

# Finite element modelling of the interphase region on the mechanical behaviour of a composite containing micrometer sized spherical particles

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## Abstract

When a particle or filler is embedded into a polymer matrix, a third region is generated between the filler and matrix due to their chemical reaction between. This region is known as the interphase region. The properties of the interphase are affected by the material properties of the particles and the polymer matrix as well as the volume fraction of the composite. We implement the interphase mathematical model into the finite element method algorithm to investigate the effect of the interphase region on the deformation behaviour of the composite. The properties of the interphase at the matrix boundary are assumed to be smooth and continuous, while discontinuous at the particle boundary. The finite element algorithm employs Galerkin weak formulation with quadratic shape functions. The displacement, strain and stress of the composite are calculated along the radial axis originated from the

centre of a particle. The finite element modelling results are presented.

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## 1 Introduction

Adding fillers to a polymer matrix was only meant to decrease the price of the product. However, soon it was discovered that the properties of the resultant composite altered significantly from the properties of the matrix polymer, such as flame retardancy [5], improved overall composite properties, decreased specific heat, increased heat conductivity, increased stiffness properties and strength [1, 5].

The overall properties of a particle-reinforced composite are difficult to determine due to the complex bonding between the filler and matrix. In some

research a nonrealistic perfect bonding between the filler and matrix was assumed [6, 9]. A third region, the interphase, is generated when fillers are added to a polymer matrix. The effects of the interphase on the overall properties of the composite include changing the elastic properties [1, 3], thermal expansion coefficient [4] and stress and strain behaviors [8]. In the authors' previous work [1, 7], an interphase mathematical model was established based on a one dimensional, three phase, composite model. The mathematical model describes the properties and behaviour of the inhomogeneous interphase region between a spherical micrometer sized particle and a polymer matrix. Using the interphase mathematical model, the shear modulus and Young's modulus were calculated and compared well with experimental results.

The one dimensional interphase mathematical model is implemented into the Finite Element Method (FEM) algorithm based on the Galerkin Weak formulation with quadratic shape functions. The displacement, strain and stress distributions in the composite were obtained and presented.

## 2 The interphase model

The model of a representative portion of a three phase composite is shown in Figure 1(a). When a spherical filler of radius  $r_f$  is added to a polymer matrix, an annular interphase region with the inner radius  $r_f$  and outer radius  $r_i$  is created between the filler and the polymer matrix, of which the outer radius is  $r_m$  (Figure 1(a)). The material properties of the filler and polymer matrix, such as the Young's Modulus, Poisson ratios and coefficients of thermal expansion, are isotropic and of different constant values. However, the properties of the interphase region are inhomogeneous [2, 4] and some functions of the spatial variable  $r$  ( $r_f \leq r \leq r_i$ ). The rate of change of an interphase property at the boundary of the filler side is non-smooth and discontinuous; while at the matrix side is smooth and continuous. Letting  $F_{\text{int}}(r)$  represent a property of the interphase region, the smooth nature of  $F_{\text{int}}(r)$  at the bound-

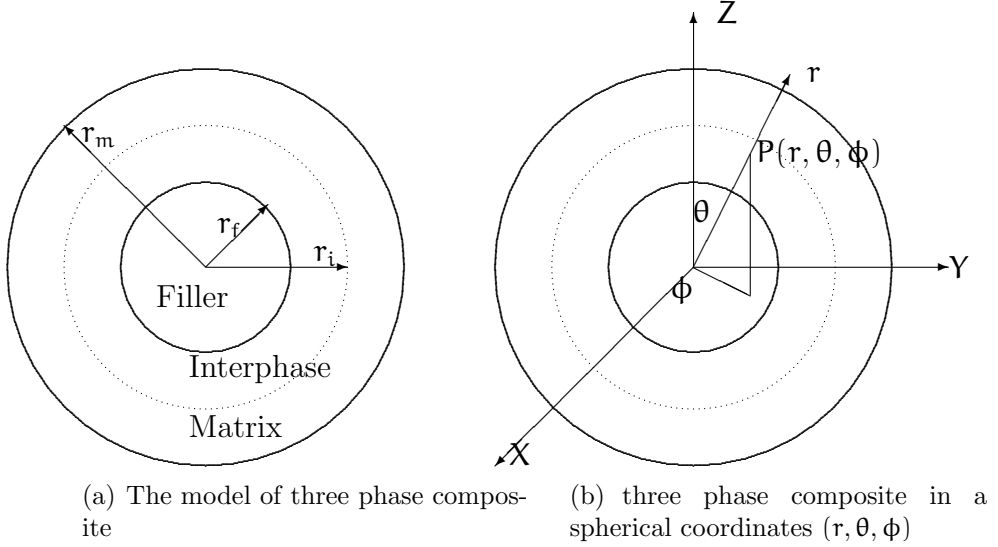


FIGURE 1: Three phase spherical filler composite.

