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A micromechanical model for effective elastic properties of particulate composites with imperfect interfacial bonds

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Abstract

A micromechanical model for effective elastic properties of particle filled acrylic composites with imperfect interfacial bonds is proposed. The constituents are treated as three distinct phases, consisting of agglomerate of particles, bulk matrix and interfacial transition zone around the agglomerate. The influence of the interfacial transition zone on the overall mechanical behavior of composites is studies analytically and experimentally. Test data on particle filled acrylic composites with three different interfacial properties are also presented. The comparison of analytical simulation with experimental data demonstrated the validity of the proposed micromechanical model with imperfect interface. Both the experimental results and analytical prediction show that interfacial conditions have great influence on the elastic properties of particle filled acrylic composites.

Keywords: Micromechanics; Particulate composite; Interfaces; Interfacial bond; Elastic modulus

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1. Introduction and Literature Survey

It is well known that interface imperfections can significantly affect the mechanical properties and failure mechanisms as well as the strength of the particulate composites. The nature of the bond between filler particles and the matrix material has a significant affect on the mechanical behavior of particulate composites. Most analytical and numerical models assume that the bond between the filler and matrix is perfect and can be modeled using the continuity of tractions and displacements across a discrete interface. When the perfect interface conditions are present, the primary result is the well-known Eshelby (1957) solution of the

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ellipsoidal inclusion problem. However, internal defects and imperfect interfaces are well known to exist in composites and the incorporation of such phenomena into the general theory requires modification and relaxation of the continuity of displacements between the constituents. The imperfect interface bond may be due to a very compliant thin interfacial layer known as interphase or interface damage. Such an interfacial zone may have also been introduced deliberately by coating the filler particles in order to control the mechanical response and fatigue life of the composite. It may also develop during the manufacturing process due to chemical reactions between the contacting particles and matrix material or due to interface damage from cycling thermo-mechanical loading. The strength of the bond at the particle-matrix interface controls the fatigue life of the composite significantly. By controlling the stress–strain response and ductility of the interphase region, it is possible to control overall behavior of the composite.

The influence of an interfacial zone on composite mechanical and thermal behavior has already been investigated by a number of researchers, such as Walpole (1978), Benveniste (1985), Ghahremani (1985), Mura et al. (1985), Achenbach and Zhu (1989, 1990), and Hashin (1990, 1991a), Basaran et al. (2004) and many others. There have also been several investigations in the literature that concentrate on the effect of imperfect interfaces in composites, which may be appropriate in the case of thin coatings on inclusions. In the related studies, two analytical models have been proposed. First is the spring layer model, which involves a very thin interfacial zone of unspecified thickness (Benveniste, 1985; Achenbach and Zhu, 1989, 1990 and Hashin (1990, 1991a)). In this model it is assumed that the radial and the tangential tractions are continuous across the interphase for reasons of equilibrium, but because of the presence of the interphase, the displacements may be discontinuous from the filler particle to the matrix. The model assumes that the tractions are proportional to the corresponding displacement discontinuities. The proportionality constants therefore characterize the stiffness and strength of the interphase.

According to the spring layer model, two different homogeneous media, matrix labeled as 1 and particle labeled as 2, are joined by an imperfect interface S_{12}, as shown in Fig. 1. With respect to a local orthogonal system, the following relations describe the imperfect interface (Hashin, 1991a)

$$T_n^{(1)} = T_n^{(2)} = D_n[u_n], \quad [u_n] = u_n^{(1)} - u_n^{(2)} \tag{1a}$$

$$T_s^{(1)} = T_s^{(2)} = D_s[u_s], \quad [u_s] = u_s^{(1)} - u_s^{(2)} \tag{1b}$$

$$T_t^{(1)} = T_t^{(2)} = D_t[u_t], \quad [u_t] = u_t^{(1)} - u_t^{(2)} \tag{1c}$$

where n is the normal direction and s, t are the tangent directions. D_n, D_s, and D_t are the interface spring stiffness constants. Infinite values of these parameters imply that there are no interface displacement jumps and the perfectly bonded interface conditions exist. At the other extreme, zero values of D_n, D_s, and D_t imply that the interface tractions do not exist and the filler is debonded from the adjoining matrix media.

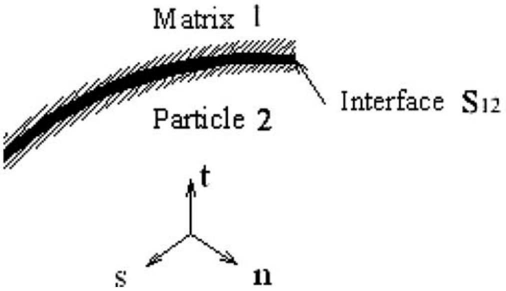

Fig. 1. Spring layer model (After Hashin, 1991a).

