress–strain relationship obtained from the unified damage coupled plastic model is compared with the Ramberg–Osgood plastic model and also compared with experimental data as shown in Figs. 5 and 6 at 24 °C and 75 °C, respectively. For simulations, 75 °C is used as the highest temperature because this particular material is not designed for or expected to experience much higher temperatures in its service life. The simulation results are obtained based on the assumption that the thickness of interface region is 1% of the radius of a filler particle and the elastic modulus of interface region is 50% of that of the matrix PMMA. It is seen that the damage effects must be accounted to be able to simulate the material behavior properly.

The influence of CTE mismatch between the filler particle and matrix on the overall uniaxial behavior of the composite is shown in Figs. 7 and 8 at 24 °C and 75 °C, respectively. $T_c$ is the temperature that the residual

![Fig. 7. The effect of CTE mismatch between matrix and particle on the overall stress–strain relation of particulate composite at 24 °C.](image1)

![Fig. 8. The effect of CTE mismatch between matrix and particle on the overall stress–strain relation of particulate composite at 75 °C.](image2)
stresses are set in the microstructure due to the CTE mismatch during cooling the composite from its curing temperature. These simulation results are obtained based on the assumption that the thickness of interface is 1% of the radius of a particle and the elastic modulus of interphase is 50% of that of PMMA.

The influence of stiffness of the interphase around the filler particle on the overall uniaxial behavior of the composite is shown in Figs. 9 and 10 at 24 °C and 75 °C, respectively. These results are obtained by only changing Young’s modulus of the interphase as the percent of that of matrix and keeping other parameters fixed (\(T_c = 100 \, ^\circ C\) and the thickness of interphase is 1% of the radius of particle). As shown in Figs. 9 and 10, when the interphase between the filler and matrix becomes more flexible (smaller elastic modulus) and stress in the matrix is reduced, which is in agreement with traditional weighted average stress equations for composites. As the interface gets more flexible, there is less contribution from the filler to the overall modulus, because the filler is a much stronger material then the matrix.

The influence of thickness of the interphase around the particle on the overall uniaxial behavior of the composite is shown in Figs. 11 and 12 at 24 °C and 75 °C, respectively. These results are obtained by only changing
the thickness of the interphase as a percent of the radius of the particle and keeping all other parameters fixed ($T_c = 100 \, ^\circ C$ and the elastic modulus of interphase is about 50% of that of PMMA). The results indicate that, as the interphase between the matrix and the filler gets thicker, the interface becomes softer. A more flexible interface makes the composite more flexible (meaning smaller stress level for the same strain). Having a more flexible interface also makes the composite more ductile, similar to the ductile matrix.

8. Conclusions

A unified damage coupled viscoplastic micro-mechanics constitutive model is proposed for a particulate composite. The proposed model accounts for the CTE mismatch between the filler and the matrix. Moreover, the model accounts for the stiffness of the interface region between the filler particles and the matrix. The interface is modeled as a layer with thickness and elastic properties. Then, the model is unified with a thermo-
dynamics based damage mechanics formulation. Comparison with test data indicates that the model is stable and predicts the test data well.

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References