ary of the polymer matrix side gives  $dF_{\text{int}}(r_i)/dr = 0$ . Thus, we defined the governing equation of the interphase for the property [1]

$$\frac{dF_{\text{int}}(r)}{dr} = Ar^\eta(r_i - r)e^{\gamma r}, \quad r \in [r_f, r_i], \quad (1)$$

where parameters  $\eta$  and  $\gamma$  are arbitrary integers, and for simplicity,  $\eta = 1$  and  $\gamma = 1$  were chosen [1]. The boundary conditions for equation (1) are

$$F_{\text{int}}(r_f) = kF_f, \quad F_{\text{int}}(r_i) = F_m, \quad (2)$$

where  $F_f$  is a property of the filler;  $F_m$  is a property of the polymer matrix; and  $k$  is a positive constant. A property of the composite is discontinuous at the boundary of the filler and interphase except for  $k = 1$ , for which it is continuous as  $F_{\text{int}}(r) = F_f$ . Using the ratio  $J = F_{\text{int}}(r_f)/F_m$  to describe the state of discontinuity at the boundary of the filler and interphase [4], for a given  $J$ , the value of  $k = J \cdot F_m/F_f$ . The arbitrary constant  $A$  in equation (1) is determined once the state of discontinuity at the boundary of the filler and interphase is defined.

Solving equation (1) subject to the boundary conditions (2), the following solution is obtained

$$F_{\text{int}}(\mathbf{r}) = A\{e^r [-r^2 + (r_i + 2)(r - 1)] + e^{r_i}(2 - r_i)\} + F_m, \quad \mathbf{r} \in [r_f, r_m], \quad (3)$$

where

$$A = \frac{kF_f - F_m}{e^{r_f} [-r_f^2 + (r_i + 2)(r_f - 1)] + e^{r_i}(2 - r_i)}.$$

Equation (3) describes a property of the interphase region as a function of the spatial variable  $r$ .

### 3 Formulation of displacement field

The three phase composite model is analysed in the spherical polar coordinates  $(r, \theta, \phi)$  shown in Figure 1(b). Suppose that, by some means, the composite is deformed so that its points undergo displacement. Let the displacement of an arbitrary point be  $\vec{\mathbf{u}} = (\mathbf{u}_r, \mathbf{u}_\theta, \mathbf{u}_\phi)$  referred to axes  $r$ ,  $\theta$  and  $\phi$ , respectively. When the temperature is constant and deformation is spherically symmetric, the displacement of the composite is independent of  $\theta$  and  $\phi$ . The strain  $\varepsilon$  is

$$\varepsilon_r = \frac{d\mathbf{u}_r}{dr}, \quad \varepsilon_\theta = \varepsilon_\phi = \frac{\mathbf{u}_r}{r}, \quad \varepsilon_{\theta\phi} = \varepsilon_{\phi r} = \varepsilon_{r\theta} = 0, \quad (4)$$

where  $\varepsilon_r$ ,  $\varepsilon_\theta$  and  $\varepsilon_\phi$  are the normal strains and  $\varepsilon_{\theta\phi}$ ,  $\varepsilon_{\phi r}$  and  $\varepsilon_{r\theta}$  are the shear strains. Consequently, the stress is determined from Hooke's law:

$$\begin{aligned} \sigma_{\theta\phi} &= \sigma_{r\phi} = \sigma_{r\theta} = 0, \\ \sigma_r &= \frac{E}{(1 + \nu)(1 - 2\nu)} [(1 - \nu)\varepsilon_r + \nu\varepsilon_\theta + \nu\varepsilon_\phi], \\ \sigma_\theta &= \sigma_\phi = \frac{E}{(1 + \nu)(1 - 2\nu)} [\varepsilon_\theta + \nu\varepsilon_r], \end{aligned} \quad (5)$$

where  $\sigma_r$ ,  $\sigma_\theta$  and  $\sigma_\phi$  are the axial stresses, while  $\sigma_{\theta\phi}$ ,  $\sigma_{\phi r}$  and  $\sigma_{r\theta}$  are the shear stresses.  $E$  and  $\nu$  are the Young's modulus and Poisson ratio, respectively.