Finite positive values for the interface parameters define an imperfect interface, which lies between two extreme cases mentioned above.

A considerable amount of literature is available on composites with coated reinforcements, where such materials are viewed as three phase composites. The alternative approach of modeling the interface describes the interphase as a layer between particles and matrix of a specified thickness and of elastic constants different from those of the matrix and the particles (Benveniste, 1985; Achenbach and Zhu, 1989, 1990; Hashin, 1990). The literature shows that a thin flexible interphase layer is equivalent to the spring layer model for the imperfect interface when the interface parameters have been determined in terms of interphase properties and thickness (Hashin, 1991a). The effect of the coating, when it is thin, can be conveniently expressed in terms of the imperfect interface. It is indeed possible, as shown in Hashin (1990, 1991a), to express imperfect interface parameters simply and directly in terms of the interphase (coating) characteristics. One of our objectives in this paper is to show how to take into account the pronounced influence that even a thin coating may exert upon the overall elastic moduli of particulate composites.

2. Micromechanical model for elastic property determination

In many particulate composites, a thin layer of 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 assumption defines a three-phase composite that includes particles, thin interphase, and the matrix as shown in Fig. 2. Literature is very rich in constitutive models for particle filled composite materials with perfectly bonded interfaces. To be able to use the composite material constitutive models developed for perfectly bonded interfaces for composites with imperfect interface, the effective mechanical and thermal properties of the composite sphere assemblage (CSA), which consists of the particle and interphase layer of thickness δ, must be determined. Ju and Chen (1994a,b) micromechanical model for perfectly bonded interface particulate composites is well known as the most effective medium approach based on generalizations of the Eshelby method, Eshelby (1957). This model would be particularly useful for situations with random microstructures, which is the case for the particular composite we are interested in. In this paper, Ju and Chen (1994a,b) model is modified to incorporate the imperfect interfacial bonding conditions.

In the next section, we will find the theoretical solution for effective thermomechanical properties of the CSA consisting of an elastic spherical particle and an elastic interphase layer (with a thickness of δ), where 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 μ.

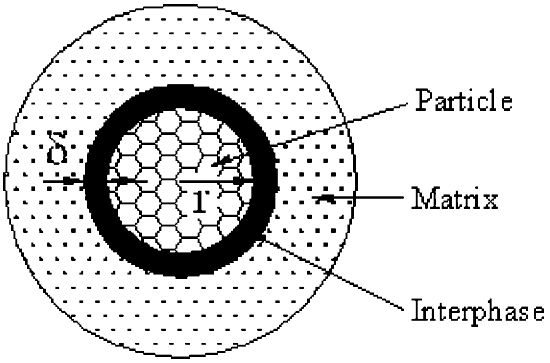

Fig. 2. Three-phase composite system.

3. Effective thermomechanical properties of CSA

Geometrical approximations are often made in obtaining effective thermomechanical properties of heterogeneous materials, because of the complications involved. One of the most common idealized geometric models is the composite sphere assemblage (CSA) model proposed by Kerner (1956) and Van der Poel (1958) as shown in Fig. 3. Smith (1974, 1975), Christensen and Lo (1979), Hashin and Rosen (1964) and Hashin (1962, 1963, 1968, 1990, 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. In the following formulae, k represents the bulk modulus, μ represents shear modulus, α represents coefficient of the 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^* for the CSA as obtained by Hashin (1962) is given by

$$k^* = k_\mathrm{i} + \frac{\phi}{\frac{1}{k_\mathrm{f}-k_\mathrm{i}} + \frac{3(1-\phi)}{3k_\mathrm{i}+4\mu_\mathrm{i}}} \tag{2}$$

where

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

where r is the radius of the filler particle and δ is the thickness of interphase.

Hashin and Shtrikman's formulae (1963) and Walpole's formulae (1966) provide upper and lower bounds for the bulk modulus of the CSA. Eq. (2) is same as the highest lower bound.

3.2. Effective coefficient of thermal expansion (ECTE)

The effective coefficient of thermal expansion α^* for the CSA is given by Levin (1967)

$$\alpha^* = \alpha_\mathrm{i} + \frac{\alpha_\mathrm{f}-\alpha_\mathrm{i}}{\frac{1}{k_\mathrm{f}} - \frac{1}{k_\mathrm{i}}}\left(\frac{1}{k^*} - \frac{1}{k_\mathrm{i}}\right) \tag{3}$$

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 as follows:

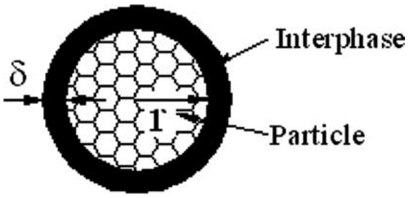

Fig. 3. Composite spherical assemblage (CSA) model.

