THEORETICAL AND COMPUTER MODELLING OF DEBONDING MECHANISMS IN FIBROUS MICRO- AND NANO-COMPOSITES

J.J. Rushchitsky[1] and I.A. Guz[2]

[1]Department of Dynamics of Polyaggregate Systems, Timoshenko Institute of Mechanics, Kiev, 03680, Ukraine.

[2]School of Engineering and Physical Sciences, University of Aberdeen, Scotland, UK.

ABSTRACT

Debonding is one of common failure mechanisms in fibre-reinforced composite materials (Broutman and Krock [1]). Both shear forces acting along or perpendicularly to the fibres and tensile forces acting perpendicularly to the fibres can contribute to the breakdown of interfacial adhesion between the fibres and the matrix. In this paper, deformation due to plane harmonic waves propagating along the fibres and polarised perpendicular direction is considered. To describe the behaviour of the material, a second-order continuum theory, namely the theory of two-component elastic mixtures (Guz, Rushchitsky [2-4]), is used. Analytical solution to the problem is derived and then used to study wave propagation phenomena in fibre reinforced composite materials with epoxy matrix. Four types of fibres are considered: Thornel-300 carbon fibres; carbon whiskers; zigzag carbon nanotubes; chiral carbon nanotubes. Of particular interest is the case when two waves are propagating in the material in-phase or in anti-phase, with the amplitudes strongly dependent on the frequency. Theoretical analysis and numerical results indicate that in unidirectional fibre-reinforced micro- and nanocomposites, the second mode of the transverse wave, propagating along the fibres and polarised perpendicularly to the fibre direction, can be critical to the strength of the material at high frequencies. This mode generates anti-phase vibrations in the composite constituents and forces that could cause interfacial debonding. This phenomenon can be classified as a new mechanism of debonding in fibre-reinforced composite materials.

1 THEORETICAL ANALYSIS OF TRANSVERSE WAVE USING TWO-COMPONENT ELASTIC MIXTURE THEORY

The two-component elastic mixture theory was introduced for description of wave propagation phenomena within the micromechanics of materials. The theory captures well the dynamics of elastic deformation of materials with microstructure (Rushchitsky [5], Bedford et al [6], Bedford and Drumheller [7], McNiven and Mengi [8], Rushchitsky [9, 10]) and is in a good agreement with experimental data for metal matrix and polymer matrix composites.

Within this theory, the two-component mixture is treated as two interpenetrating and interacting continua. Kinematics of deformation is described by two partial displacement vectors $\vec{u}^{(\alpha)}\ \left(u_i^{(\alpha)} = u_i^{(\alpha)}(x_1, x_2, x_3, t),\ \alpha = 1; 2\right)$; consequently, there are two partial strain tensors $\varepsilon_{ik}^{(\alpha)}$, relative displacement vector $\vec{u}^{(1)} - \vec{u}^{(2)}$, and two partial stress tensors $\sigma_{ik}^{(\alpha)}$. Each partial quantity is obtained by averaging the corresponding quantity for the matrix or the fibres over the volume of the whole material. Equations of dynamics for free waves in a two-component elastic mixture have the form of the following six coupled equations

$$C_{iklm}^{(\alpha)} u_{l,mk}^{(\alpha)} + C_{iklm}^{(3)} u_{l,mk}^{(\delta)} + \beta_k \left(u_k^{(\delta)} - u_k^{(\alpha)}\right) = \left(\rho_{\alpha\alpha} + \rho_{12}^{(k)}\right)\ddot{u}_k^{(\alpha)} - \rho_{12}^{(k)}\ddot{u}_k^{(\delta)} \tag{1}$$

Three tensors of elastic constants $C_{iklm}^{(k)}$, two partial densities $\rho_{\alpha\alpha}$, vector of interacting forces β_k, and vector of inertial interaction $\rho_{12}^{(k)}$ form the full set of physical constants in the theory of mixtures. In the case of a transversally isotropic mixture, often used to describe fibre-reinforced materials, the full set consists of 21 constants.

The following analysis examines the plane waves in mixtures. The plane wave is described by partial displacements $\vec{u}^{(\alpha)}(x,t) = \vec{u}^{(\alpha)o} \cdot e^{i(\xi - \omega t)}$, which differ by amplitudes only. Here $\vec{u}^{(\alpha)o} = \left\{u_1^{(\alpha)o}, u_2^{(\alpha)o}, u_3^{(\alpha)o}\right\}$ are the initials amplitudes, $\xi = \vec{k} \cdot \vec{r}$, and $\vec{r}$ is the radius-vector of the point $x = (x_1, x_2, x_3)$. Christoffel's equation is transformed into a set of two equations, which allows one to write down wave equations for various cases depending on the symmetry of the mixture, wave polarisation and directions of wave propagation.