Substituting equation (4) into equation (5) gives the stress in terms of displacement in the radial direction only

$$\sigma_r = X \frac{du_r}{dr} + Y \frac{u_r}{r}, \quad \sigma_\theta = Z \frac{du_r}{dr} + W \frac{u_r}{r},$$

where

$$X = X_\omega = \frac{(1 - \nu_\omega)E_\omega}{(1 + \nu_\omega)(1 - 2\nu_\omega)}, \quad Y = Y_\omega = \frac{2\nu_\omega E_\omega}{(1 + \nu_\omega)(1 - 2\nu_\omega)},$$

$$W = W_\omega = \frac{X_\omega}{1 - \nu_\omega}, \quad Z = Z_\omega = \frac{Y_\omega}{2} \quad \text{for } \omega = f, i, m.$$

Note that the material properties of the filler and polymer matrix  $E_f$ ,  $E_m$ ,  $\mu_f$  and  $\mu_m$  are known constants, while the Young's modulus and Poisson ratios of the interphase  $E_i$  and  $\mu_i$  are the variables of  $r$  and determined by equation (3).

The equilibrium equation for the stress distribution of the composite is

$$\frac{d}{dr} \left( X \frac{du_r}{dr} \right) + \frac{d}{dr} \left( Y \frac{u_r}{r} \right) + \frac{2(X - Z)}{r} \frac{du_r}{dr} + \frac{2(Y - W)}{r^2} u_r = 0, \quad (6)$$

$$u_r(0) = 0, \quad u_r(r_m) = \bar{\epsilon} r_m.$$

## 4 The finite element algorithm

The interphase mathematical model is implemented into the FEM algorithm to calculate the displacement along the radial axis ( $r$  direction). We discretize the  $r$  domain into  $N$  quadratic elements with a total nodal number of  $2N + 1$ , Taking the piecewise quadratic functions  $\psi_p(r)$  as the weighting functions and  $r_p$  the nodal coordinate of the corresponding node  $p$ , the weak formulation of equation (6) is

$$\int_{r_1}^{r^{2N+1}} \left[ \frac{d}{dr} \left( X \frac{du_r}{dr} \right) + \frac{d}{dr} \left( Y \frac{u_r}{r} \right) + \frac{2(X-Z)}{r} \frac{du_r}{dr} + \frac{2(Y-W)}{r^2} u_r \right] \psi_p(r) dr = 0, \quad (7)$$

for  $p = 1, 2, \dots, 2N + 1$ . Considering the essential boundary conditions at both ends, we simplify equation (7) by integrating by parts to obtain

$$\int_{r_1}^{r^{2N+1}} \left[ \left( \frac{2(X-Z)}{r} \frac{du_r}{dr} + \frac{2(Y-W)}{r^2} u_r \right) \psi_p(r) - \left( X \frac{du_r}{dr} + Y \frac{u_r}{r} \right) \frac{d\psi_p(r)}{dr} \right] dr = 0. \quad (8)$$

The approximate solution is assumed to be  $u_r = \sum_{q=1}^{2N+1} U_q \psi_q(r)$  where  $U_q$  ( $q = 1, 2, \dots, 2N + 1$ ) are the nodal displacements. Substituting into equation (8) yields the following set of discretised equations

$$M_{p,q} U_q = L_p \quad \text{for } p, q = 1, 2, \dots, 2N + 1, \quad (9)$$

where

$$M_{p,q} = \int_{r_1}^{r^{2N+1}} \left[ \left( \frac{2(X-Z)}{r} \frac{d\psi_q(r)}{dr} + \frac{2(Y-W)}{r^2} \psi_q(r) \right) \psi_p(r) - \left( X \frac{d\psi_q(r)}{dr} + \frac{Y}{r} \psi_q(r) \right) \frac{d\psi_p(r)}{dr} \right] dr$$

and

$$L_p = \begin{cases} 0, & \text{if } p \neq 2N + 1, \\ \bar{\epsilon} r_m, & \text{if } p = 2N + 1. \end{cases}$$

Note that the regions of the filler, interphase and matrix are discretised into  $n$ ,  $m$  and  $l$  elements with the equal length  $h_1 = r_f/n$ ,  $h_2 = (r_i - r_f)/m$  and  $h_3 = (r_m - r_i)/l$  in each region, respectively.