$$A\left(\frac{\mu^*}{\mu_i}\right)^2 + B\left(\frac{\mu^*}{\mu_i}\right) + D = 0 \tag{4}$$

where

$$A = 8[\mu_f/\mu_i - 1](4 - 5v_i)\eta_1\phi^{10/3} - 2[63(\mu_f/\mu_i - 1)\eta_2 + 2\eta_1\eta_3]\phi^{7/3} + 252[\mu_f/\mu_i - 1]\eta_2\phi^{5/3} - 50[\mu_f/\mu_i - 1](7 - 12v_i + 8v_i^2)\eta_2\phi + 4(7 - 10v_i)\eta_2\eta_3 \tag{5}$$

$$B = -4[\mu_f/\mu_i - 1](1 - 5v_i)\eta_1\phi^{10/3} + 4[63(\mu_f/\mu_i - 1)\eta_2 + 2\eta_1\eta_3]\phi^{7/3} - 504[\mu_f/\mu_i - 1]\eta_2\phi^{5/3} + 150[\mu_f/\mu_i - 1](3 - v_i)v_i\eta_2\phi + 3(15v_i - 7)\eta_2\eta_3 \tag{6}$$

$$D = 4[\mu_f/\mu_i - 1](5v_i - 7)\eta_1\phi^{10/3} - 2[63(\mu_f/\mu_i - 1)\eta_2 + 2\eta_1\eta_3]\phi^{7/3} + 252[\mu_f/\mu_i - 1]\eta_2\phi^{5/3} + 25[\mu_f/\mu_i - 1](v_i^2 - 7)\eta_2\phi - (7 + 5v_i)\eta_2\eta_3 \tag{7}$$

with

$$\eta_1 = [\mu_f/\mu_i - 1](49 - 50v_f v_i) + 35(\mu_f/\mu_i)(v_f - 2v_i) + 35(2v_f - v_i) \tag{8}$$

$$\eta_2 = 5v_f[\mu_f/\mu_i - 8] + 7(\mu_f/\mu_i + 4) \tag{9}$$

$$\eta_3 = (\mu_f/\mu_i)(8 - 10v_i) + (7 - 5v_i) \tag{10}$$

where $\phi = \left(\frac{r}{r+\delta}\right)^3$.

Based on the Van der Poel's (1958) formula for the shear modulus of a particulate composite, Smith (1974, 1975) also proposed the condition for determining the effective shear modulus of the CSA as follows:

$$\alpha\left(\frac{\mu^*}{\mu_i} - 1\right)^2 + \beta\left(\frac{\mu^*}{\mu_i} - 1\right) + \gamma = 0 \tag{11}$$

where

$$\alpha = [4P(7 - 10v_i) - S\phi^{7/3}][Q - (8 - 10v_i)(M - 1)\phi] - 126P(M - 1)\phi(1 - \phi^{2/3})^2 \tag{12}$$

$$\beta = 35(1 - v_i)P[Q - (8 - 10v_i)(M - 1)\phi] - 15(1 - v_i)[4P(7 - 10v_i) - S\phi^{7/3}](M - 1)\phi \tag{13}$$

$$\gamma = -525P(1 - v_i)^2(M - 1)\phi \tag{14}$$

with

$$M = \mu_f/\mu_i \tag{15}$$

$$P = (7 + 5v_f)M + 4(7 - 10v_f) \tag{16}$$

$$Q = (8 - 10v_i)M + (7 - 5v_i) \tag{17}$$

$$S = 35(7 + 5v_f)M(1 - v_i) - P(7 + 5v_i) \tag{18}$$

where $\phi = \left(\frac{r}{r+\delta}\right)^3$

Eqs. (4) and (11) are considered (and verified) to be as equivalent in the determination of the effective shear modulus of the CSA. Using the above equations and the quadratic formula, one can determine the exact solution for the effective shear modulus of the CSA model. One of the roots is negative and is extraneous. The positive root provides the value of the effective shear modulus.

3.4. Effective Young's modulus and Poisson's ratio

Effective Young's modulus and Poisson's ratio for the CSA can be calculated from the well-known expressions

$$E^* = \frac{9k^*\mu^*}{3k^* + \mu^*} \tag{19}$$

$$v^* = \frac{3k^* - 2\mu^*}{6k^* + 2\mu^*} \tag{20}$$

3.5. Numerical examples

In order to illustrate the effects of imperfect interface and interphase thickness on the overall effective mechanical and thermal properties of CSA, we consider a special case of CSA that consists of spherical alumina trihydrate (ATH) filler particle and interphase material Dupont CA-14. The following two-phase properties are used:

$$E_{\mathrm{f}} = 70\ \mathrm{GPa}, \quad v_{\mathrm{f}} = 0.24, \quad \alpha_{\mathrm{f}} = 13 \times 10^{-6}/^{\circ}\mathrm{C}$$

$$v_{\mathrm{i}} = 0.31, \quad \alpha_{\mathrm{i}} = 70 \times 10^{-6}/^{\circ}\mathrm{C}, \quad \delta/r = 0.01 \quad \text{and} \quad \delta/r = 0.001$$

