The uniaxial tension of particulate composite materials with nonlinear interface debonding

H. Tan a, Y. Huang a,*, C. Liu b, G. Ravichandran c, H.M. Inglis d, P.H. Geubelle d

a Department of Mechanical and Industrial Engineering, University of Illinois at Urbana–Champaign, Urbana, IL 61801, USA
b Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
c Graduate Aeronautical Laboratory, California Institute of Technology, Pasadena, CA 91125, USA
d Department of Aerospace Engineering, University of Illinois at Urbana-Champaign, Urbana, IL 61801, USA

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Abstract
Debonding of particle/matrix interfaces can significantly affect the macroscopic behavior of composite material. We have used a nonlinear cohesive law for particle/matrix interfaces to study interface debonding and its effect on particulate composite materials subject to uniaxial tension. The dilute solution shows that, at a fixed particle volume fraction, small particles lead to hardening behavior of the composite while large particles yield softening behavior. Interface debonding of large particles is unstable since the interface opening (and sliding) displacement(s) may have a sudden jump as the applied strain increases, which is called the catastrophic debonding. A simple estimate is given for the critical particle radius that separates the hardening and softening behavior of the composite.

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1. Introduction
Solid propellants and high explosives can be considered as composite materials with energetic particles in polymeric binder matrix (e.g., Bennett et al., 1998; Kimura and Oyumi, 1998; Ide et al., 1999; Liu, 2003; Balzer et al., 2004). These energetic materials display strong particle size effects. For example, large particles debond earlier than small ones in high explosives (Rae et al., 2002a). A mix of large and small particles gives much higher explosiveness than small particles only at a fixed volume fraction of energetic particles (Kimura and Oyumi, 1998; Fleming et al., 1985). Particle size strongly influences the behavior of high explosives, but the classical composite theories cannot predict the size effect since the theories involve no intrinsic material lengths (e.g., Budiansky, 1965; Hill, 1965; Mori and Tanaka, 1973; Christensen and Lo, 1979; Christensen, 1990; Huang et al., 1994a,b, 1995; Huang and Hu, 1995).
Debonding of particle/matrix interfaces can significantly affect the macroscopic behavior of solid propellants and high explosives. Hotspot may form from the localized sudden interface debonding, thus trigger detonation of high explosives under low-level loading. Interface debonding in plastic bonded explosives has been numerically studied (e.g., Bardenhagen et al., 1998, 2000) and experimentally observed (e.g., Liu, 2004). Debonded interfaces, which always start from large particles, have openings on the order of 100 μm that are comparable to, or even larger than, the average particle size (Liu, 2004). Debonded interfaces also become the path of crack propagation in high explosives (e.g., Wiegand and Pinto, 1996; Rae et al., 2002a,b) and solid propellants (e.g., Ho and Fong, 1987; Sciammarella and Sciammarella, 1998; Ide et al., 1999).

Debonding of particle/matrix interfaces can usually be characterized by a nonlinear cohesive law (Needleman, 1987), which gives stress tractions in terms of displacement discontinuities across the interface. Tan et al. (2005b) combined experiments and micromechanics models to determine the cohesive law for particle/matrix interfaces in the high explosive PBX 9501. The cohesive law displays three stages, namely the linear hardening (stage I), softening (stage II), and complete debonding (stage III), as illustrated in Fig. 1. The cohesive properties, including the linear hardening modulus, cohesive strength and cohesive energy, have been obtained for PBX 9501. Using such a cohesive law, Tan et al. (2005a, 2006) studied the effect of nonlinear interface debonding on macroscopic constitutive behavior of the high explosive PBX 9501. Specifically, small particles lead to hardening behavior of PBX 9501 while large particles yield softening behavior. Large particles may also lead to catastrophic debonding (i.e., sudden debonding even under static load) that may trigger the reaction or detonation of high explosives. Tan et al. (2005a,b), however, only studied nonlinear interface debonding in composites subject to hydrostatic tension, for which the interface shear becomes insignificant and the analytic solution can be obtained.

The uniaxial tensile test is an important experimental method to measure the mechanical properties of materials. We study in this paper the effect of nonlinear interface debonding on the macroscopic behavior of composites subject uniaxial tension. The effect of interface shear is important and must be accounted for. The paper is outlined as follows. A micromechanics model accounting for interface debonding is established in Section 2 for a composite subject to general loadings, and is applied to uniaxial tension in Section 3. Section 4 gives the dilute solution, and a specific cohesive law for the high explosive PBX 9501 is adopted in Section 5. The numerical results in Section 6 clearly show strong particle size effect on the macroscopic behavior of the composite.