The axial strain and axial stress are found using  $\epsilon_r = \partial u_r / \partial r$  and  $\sigma = E \epsilon$ , once equation (9) is solved for the displacement.

TABLE 1: Young’s modulus and Poison ratio for E-glass and BISGMA/TEGDMA epoxy

Property	E-glass filler	BISGMA/TEGDMA epoxy
Young’s modulus E (GPa)	75	1.7
Poison ratio $\nu$	0.24	0.35

## 5 Result and discussion

### 5.1 Material properties

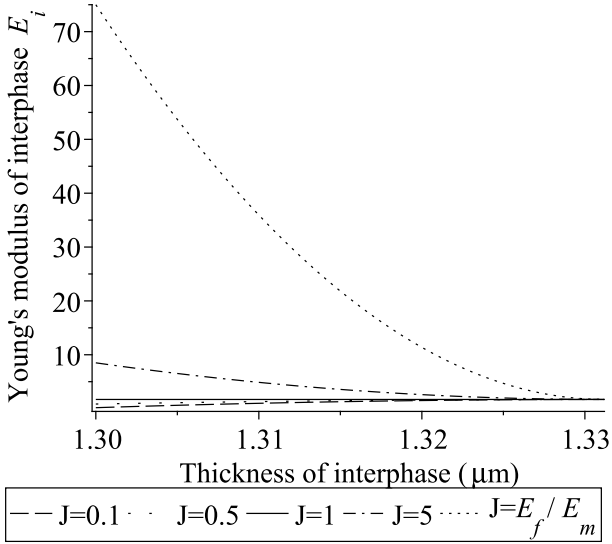
The biocompatible composite, E-glass particle reinforced BISGMA/TEGDMA epoxy, is used in the present study [1, 6]. The average diameter of the E-glass particles is 2.6  $\mu\text{m}$ . The Young’s moduli and Poison ratios of the E glass particles and the BISGMA/TEGDMA polymers are listed in Table 1. According to these properties, the average thickness of the interphase is 2.4% of the radius of the filler [1, 6]. For an iso-displacement loading, a small strain of  $\bar{\epsilon} = 0.1\%$  is used [6].

### 5.2 Results and discussions

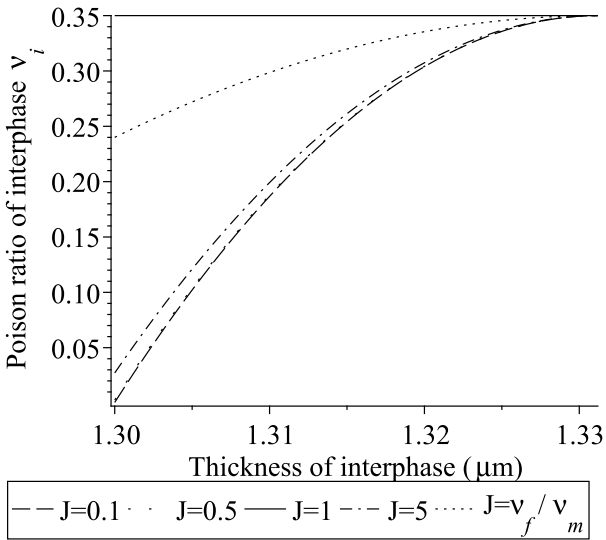
Figures 2(a) and 2(b) show the variations of Young’s modulus and Poison ratio over the interphase region for different J values. The volume fraction of the filler is assumed as 20%. When  $J > 1$ , the Young’s modulus of the interphase decreases moving away from the particle, while the Poison ratio increases. When  $J < 1$ , the Young’s modulus only increases slightly but the Poison ratio increases rapidly. In both cases, the Young’s modulus and Poison ratio are continuous and smoothly merge with the polymer matrix.

Figures 3(a) and 3(b) show the normalized radial displacement  $u_r/u(r_m)$  versus the normalized radial distance  $r/r_m$  from the centre of the filler to