The non-dimensional interface parameter is defined as $q = E_{\mathrm{f}}/E_{\mathrm{i}}$. Zero value of q implies that there are no interface displacement jumps and the perfectly bonded interface conditions exist. At the other extreme, infinite value of q implies that the interface tractions do not exist and the filler is debonded from the adjoining matrix media. Finite positive values for q define an imperfect interface, which lies between two extreme cases mentioned above. We focus on the effects of degradation the interface bond from the initial perfect bonding to completely debonding and the thickness of the interphase on the overall mechanical and thermal properties of the CSA. A plot of effective thermal expansion coefficient as a function of q is shown in Fig. 4 for different interphase thicknesses. Thickness of interphase has a great influence on the overall effective CTE of CSA. It was also observed that weakening the bond decreases the effective CTE until it asymptotically attains the CTE value of ATH. The reason for this is that when the interphase doses not exist, the effective CTE of the material is exactly the CTE of the filler.

Figs. 5–7 show the effects of the interphase bond modulus and thickness on the effective shear modulus, effective Young's modulus, and effective bulk modulus. The stiffness of the bond has a strong effect on the degradation of the shear modulus, Young's modulus, and bulk modulus. As the interface becomes thinner, for the same E_{i} value, the effective shear modulus increases. On the other hand for the same interphase thickness, as the elastic modulus value E_{i} of the interphase decreases, the effective shear modulus decrease. This is also true for both the effective bulk modulus and Young's modulus.

Fig. 8 shows the effects of the interphase bond modulus and thickness on the effective Poisson's ratio. Effective Poisson's ratio seems to be independent of the interphase stiffness when $q \leqslant 10^2$ and $q \geqslant 10^4$.

We analytically and numerically evaluated the effective elastic properties and the coefficient of thermal expansion of the composite sphere assemblage. Simulation results show that the bulk and shear moduli are generally insensitive to the value of the Poisson's ratio of interphase. On the other hand, numerical results

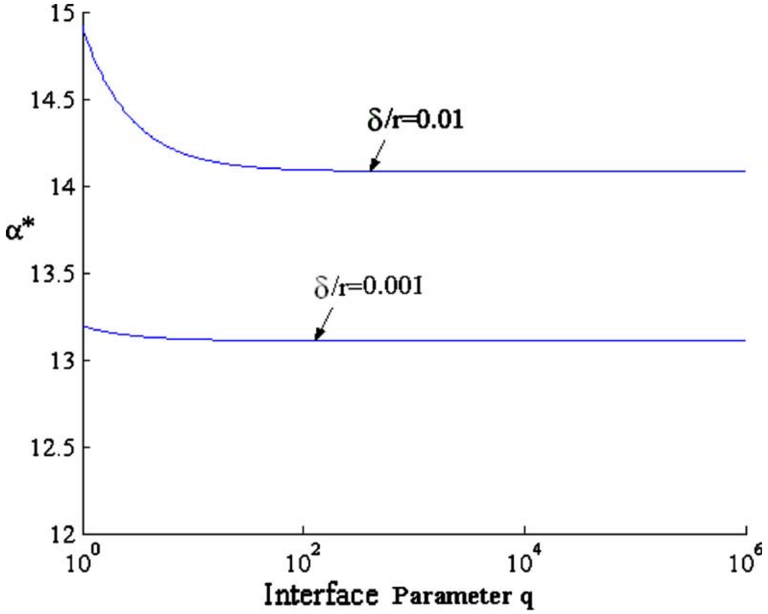

Fig. 4. Variation of effective CTE with the interface parameter q.