2. A micromechanics model accounting for particle/matrix interface debonding

A micromechanics model is presented in this section to account for the effect of nonlinear interface debonding on the constitutive behavior of the composite. It is applied to study uniaxial tension in Section 3.

2.1. Strain energy

The macroscopic stress $\sigma$ and strain $\varepsilon$ of the composite are distinguished from the microscopic stress $\tilde{\sigma}$ and strain $\tilde{\varepsilon}$ in each constituent (particles and matrix). The stress and strain on the microscale are nonuniform due
to material inhomogeneities, and they satisfy the constitutive law for the corresponding constituent. On the contrary, the macroscopic stress and strain represent the collective, homogenized behavior of the composite, and are uniform in the representative volume element.

For a composite consisting of particles with volume fraction $f$ embedded in the matrix, the macroscopic stress–strain relation can be expressed as (e.g., Benveniste and Aboudi, 1984; Tan et al., 2005a, 2006)

$$
\bar{\varepsilon} = M^m : \bar{\sigma} + f \{(M^p - M^m) : \sigma^p + \varepsilon^{\text{int}}\},
$$

where the matrix and particles are linear elastic with the elastic compliance tensor $M^m$ and $M^p$, respectively; $\sigma^p = \frac{1}{V_p} \int_{\Omega_p} \sigma \, dV$ is the average stress in particles, $\Omega$ is the composite volume and $\Omega^p = \Omega \Omega_p$ is the particle volume; $\varepsilon^{\text{int}}$ is the additional strain due to interface debonding, and is related to the displacement discontinuity $[u] = \bar{u}^m - \bar{u}^p$ across the particle/matrix interface $S^{\text{int}}$ by

$$
\varepsilon^{\text{int}} = \frac{1}{2 \Omega^p} \int_{\Omega^p} ([u] \otimes n + n \otimes [u]) \, dA.
$$

Here $n$ is the unit normal vector on the interface pointing into the matrix.

Interface debonding can be characterized by a cohesive law, which gives the cohesive energy (per unit interface area) in terms of interface displacement discontinuity $[u]$,

$$
\phi = \phi([u]).
$$

Section 2.2 gives $\phi$ for the high explosive PBX 9501 determined by Tan et al. (2005b). Ferrante et al. (1982) gave $\phi$ for bimetallic interfaces.

The composite strain energy $U$ is composed of strain energy in the matrix and particles, and the cohesive energy in particle/matrix interfaces

$$
U = \frac{1}{2} \int_{\Omega} \sigma : \varepsilon \, dV + \int_{\Omega^p} \phi \, dA.
$$

Using the divergence theorem, we rewrite the above equation as

$$
U = \frac{1}{2} \bar{\sigma} : \bar{\varepsilon} + \int_{\Omega^p} \left\{ \phi - \frac{1}{4} \sigma^{\text{int}} : ([u] \otimes n + n \otimes [u]) \right\} \, dA,
$$

where $\sigma^{\text{int}}$ is the stress at particle/matrix interfaces.

### 2.2. A cohesive law for particle/matrix interfaces in the high explosive PBX 9501

Tan et al. (2005b) measured the cohesive law for particle/matrix interfaces in the high explosive PBX 9501. As shown in Fig. 1, the cohesive law is well characterized by three stages, namely the linear hardening, softening, and complete debonding stages. Each stage gives approximately a straight line. The cohesive law involves three parameters, namely the interface cohesive strength $\sigma_{\text{max}}$, linear modulus $k$, and softening modulus $\tilde{k}$ of the interface. For the high explosive PBX 9501, $\sigma_{\text{max}} = 1.66$ MPa, $k = 1.55$ GPa/μm and $\tilde{k} = 17$ MPa/mm. The normal stress at the interface $\sigma^{\text{int}}$ is then related to the opening displacement $[u]$ by

$$
\sigma^{\text{int}} = k[u], \quad [u] < \sigma_{\text{max}} / k, \quad \text{stage I},
$$

$$
\sigma^{\text{int}} = (1 + \tilde{k}/k) \sigma_{\text{max}} - \tilde{k}[u], \quad \sigma_{\text{max}} / k < [u] < \sigma_{\text{max}} (1/k + 1/\tilde{k}), \quad \text{stage II},
$$

$$
\sigma^{\text{int}} = 0, \quad [u] > \sigma_{\text{max}} (1/k + 1/\tilde{k}), \quad \text{stage III}.
$$

Its integration gives cohesive energy

$$
\phi = k[u]^2 / 2, \quad [u] < \sigma_{\text{max}} / k, \quad \text{stage I},
$$

$$
\phi = k[u]^2 / 2 - (k + \tilde{k})([u] - \sigma_{\text{max}} / k)^2 / 2, \quad \sigma_{\text{max}} / k < [u] < \sigma_{\text{max}} (1/k + 1/\tilde{k}), \quad \text{stage II},
$$

$$
\phi = \sigma_{\text{max}}^2 (1/k + 1/\tilde{k}) / 2, \quad [u] > \sigma_{\text{max}} (1/k + 1/\tilde{k}), \quad \text{stage III}.
$$
It is important to point out that the cohesive law in Eqs. (6) and (7) is limited to the opening displacement only. It is extended to account for the sliding displacement in Section 5.