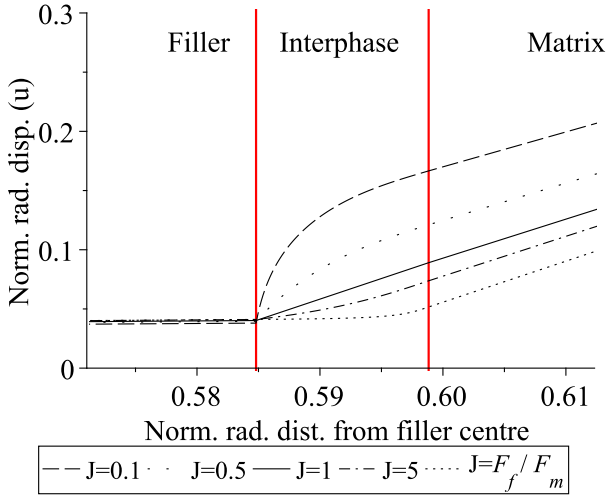


(a) Young's modulus

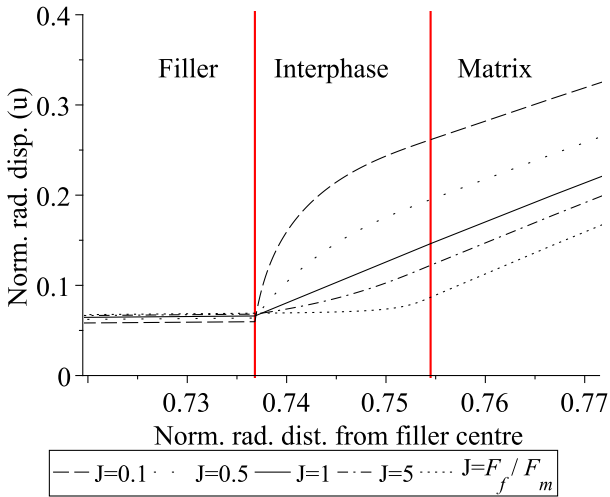


(b) Poison ratio

FIGURE 2: Variations of Young's modulus (a) and Poison ratio (b) over the interphase region. Interphase thickness 2.4%



(a) Volume fraction of fillers: 20%

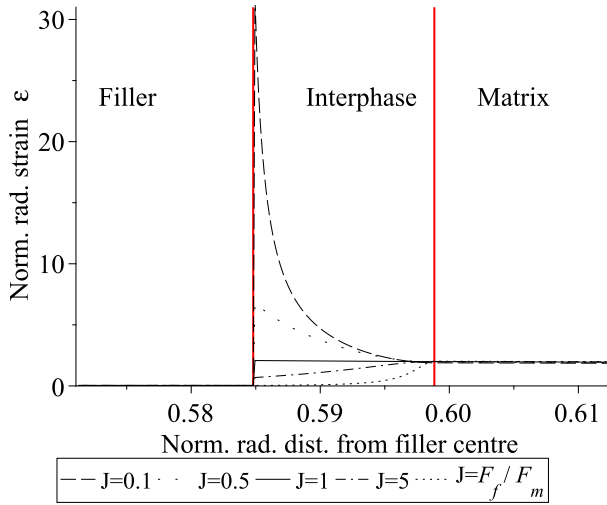


(b) Volume fraction of fillers: 40%

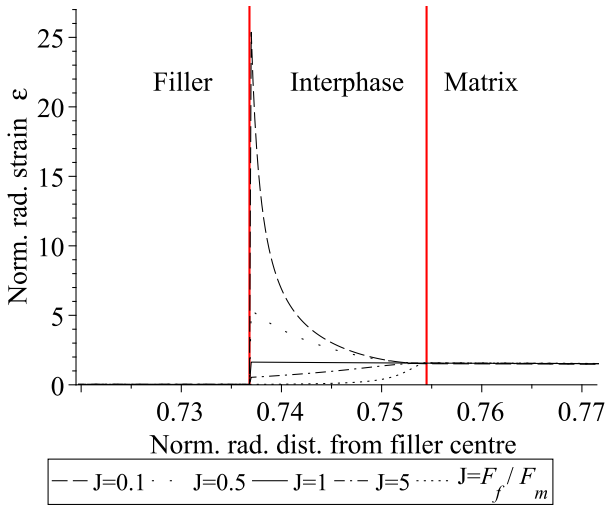
FIGURE 3: Normalised radial displacement against radial distance.

the outer surface of the matrix for the volume fractions 20% and 40%, respectively. Figure 3(a) shows that the displacement is independent of  $r$  in the filler region, though an increase of the  $J$  value results in a slight decrease of the displacement. In the interphase region, the displacement becomes a function of  $r$  and it increases more significantly for the cases of  $J < 1$ . The displacement is continuous at both boundaries of the filler-interphase and interphase-matrix regardless of the  $J$  value. However, the displacement at the boundary of the filler and interphase is non-smooth, while smooth at the interphase-matrix boundary. Only for the case of  $J = F_f/F_m$ , the displacement is continuous and smooth at both boundaries of the filler-interphase and interphase-matrix. Figures 3(a) and 3(b) also show that the volume fraction of fillers of a composite has a great influence on the displacement in the composite. The larger the volume fraction of fillers is, the greater increase in the magnitude of displacement.