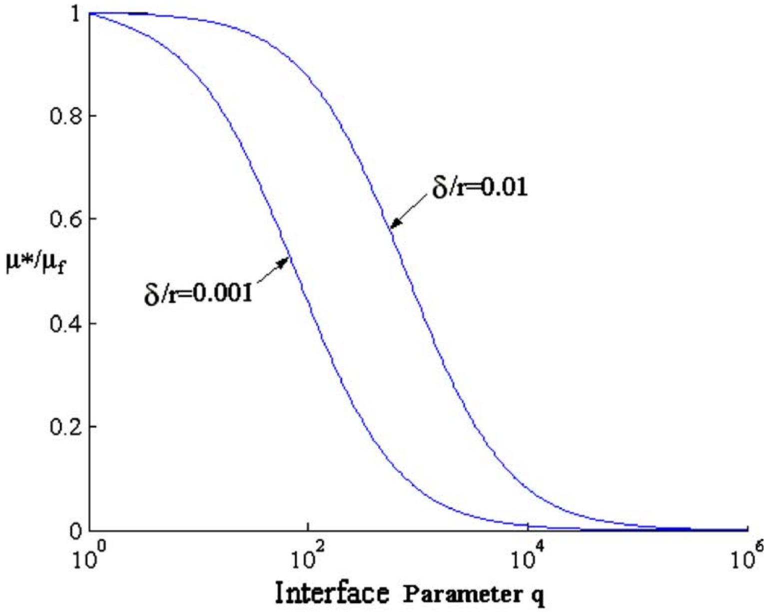

Fig. 5. Variation of effective shear modulus with the interface parameter q.

show that the nature of the interphase has significant effects on stiffness and the thermal expansion coefficient of the CSA.

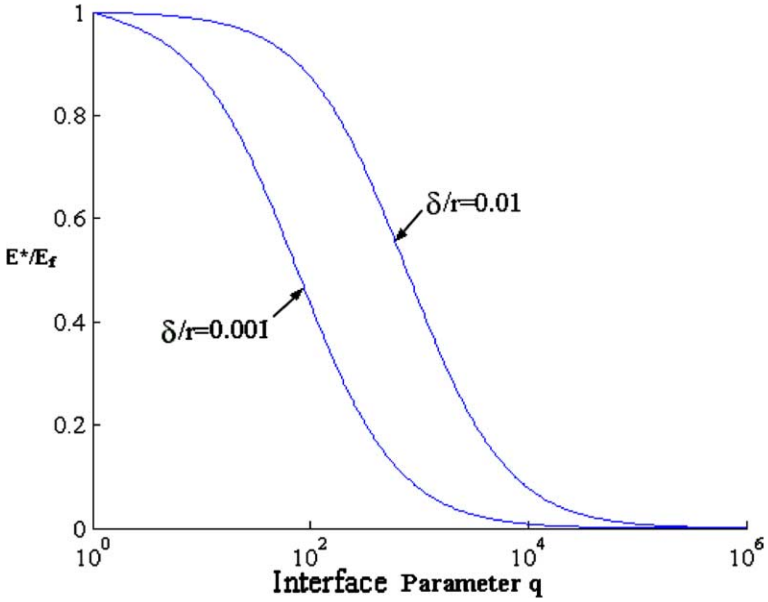

Fig. 6. Variation of effective Young's modulus with the interface parameter q.

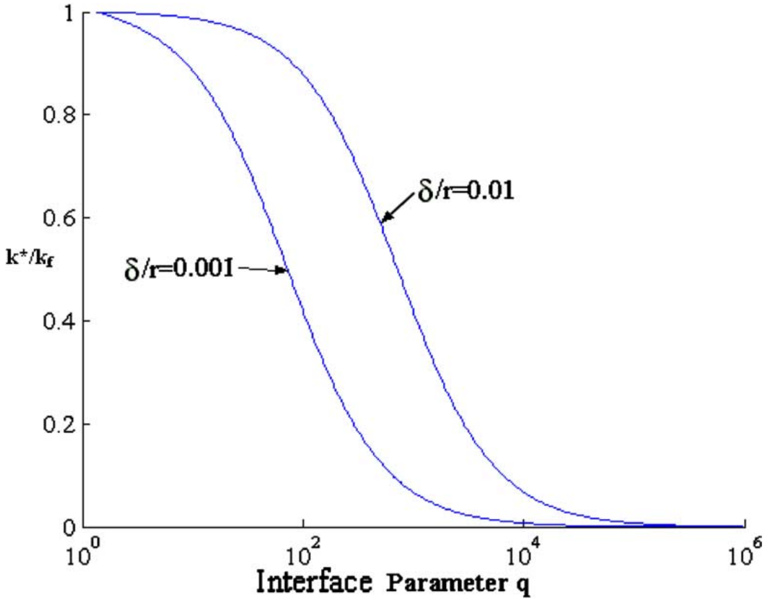

Fig. 7. Variation of effective bulk modulus with the interface parameter q.