3. Uniaxial tension

We consider a composite material with spherical particles subject to uniaxial tension. The particles radius is $a$, and volume fraction is $f$. For uniaxial tension $\sigma = \sigma_{33}$ along the $x_3$ direction, the axial strain $\varepsilon = \varepsilon_{33}$ is obtained from Eq. (1) as

$$\varepsilon = \frac{\sigma}{E_m} + f \left[ \left( \frac{1}{2\mu_p} \right) \sigma_{33} + \left( \frac{1}{E_p} - \frac{1}{2\mu_p} \right) \sigma_{kk} \right],$$

(8)

where $E_m$ and $v_m$ are Young’s moduli (shear moduli and Poisson’s ratios) of the matrix and $E_p$ and $v_p$ are Young’s moduli (shear moduli and Poisson’s ratios) of the particles, respectively. Once a micromechanics model of homogenization is adopted (e.g., dilute solution, Mori–Tanaka method), the average stress $\sigma_p$ in particles and additional strain $\varepsilon_{\text{int}}$ due to interface debonding are related to the macroscopic strain $\varepsilon$ (and stress $\sigma$). Eq. (8) then gives the macroscopic stress–strain relation for a composite subject to uniaxial tension.

3.1. A general solution accounting for interface debonding

We present a general approach to determine the average stress $\sigma_p$ and additional strain $\varepsilon_{\text{int}}$ due to interface debonding in uniaxial tension. For each micromechanics model of homogenization (e.g., dilute solution, Mori–Tanaka method), $\sigma_p$ and $\varepsilon_{\text{int}}$ are obtained from the deformation field of a single inclusion in an infinite medium subject to uniaxial tension (along the $x_3$ direction). The corresponding deformation field is axisymmetric.

The general solution for an axisymmetric deformation field is expressed in terms of the Legendre polynomials $P_n(\cos \theta)$ in spherical coordinates ($r$, $\theta$, $\phi$) (Luré, 1964). Specifically, the interface opening displacement $[u]$ and sliding displacement $[v]$ can be expressed as

$$[u] = \sum_{n=0}^{\infty} [u_n] P_n(\cos \theta),$$

$$[v] = \sum_{n=2}^{\infty} [v_n] P'_n(\cos \theta),$$

(9)

where the summation, here and thereafter, is for even numbers only, $P'_n(\cos \theta) = \frac{\partial P_n(\cos \theta)}{\partial \theta}$, and the coefficients $[u_n]$ and $[v_n]$ are to be determined via energy minimization in Section 6. Similarly, the normal and shear stress tractions at the interface, $\sigma_{\text{int}}$ and $\tau_{\text{int}}$, can be expressed as

$$\sigma_{\text{int}} = \sum_{n=0}^{\infty} \sigma_{\text{int}}^n P_n(\cos \theta),$$

$$\tau_{\text{int}} = \sum_{n=2}^{\infty} \tau_{\text{int}}^n P'_n(\cos \theta),$$

(10)

where the coefficients $\sigma_{\text{int}}^n$ and $\tau_{\text{int}}^n$ are to be determined.

The average stresses in spherical particles are related to $\sigma_{\text{int}}^n$ and $\tau_{\text{int}}^n$ by

$$\sigma_{33}^p = 3\sigma_{0}^\text{int} \quad \text{and} \quad \sigma_{kk}^p = \sigma_{0}^\text{int} + \frac{2}{3}(\sigma_{2}^\text{int} + 3\tau_{2}^\text{int}).$$

(11)

Here $\sigma_{kk}^p$ is obtained by averaging the hydrostatic stress $\sigma_{kk}$ in the particle over the spherical angle $\theta$ and it is directly proportional to $\sigma_{0}^\text{int}$. The additional stress $\sigma_{33}^p$ in the particle has the hydrostatic part (proportional to $\sigma_{0}^\text{int}$) and deviatoric part (proportional to $\sigma_{2}^\text{int}$ and $\tau_{2}^\text{int}$). The additional strain due to interface debonding is given in terms of $[u_n]$ and $[v_n]$ by
\[
\varepsilon_{33}^{\text{int}} = \frac{[u_0]}{a} + \frac{2([u_2] + [v_2])}{5a}.
\]