Figures 4(a) and 4(b) show the strain variations in the composite for volume fractions of 20% and 40%. The strain is normalized to the strain at the outer surface of the matrix  $\varepsilon/\varepsilon_m$ . The strain in the filler region is small for all  $J$ . However, the strain distribution in the interphase region is strongly influenced by the value of  $J$ . When  $J > 1$ , the strain decreases rapidly as  $r$  increases, while the strain increases slowly with  $J < 1$ . The strain at the filler-interphase boundary is discontinuous for all values of  $J$  except the case of  $J = F_f/F_m$ , for which the strain seems continuous and smooth. The graphs also show the strain is least in the filler region. The results of the stress distribution in a composite are presented in Figures 5(a) and 5(b). As shown in Figure 5(a), the stresses in the filler region and matrix region are constants of different values, higher magnitudes in the filler region. At the boundary of filler and interphase, stresses are discontinuous for all the values of  $J$  except the case of  $J = F_f/F_m$ . The magnitude of the stress decreases as  $r$  increases towards the boundary of the interphase and matrix. Figure 5(b) presents the stress distribution when the volume fraction is increased to 40%. Overall, the stress behaves similar to the case in Figure 5(a) but the magnitudes of the stress are decreased.

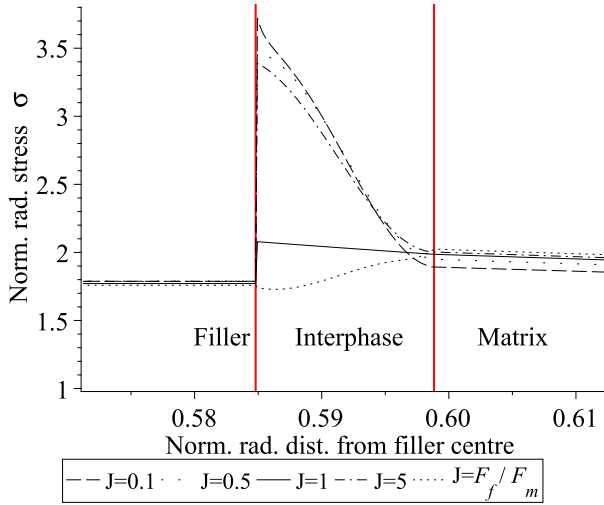


(a) Volume fraction of fillers 20%

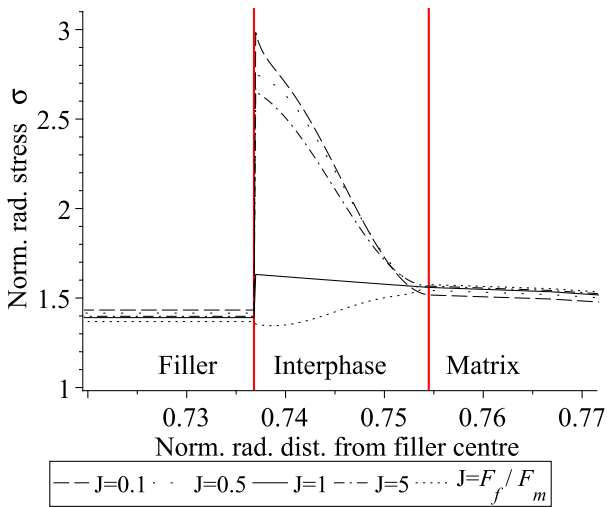


(b) Volume fraction of fillers 40%

FIGURE 4: Normalised radial strain against radial distance.



(a) Volume filler fraction 20%



(b) Volume filler fraction 40%

FIGURE 5: Normalized radial stress against radial distance.

## 6 Conclusion

The new mathematical model describing a property of the interphase between a spherical particle and polymer matrix provides a good solution for predicting the mechanical properties of a composite.

The Young's modulus and Poisson ratio vary over the interphase region as the state of discontinuity varies, measured by the J values. Overall, the Young's modulus of the composite decreases as the J value increases, while Poisson ratio increases.

The FEM solution of the mathematical model provides a good prediction of the variation of displacement, strain and stress along the radial axis over the interphase region at different states of discontinuity.

The increase of the volume fraction of the filler results in decrease of the strain and stress, and increase of the displacement in the composite.

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