4. Mechanical properties of two-phase composites with imperfect interface

In this section, Ju and Chen (1994a,b) elastic moduli micromechanical model for particulate composites with perfectly bonded interfaces is modified to account for interface imperfections. Several numerical

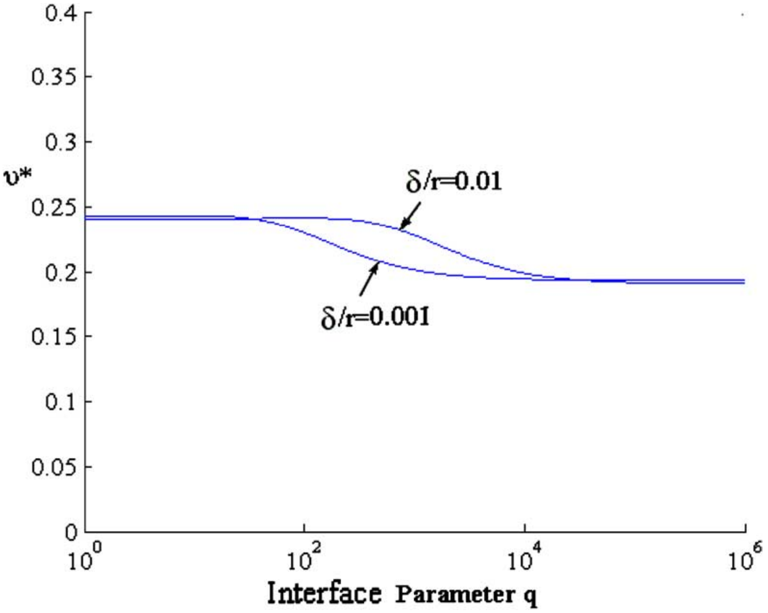

Fig. 8. Variation of effective Poisson's ratio with the interface parameter q.

simulation are conducted to show the effects of the interphase properties on the overall properties of the two-phase composite with imperfect interface. All filler particles are assumed to be spherical and both the matrix and filler particles are isotropic and elastic.

Ju and Chen (1994a,b) non-interacting solution for the effective elastic properties of two-phase composite with perfect bond can be modified for imperfect interfaces as follows; Non-interacting solution effective bulk modulus and effective shear modulus, respectively, are given by, Ju and Chen (1994a,b) as;

$$k = k_{\mathrm{m}}\left\{1 + \frac{3(1-v_{\mathrm{m}})(k^{*}-k_{\mathrm{m}})\phi_{\mathrm{f}}}{3(1-v_{\mathrm{m}})k_{\mathrm{m}} + (1-\phi_{\mathrm{f}})(1+v_{\mathrm{m}})(k^{*}-k_{\mathrm{m}})}\right\} \tag{21}$$

$$\mu = \mu_{\mathrm{m}}\left\{1 + \frac{15(1-v_{\mathrm{m}})(\mu^{*}-\mu_{\mathrm{m}})\phi_{\mathrm{f}}}{15(1-v_{\mathrm{m}})\mu_{\mathrm{m}} + (1-\phi_{\mathrm{f}})(8-10v_{\mathrm{m}})(\mu^{*}-\mu_{\mathrm{m}})}\right\} \tag{22}$$

The pairwise interacting solutions for the effective properties of two-phase composite with imperfect bond can be given as;

$$k = k_{\mathrm{m}}\left\{1 + \frac{30(1-v_{\mathrm{m}})(3\gamma_1+2\gamma_2)\phi_{\mathrm{f}}}{3\beta_1+2\beta_2-10(1+v_{\mathrm{m}})(3\gamma_1+2\gamma_2)\phi_{\mathrm{f}}}\right\} \tag{23}$$

$$\mu = \mu_{\mathrm{m}}\left\{1 + \frac{30(1-v_{\mathrm{m}})\gamma_2\phi_{\mathrm{f}}}{\beta_2-4(4-5v_{\mathrm{m}})\gamma_2\phi_{\mathrm{f}}}\right\} \tag{24}$$

where

$$\beta_1 = 2(5v_{\mathrm{m}}-1) + 10(1-v_{\mathrm{m}})\left(\frac{k_{\mathrm{m}}}{k^{*}-k_{\mathrm{m}}} - \frac{\mu_{\mathrm{m}}}{\mu^{*}-\mu_{\mathrm{m}}}\right) \tag{25}$$

$$\beta_2 = 2(4-5v_{\mathrm{m}}) + 15(1-v_{\mathrm{m}})\frac{\mu_{\mathrm{m}}}{\mu^{*}-\mu_{\mathrm{m}}} \tag{26}$$

and

$$\gamma_1 = \frac{5\phi_f}{8\beta_2^2}\left\{(13-14v_m)v_m - \frac{8\beta_1}{3\beta_1+2\beta_2}(1-2v_m)(1+v_m)\right\} \tag{27}$$

$$\gamma_2 = \frac{1}{2} + \frac{5\phi_f}{16\beta_2^2}\left\{(25-34v_m+22v_m^2) - \frac{6\beta_1}{3\beta_1+2\beta_2}(1-2v_m)(1+v_m)\right\} \tag{28}$$

where k^* and μ^* are the effective bulk modulus and shear modulus of CSA, ϕ_f is the filler particle volume fraction.