Here the additional strain \(\varepsilon_{33}^{\text{int}}\) due to interfacial debonding also has the volumetric part (proportional to \([u_0]\)) and deviatoric part (proportional to \([u_2]\) and \([v_2]\)). Eq. (8) then becomes

\[
\varepsilon = \frac{\bar{\sigma}}{E_m} + f \left\{ \left( \frac{1}{3K_p} - \frac{1}{3K_m} \right) \sigma_0^{\text{int}} + \left( \frac{1}{\mu_p} - \frac{1}{\mu_m} \right) \frac{\sigma_0^{\text{int}} + 3\tau_0^{\text{int}}}{5} + \frac{[u_0]}{a} + \frac{2([u_2] + [v_2])}{5a} \right\},
\]

where \(K_m\) and \(K_p\) are the bulk moduli of the matrix and particles, respectively. The above relation holds for all micromechanics models (e.g., dilute solution, Mori–Tanaka method) and all particle/matrix interface cohesive laws. However, \(\sigma_n^{\text{int}}, \tau_n^{\text{int}}, [u_n]\) and \([v_n]\) depend on not only the macroscopic strain \(\varepsilon\) (or stress \(\bar{\sigma}\)) but also the micromechanics model and interface cohesive law, as shown in Section 4.

3.2. Strain energy in uniaxial tension

For uniaxial tension, Eq. (5) gives the strain energy density as

\[
\frac{U}{\Omega} = \frac{1}{2} \bar{\sigma}^2 + \frac{3f}{2a} \int_0^\varepsilon \left\{ \phi - \frac{1}{2} \left( \sigma^{\text{int}}[u] + \tau^{\text{int}}[v] \right) \right\} \sin \theta \, d\theta,
\]

where \(a\) is the particle radius. For \([u]\) and \([v]\) in Eq. (9) and \(\sigma^{\text{int}}\) and \(\tau^{\text{int}}\) in Eq. (10), the strain energy density above becomes

\[
\frac{U}{\Omega} = \frac{1}{2} \bar{\sigma}^2 + \frac{3f}{2a} \int_0^{\pi/2} \phi \sin \theta \, d\theta - \sum_{n=0}^{\infty} \frac{\sigma_n^{\text{int}} [u_n] + n(n+1) \tau_n^{\text{int}} [v_n]}{2(2n+1)}.
\]

The above relations also hold for all micromechanics models and particle/matrix interface cohesive laws.

4. Dilute solution

We use the simplest micromechanics model of homogenization, namely the dilute solution that neglects the interaction among particles, to determine the coefficients \(\sigma_n^{\text{int}}\) and \(\tau_n^{\text{int}}\) in terms of \([u_n]\) and \([v_n]\). The coefficients \([u_n]\) and \([v_n]\) are then determined via energy minimization in Section 6.

An infinite matrix containing a single spherical particle of radius \(a\) is subject to remote uniaxial tension \(\bar{\sigma}\) in the dilute solution. The normal and shear stress tractions at the particle/matrix interface \(r = a\) are given in Eq. (10) as \(\sigma^{\text{int}} = \sum_{n=0}^{\infty} \sigma_n^{\text{int}} P_n(\cos \theta)\) and \(\tau^{\text{int}} = \sum_{n=1}^{\infty} \tau_n^{\text{int}} P_n(\cos \theta)\). For the spherical particle subject to the above normal and shear stress tractions, the displacement field \(u_\theta(r, \theta)\) and \(u_\theta(r, \theta)\) in the particle has been obtained analytically in terms of \(\sigma_n^{\text{int}}\) and \(\tau_n^{\text{int}}\) (Luré, 1964). The matrix is subject to remote uniaxial tension \(\bar{\sigma}\), and the above normal and shear stress tractions on its inner surface \(r = a\) (i.e., particle/matrix interface). The displacement field \(u_\theta^m(r, \theta)\) and \(u_\theta^m(r, \theta)\) in the matrix has also been obtained analytically in terms of \(\sigma_n^{\text{int}}\) and \(\tau_n^{\text{int}}\) (Luré, 1964). The displacement jump across the particle/matrix interface requires

\[
\begin{align*}
\left. u_\theta^p \right|_{(a+0, \theta)} - \left. u_\theta^p \right|_{(a-0, \theta)} &= [u] = \sum_{n=0}^{\infty} [u_n] P_n(\cos \theta), \\
\left. u_\theta^m \right|_{(a+0, \theta)} - \left. u_\theta^m \right|_{(a-0, \theta)} &= [v] = \sum_{n=2}^{\infty} [v_n] P_n'(\cos \theta),
\end{align*}
\]

which gives the following two linear algebraic equations to solve \(\sigma_n^{\text{int}}\) and \(\tau_n^{\text{int}}\) in terms of \([u_n]\) and \([v_n]\)

\[
\begin{align*}
M_n^u \sigma_n^{\text{int}} + n(n+1)M_n^\tau^{\text{int}} &= \frac{1 - v_m}{4(1 + v_m)\mu_m} \bar{\sigma}_\delta_{\alpha\theta} + \frac{5(1 - v_m)}{(7 - 5v_m)\mu_m} \bar{\sigma}_\delta_{\theta\theta} - \frac{[u_n]}{a}, \\
M_n^\tau \sigma_n^{\text{int}} + (M_n^u + M_n^\tau) \tau_n^{\text{int}} &= \frac{5(1 - v_m)}{2(7 - 5v_m)\mu_m} \bar{\sigma}_\delta_{\theta\theta} - \frac{[v_n]}{a},
\end{align*}
\]
where \( \delta \) is a Kronecker delta, and
\[
M_u' = \frac{2n^2 - 1 - (2n^2 - n - 2)v_p}{2(n - 1)(n^2 + n + 1 + (2n + 1)v_p)\mu_p} + \frac{2n^2 + 4n + 1 - (2n^2 + 5n + 1)v_m}{2(n + 2)(n^2 + n + 1 - (2n + 1)v_m)\mu_m},
\]
\[
M_v' = \frac{2 - n + (2n - 1)v_p}{2(n - 1)(n^2 + n + 1 + (2n + 1)v_p)\mu_p} + \frac{n + 3 - (2n + 3)v_m}{2(n + 2)(n^2 + n + 1 - (2n + 1)v_m)\mu_m}.
\]

Only \([u_n]\) and \([v_n]\) remain to be determined via energy minimization in Section 6.

Using Eq. (17), we can simplify the stress–strain relation of the composite in Eq. (13) to
\[
\sigma = E_0 \left( \frac{1 - K_m/K_0 [u_0]}{1 - K_m/K_0} - \frac{1 - \mu_m/\mu_0}{1 - \mu_m/\mu_p} \frac{2[u_2] + 6[v_2]}{5a} \right),
\]
where \(E_0, K_0\) and \(\mu_0\) are respectively the dilute solution of the Young’s, bulk and shear moduli of a composite without interface debonding \(([u] = [v] = 0)\), and are given in the Appendix. The strain energy density in Eq. (15) is then written in terms of \([u_n]\) and \([v_n]\) as
\[
\frac{U}{\Omega} = \frac{\sigma^2}{2E_0} + \frac{3f}{2a^2} \sum_{n=0}^{\infty} \left( \frac{M_u' + M_v'}{M_u' + M_v'}\right)[u_n]^2 - 2n(n + 1)M_u'[u_n][v_n] + n(n + 1)M_v'[v_n]^2 + \frac{3f}{a} \int_0^{\pi/2} \phi \sin \theta \, d\theta.
\]
The cohesive energy \(\phi\) is expressed in terms of \([u_n]\) and \([v_n]\) in the next section.

### 5. A cohesive law for particle/matrix interfaces

The cohesive law in Section 2.2, which involves only the interface opening displacement \([u]\), is extended in this section to account for the sliding displacement \([v]\) via the following combined measure (e.g., Tvergaard and Hutchinson, 1993)
\[
\lambda = \sqrt{ \left( \frac{[u]}{\delta_{\text{open}}} \right)^2 + \left( \frac{[v]}{\delta_{\text{slide}}} \right)^2 },
\]
where \(\delta_{\text{open}}\) and \(\delta_{\text{slide}}\) are the critical opening and sliding displacements of the interface, respectively, and \(\lambda = 1\) corresponds to complete interface debonding.

The interface cohesive energy \(\phi\) depends only on \(\lambda\) (e.g., Tvergaard and Hutchinson, 1993),
\[
\phi = \phi(\lambda).
\]

For the interface cohesive law in Eq. (7), \(\phi\) takes the form
\[
\phi = \frac{1}{2} \delta_{\text{open}}^2 k \lambda^2, \quad \lambda < \frac{k}{k + k}, \quad \text{stage I},
\]
\[
\phi = \frac{1}{2} \delta_{\text{open}}^2 \left[ k \lambda^2 - (k + k) \left( \lambda - \frac{k}{k + k} \right)^2 \right], \quad \frac{k}{k + k} < \lambda < 1, \quad \text{stage II},
\]
\[
\phi = \frac{1}{2} \delta_{\text{open}}^2 \frac{k k}{k + k}, \quad \lambda > 1, \quad \text{stage III},
\]
where \(\delta_{\text{open}} = \sigma_{\text{max}} \left( \frac{1 + \frac{1}{\epsilon}}{\frac{1}{\epsilon}} \right)\).