The material studied in this work is a special composite with the constituent materials of ATH particle fillers and PMMA matrix with different interfacial properties. Test samples with different interfacial adhesion properties were manufactured for this project. Table 1 shows the chemical and physical composition of the specimens. The following phase properties are used for the numerical analysis.

ATH filler: $E_f = 70$ GPa, $v_f = 0.24$
PMMA matrix : $E_m = 3.5$ GPa, $v_m = 0.31$
Filler particle volume fraction is $\phi_f = 0.48$
Non-dimensional interface parameter is defined as $q = E_f/E_i$

Three different composite materials, (named as A, B, C) were manufactured for this project. Composite material A has the strongest interfacial adhesion among the three composites we studied due to addition of a special adhesion promoting additive, Dupont CA-14, to the surface of the filler ATH agglomerate, where the value of non-dimensional interface parameter q can be assumed to be 1. The interfacial adhesion strength of composite C is the weakest among the three composites due to the debonding promoting additive, Dupont CA-11, applied to the surface of the filler ATH agglomerate. The value of non-dimensional interface parameter q for composite C can be assumed to be a very large number. The interfacial strength of the Composite B is moderate and is between the interfacial strength of composite A and composite C. The value of non-dimensional interface parameter q for composite B can be assumed to be between that values for composites A and C. The elastic Young's moduli for composites A, B and C are also determined by uniaxial tensile tests at a strain rate of 1×10^{-4} s^{-1} according to ASTM D638-98.

The effects of the interface properties on the effective Young's modulus of two-phase composite are shown in Figs. 9–11. Figures also compare experimental test data with analytical model simulations for samples with different interface properties. Results shown in Figs. 9–11 indicate that Pairwise interacting solution yields a better approximation of the overall elastic modulus. These figures present the effective Young's modulus as a function of interphase region elastic modulus (q) and for different interphase thicknesses. As the interphase region gets thinner, effective Young's modulus value remains close to perfectly bonded composite material properties. This is due to the fact that, for the same value of E_i, thinner interphase region yields higher stiffness in the interphase region, thicker interphase is more flexible.

Table 1
The chemical and physical composition of the materials tested

Composite material	Filler particle coating for interfacial bond	Constituents & Volume fraction		Average filler particle size (μm)
		Matrix (PMMA)	Filler particle (ATH)	
A	Adhesion promoting additive Dupont CA-14	52%	48%	35
B	No additives	52%	48%	35
C	Debonding promoting additive Dupont CA-11	52%	48%	35

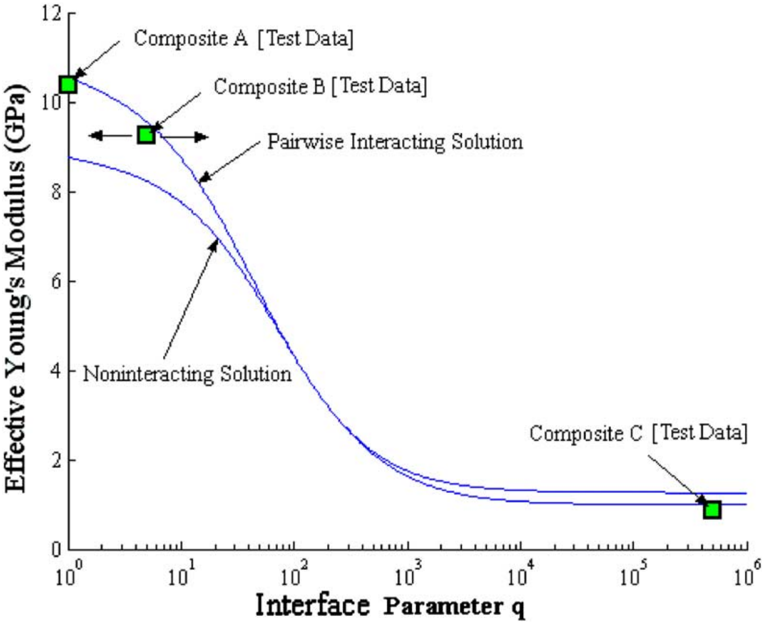

Fig. 9. Effective Young's modulus vs. interphase parameter at volume fraction of 48% with $\delta/r = 0.1$.