The normal and shear stresses at the particle/matrix interface are given by
\[
\sigma^\text{int} = \frac{\partial \phi}{\partial [u]} = \frac{\phi'(\lambda)}{\delta_{\text{open}}^2 \lambda} [u],
\]
\[
\tau^\text{int} = \frac{\partial \phi}{\partial [v]} = \frac{\phi'(\lambda)}{\delta_{\text{open}}^2 \lambda} \left( \frac{\delta_{\text{open}}}{\delta_{\text{slide}}} \right)^2 [v],
\]
\]
where
\[
\frac{\phi'}{\delta_{\text{open}}^2 \lambda} = k, \quad \lambda < \frac{\tilde{k}}{k + \tilde{k}}, \quad \text{stage I},
\]
\[
\frac{\phi'}{\delta_{\text{open}}^2 \lambda} = \tilde{k} \frac{1 - \lambda}{\lambda}, \quad \frac{\tilde{k}}{k + \tilde{k}} < \lambda < 1, \quad \text{stage II},
\]
\[
\frac{\phi'}{\delta_{\text{open}}^2 \lambda} = 0, \quad \lambda > 1, \quad \text{stage III}.
\]

Fig. 2. Legendre coefficients versus the macroscopic strain $\bar{\varepsilon}$. (a) coefficients $[u_n]$ for interface opening displacement. (b) coefficients $[v_n]$ for interface sliding displacement.
6. Results

The strain energy in Eq. (20) is now a function of the macroscopic strain $\varepsilon$, $[u_n]$ and $[v_n]$ after the macroscopic stress $\varepsilon$ is substituted by Eq. (19). For a given macroscopic strain $\varepsilon$, the strain energy is the same as the potential energy such that the principle of minimum potential energy gives the following equations to determine the coefficients

$$\frac{\partial (U/\Omega)}{\partial [u_n]} = 0,$$

$$\frac{\partial (U/\Omega)}{\partial [v_n]} = 0.$$  \hspace{1cm} (26)

For simplicity we take the critical interface sliding displacement to be the same as the opening displacement $\delta_{\text{slide}} = \delta_{\text{open}}$ in the following.

6.1. Linear stage of interface debonding

The particle/matrix interface is in the linear stage (stage I in Fig. 1) for small macroscopic strain $\varepsilon$. The cohesive energy can be written as $\phi = \frac{k}{2} ([u]^2 + [v]^2)$, which gives

$$\int_0^{\pi/2} \phi \sin \theta \, d\theta = \frac{k}{2} \sum_{n=0}^{\infty} \frac{1}{2n+1} \left\{ [u_n]^2 + n(n+1)[v_n]^2 \right\}.$$  \hspace{1cm} (27)

The minimization of energy in Eq. (20) gives vanishing $[u_n]$ and $[v_n]$ except

$$[u_0] = \frac{\frac{1}{3K_m} + \frac{1}{4\mu_m}}{1 + ka \left( \frac{1}{3K_p} + \frac{1}{4\mu_m} \right)} \frac{a\bar{\sigma}}{3}, \quad [u_2] = 2[v_2] = \frac{\frac{5(1 - v_m)}{(7 - 5v_m)\mu_m}}{1 + ka \left( \frac{1}{2\mu_p} + \frac{4 - 5v_m}{(7 - 5v_m)\mu_m} \right)} \frac{a\bar{\sigma}}{3}.$$  \hspace{1cm} (28)

The linear relation between the macroscopic stress and strain in this stage can be written as

$$\sigma = E\varepsilon.$$  \hspace{1cm} (29)

![Fig. 3. The stress–strain curve for a composite containing small particles (radius $a = 4$ microns).](image-url)
where $\bar{E}$ is the composite Young’s modulus accounting for linear interface debonding, and is given by

$$\frac{1}{\bar{E}} = \frac{1}{E_0} + \frac{1}{3} \frac{1 - K_m/K_0}{1 - K_m/K_p} \left[ \frac{1}{3K_m} + \frac{1}{4\mu_m} \right] + \frac{1 - \mu_m/\mu_0}{1 + ka \left( \frac{1}{3K_p} + \frac{1}{4\mu_m} \right)} \left( \frac{5(1 - \nu_m)}{(7 - 5\nu_m)\mu_m} \right).$$

(30)

6.2. Softening stage of interface debonding

Once the macroscopic strain $\bar{\gamma}$ exceeds a critical value, the top and bottom points of the interface ($\theta = 0$ and $\pi$) reach the softening stage (stage II in Fig. 1) first. This critical macroscopic strain is determined from $[u_0] + [u_2] = \frac{\eta_{\text{max}}}{k}$ as

![Angular distribution of the opening and sliding displacements at interfaces between the matrix and small particles (radius $a = 4 \mu$m).](image)

Fig. 4. Angular distribution of the opening and sliding displacements at interfaces between the matrix and small particles (radius $a = 4 \mu$m). (a) Opening [$u$] and (b) sliding [$v$].
The corresponding macroscopic stress is obtained from the Young’s modulus in Eq. (29) as \( \bar{\sigma}^{I-II} = E^{I-II} \). For \( \bar{\varepsilon} > \bar{\varepsilon}^{I-II} \), the analysis becomes nonlinear due to softening in interface debonding. We use the conjugate gradient method to determine \([u_n]\) and \([v_n]\) by minimizing the energy in Eq. (20).

The material properties are taken from the high explosive PBX 9501. The elastic bulk and shear moduli of particles are \( K_p = 12.5 \) GPa and \( \mu_p = 5.43 \) GPa (Zaug, 1998). The matrix Young’s modulus is \( E_m = 1 \) MPa, and Poisson’s ratio \( \nu_m = 0.499 \) (Cady et al., 2000; Mas et al., 2001). The parameters in the interface cohesive law for the high explosive PBX 9501 are \( \sigma_{\text{max}} = 1.66 \) MPa, \( k = 1.55 \) GPa/\( \mu m \) and \( \tilde{k} = 17 \) MPa/\( \mu m \) (Tan et al., 2005b), which give the critical opening displacement \( \delta_{\text{open}} = 98 \mu m \). The particle volume fraction is taken as \( f = 10\% \). In the following we focus on the particle size effect since small particles lead to very different behavior from large particles at the same particle volume fraction.

6.2.1. Small particles

We take small particle radius \( a = 4 \mu m \). The critical macroscopic strain for the interface to reach softening is \( \bar{\varepsilon}^{I-II} \approx 0.62 \), and the corresponding macroscopic stress is \( \bar{\sigma}^{I-II} \approx 0.50\sigma_{\text{max}} \). Fig. 2 shows the coefficients \([u_n]\) and \([v_n]\) versus the normalized macroscopic strain \( \bar{\varepsilon}/\bar{\varepsilon}^{I-II} \). The coefficients \([u_{20}], [u_{22}], ..., [v_{20}], [v_{22}], ... \), are very close to zero, which suggests that the numerical solution has converged.

Fig. 3 shows the normalized macroscopic stress \( \bar{\sigma}/\bar{\sigma}^{I-II} \) versus macroscopic strain \( \bar{\varepsilon}/\bar{\varepsilon}^{I-II} \) for \( a = 4 \mu m \). It is a straight line before the interface reaches the softening stage. Nonlinearity results from the softening in interface debonding. The points marked by A, B, C, D and E correspond to strains \( \bar{\varepsilon}^{I-II}, 1.25\bar{\varepsilon}^{I-II}, 1.5\bar{\varepsilon}^{I-II}, 1.75\bar{\varepsilon}^{I-II} \) and \( 2\bar{\varepsilon}^{I-II} \), respectively. Fig. 4 shows the distribution of interface opening displacement \([u]\) and sliding displacement \([v]\) (versus the angle \( \theta \)) for the five points A, B, C, D and E in Fig. 3. There is no visible opening (sliding) displacement(s) for point A since the interface just reaches the softening stage. For points B, C, D and E, the opening (sliding) displacement(s) increases monotonically with the

Fig. 5. The stress–strain curve for a composite containing large particles (radius \( a = 125 \mu m \).
macroscopic strain $\bar{e}$. The debonding region also increases with $\bar{e}$, but the interface is still not completely debonded at $\bar{e} = 2\bar{e}_{I-II}$.

6.2.2. Large particles

We take large particle radius $a = 125 \, \mu\text{m}$. The critical macroscopic strain (and stress) for the interface to reach softening is the same as that in Section 6.2.1 for small particles, $\bar{e}_{I-II} \approx 0.62$ ($\bar{\sigma}_{I-II} \approx 0.50\sigma_{\text{max}}$), which suggests that the particle size has essentially no effect before the interface reaches the softening stage. For large particles, more terms are needed in Eq. (9) since only coefficients of higher-order terms $[u_{28}], [u_{30}], \ldots$, and $[v_{28}], [v_{30}], \ldots$, are close to zero.