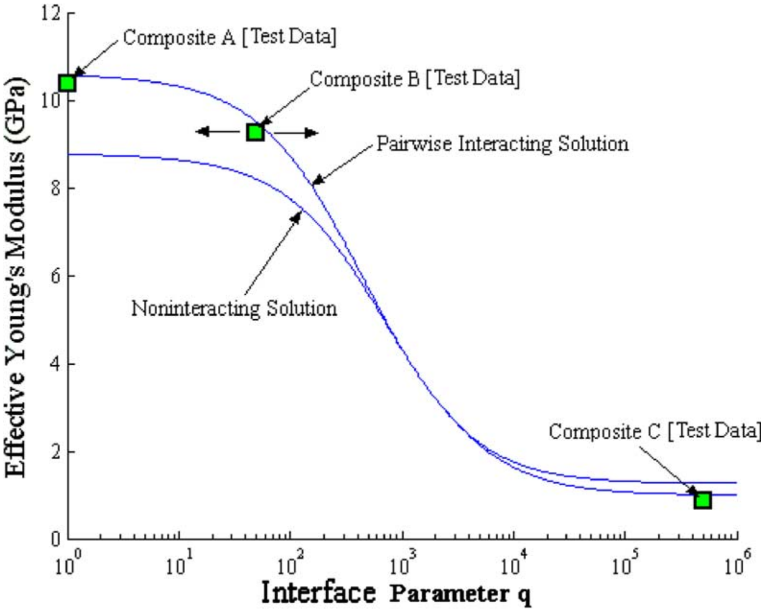

Fig. 10. Effective Young's modulus vs. interphase parameter at volume fraction of 48% with $\delta/r = 0.01$.

It is expected that the interface parameters will be different in tension and compression. Such an effect is easily incorporated into the effective bulk modulus and thermal expansion coefficient analysis. It will change the bulk moduli for isotropic tension and compression, as well as the thermal expansion coefficients

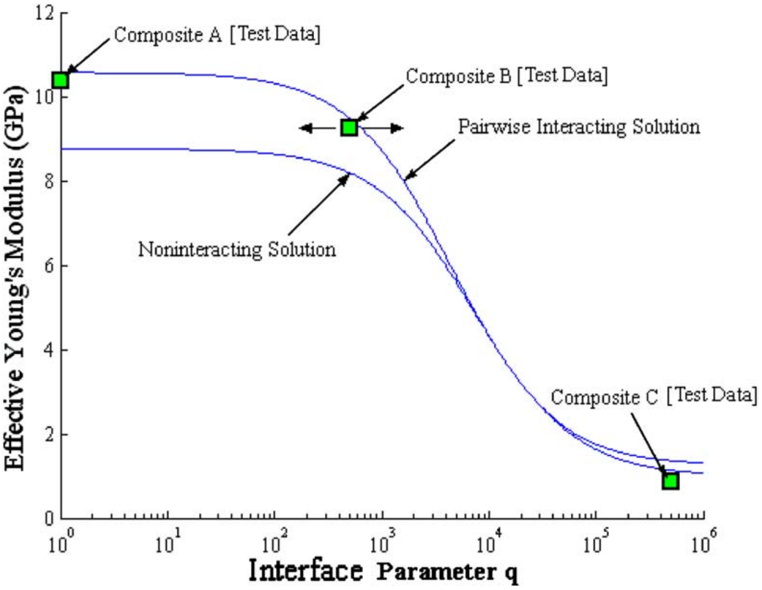

Fig. 11. Effective Young's modulus vs. interphase parameter at volume fraction of 48% with $\delta/r = 0.001$.

for heating and cooling. The sign of the radial interphase stress can determine the choice among the different interface parameters. But this presents a rather difficult problem in which there are different interface regions defined by tensile and compressive normal tractions and their boundaries are a priori unknown. Basaran et al. (2004) have shown experimentally that composites A, B and C when undergoing compressive stresses, behave as if they have perfectly bonded interfaces. Hence this experimental finding helps us to simplify the modeling process.

As a related matter, it should be mentioned that if the elasticity problem with interface conditions is regarded in a mathematical fashion without reference to the physical situation it describes, there arises the following difficulty: the normal displacement continuity cannot be negative for this since it would imply penetration of the matrix into the inclusion. The mathematical remedy for this problem would be to require perfect normal bonding when the normal traction is compressive and imperfect bonding for the case of tension. This would complicate the problem enormously. But, if there is a compliant thin interphase, then the normal displacement discontinuities are defined by the normal deformation of this interphase, which can take place both outwardly and inwardly. The interphase contribution is approximated by an imperfect interface condition. This approximation assumes that the interface between matrix and inclusion interface can be moved inward by a small distance equal to the interphase thickness.

5. Conclusions

The proposed effective elastic moduli micromechanical model for particle filled acrylic composites with imperfect interface between particle and matrix successfully predicted that interfacial conditions have great influence on the elastic properties of composites. It is observed that increasing the interfacial thickness or reducing the interfacial strength between the filler particles and matrix would reduce the effective elastic modulus seriously, which would most likely lead to more ductile behavior and fatigue life could be improved. Increasing the elastic modulus of the interphase region leads to a higher effective elastic material properties. Based on these findings it is reasonable to conclude that by controlling the interphase region thickness and elastic properties it is possible to control the ductility and fatigue life of this particle filled composite.

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