Fig. 5 shows the normalized macroscopic stress $\bar{\sigma}/\bar{\sigma}_{I-II}$ versus macroscopic strain $\bar{e}/\bar{e}_{I-II}$ for $a = 125 \, \mu\text{m}$. Once again it is a straight line before the interface reaches the softening stage. However, the stress–strain curve

![Diagram showing interface opening and sliding displacements](image)

Fig. 6. Angular distribution of the opening and sliding displacements at interfaces between the matrix and large particles (radius $a = 125 \, \mu\text{m}$). (a) Opening [$u$] and (b) sliding [$v$].
displays a sudden drop when the macroscopic strain \( \varepsilon \) reaches 1.10\( \varepsilon^{I-II} \), and the drop in macroscopic stress \( \sigma \) is 0.086\( \sigma^{I-II} \). This sudden drop is due to unstable debonding of the particle/matrix interface, which has been observed in composite subject to hydrostatic tension, and is called the "catastrophic debonding" (Tan et al., 2005a,b). The points marked by A, B, C and D correspond to strains 1.033\( \varepsilon^{I-II} \), 1.067\( \varepsilon^{I-II} \) and 1.10\( \varepsilon^{I-II} \) prior to catastrophic debonding, while the points marked by E, F, G and H correspond to strains 1.10\( \varepsilon^{I-II} \), 1.133\( \varepsilon^{I-II} \), 1.167\( \varepsilon^{I-II} \), and 1.2\( \varepsilon^{I-II} \) after catastrophic debonding, respectively. Fig. 6 shows the distribution of interface opening displacement \( u \) and sliding displacement \( v \) (versus the angle \( \theta \)) for these points in Fig. 5. For points A, B, C and D, the opening (sliding) displacement(s) increases gradually with the macroscopic strain \( \varepsilon \). For points D and E that correspond to the same macroscopic strain 1.10\( \varepsilon^{I-II} \), there is a big jump in the opening and sliding displacements, i.e., "catastrophic debonding". This jump leads to sudden drop in the stress–strain curve in Fig. 5. After catastrophic debonding the opening and sliding displacements increase gradually again, as seen from the curves for points E, F, G and H.

6.2.3. Particle size effect

Fig. 7 shows the macroscopic stress–strain curves for different particle sizes at the same particle volume fraction. The curve for \( a = 75 \mu m \) is (approximately) at the transition between hardening and softening for the composite subject to uniaxial tension. Tan et al. (2006) obtained the following analytical expression of critical particle radius separating the hardening and softening behavior of the composite subject to hydrostatic tension

\[
a^{cr} = \frac{1}{k \left( \frac{4v_m}{\alpha_1} + \frac{1}{\alpha_p} \right)}.
\]  

(32)

It gives \( a^{cr} = 78 \mu m \) for the present material properties, and is very close to the present result (\( a = 75 \mu m \)). Therefore, Eq. (32) may be used to estimate the critical particle radius separating the hardening and softening behavior under general loadings.

7. Concluding remarks and discussions

We have used a nonlinear cohesive law for particle/matrix interfaces to study the effect of interface debonding on the macroscopic behavior of particulate composite materials subject to uniaxial tension. The dilute
solution shows that, at a fixed particle volume fraction, small particles lead to hardening behavior of the composite while large particles yield softening behavior. Interface debonding of large particles is unstable since the interface opening (and sliding) displacement(s) may have a sudden jump as the applied strain increases, which is called the catastrophic debonding. A simple estimate is given for the critical particle radius that separates the hardening and softening behavior of the composite.

It should be pointed out that the opening and sliding displacements in Figs. 4 and 6 are comparable to the particle radius. Strictly speaking, the finite deformation analysis may be necessary since the infinitesimal deformation analysis may not be accurate any more.

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Appendix

The dilute solution gives the bulk and shear moduli of a composite without interface debonding as

\[
K_0 = K_m \left\{ 1 + \frac{f\left( \frac{K_p}{K_m} - 1 \right)}{1 + \left[ \frac{1 + v_m}{3(1 - v_m)} - f\left( \frac{K_p}{K_m} - 1 \right) \right]} \right\}
\]

and

\[
\mu_0 = \mu_m \left\{ 1 + \frac{f\left( \frac{\mu_p}{\mu_m} - 1 \right)}{1 + \left[ \frac{2(4 - 5v_m)}{15(1 - v_m)} - f\left( \frac{\mu_p}{\mu_m} - 1 \right) \right]} \right\}.
\]

The Young’s modulus is given by

\[
E_0 = \frac{3\mu_0 K_0}{\mu_0 + 2K_0}.
\]

References