For fibre-reinforced materials exhibiting transverse isotropy, the transverse wave propagating along the axis of material symmetry (for unidirectional materials this will be the fibre direction) is considered. Propagation of such wave can be described by a set of two coupled equations (plane polarised waves)

$$C_{1313}^{(\alpha)} u_{1,11}^{(\alpha)} + C_{1313}^{(3)} u_{1,11}^{(\delta)} + \beta_1 \left(u_1^{(\alpha)} - u_1^{(\delta)}\right) - \rho_{12}^{(1)} \left(\ddot{u}_1^{(\alpha)} - \ddot{u}_1^{(\delta)}\right) = \rho_{\alpha\alpha}\, \ddot{u}_1^{(\alpha)}. \qquad (2)$$

The plane wave, eqn (2), propagates along the applicate axis and is polarised in the direction, perpendicular to it, i.e. vibration occurs along the abscissa axis. Equations (2) take account of all possible linear and elastic interactions: between stresses and deformations (constants $C_{1313}^{(3)}$), shear (constant β_1) and inertia (constant $\rho_{12}^{(1)}$) forces. Following the standard procedure, a dispersion equation can be obtained for the set of Christoffel's equations. Its solution has the form

$$k_1^2 = \frac{\omega^2}{2\Delta_c}\left\{\left(C_{1313}^{(1)}\rho_{22} + C_{1313}^{(2)}\rho_{11} + B\sigma_c\right) \pm \sqrt{\left(C_{1313}^{(1)}\rho_{22} + C_{1313}^{(2)}\rho_{11} + B\sigma_c\right)^2 - 4\Delta_c W}\right\}, \qquad (3)$$

$$\Delta_c = C_{1313}^{(1)}C_{1313}^{(2)} - C_{1313}^{(3)}C_{1313}^{(3)},\ B = -\rho_{12}^{(1)} + \left(\frac{\beta_1}{\omega^2}\right),\ \sigma_c = C_{1313}^{(1)} + C_{1313}^{(2)} + 2C_{1313}^{(3)},\ W = \rho_{11}\rho_{22} - B\left(\rho_{11} + \rho_{22}\right)$$

It follows from the dispersion equation, eqn (3), that the mixture is a dispersive medium, and the waves in it always propagate in two modes, i.e. two waves with different wave numbers propagating simultaneously. The positive sign in eqn (3) corresponds to the slow (acoustic) mode, while the negative sign corresponds to the fast (optic) mode. The former exists for all frequencies, and the latter for high frequencies only. Condition $W = 0$ defines the cutting frequency

$$\omega_{cut} = \sqrt{\beta_1 \frac{\rho_{11} + \rho_{22}}{\rho_{11}\rho_{22} + \rho_{12}^{(1)}\left(\rho_{11} + \rho_{22}\right)}},$$

below which the free wave as if does not exist.

Solution to eqns (2) has the form of superposition of two harmonic waves (modes)

$$u_1^{(\alpha)}(x_1,t) = u_1^{(\alpha)o} e^{-i\left(k_1^{(\alpha)} x_1 - \omega t\right)} + l\left(k_1^{(\delta)},\omega\right) u_1^{(\delta)o} e^{-i\left(k_1^{(\delta)} x_1 - \omega t\right)} \tag{4}$$

The coefficients of amplitude re-distribution $l\left(k_1^{(1)},\omega\right), l\left(k_1^{(2)},\omega\right)$ are calculated from the following formulae

$$l\left(k_1^{(1)},\omega\right) = -\frac{C_{1313}^{(3)}\left(k_1^{(1)}\right)^2 - \beta_1 - \rho_{12}^{(1)}\omega^2}{C_{1313}^{(1)}\left(k_1^{(1)}\right)^2 + \beta_1 - \left(\rho_{11} - \rho_{12}^{(1)}\right)\omega^2}, \tag{5a}$$

$$l\left(k_1^{(2)},\omega\right) = -\frac{C_{1313}^{(2)}\left(k_1^{(1)}\right)^2 + \beta_1 - \left(\rho_{22} - \rho_{12}^{(1)}\right)\omega^2}{C_{1313}^{(3)}\left(k_1^{(1)}\right)^2 - \beta_1 - \rho_{12}^{(1)}\omega^2}. \tag{5b}$$

The microstructure of the material affects many aspects of wave propagation. In each component of the mixture, both modes propagate simultaneously, each having its own amplitude. These amplitudes strongly depend on frequency as evidenced by eqn (5). One of the consequences of this dependence is known as the energy pumping from one mode to the other [6]. This paper examines another aspect, namely, the sign change with the frequency for the functions given by eqn (5). As a result of the sign change, the amplitudes of the partial vibrations (same mode vibrations in the different components of the mixture) can be of the opposite signs, thus creating conditions for the adhesion breakdown between fibres and the matrix, especially when the amplitudes are sufficiently high.

2 COMPOSITE MATERIALS USED IN NUMERICAL EXAMPLES

Four composite materials used in the numerical examples of the next section were described in detail in [2, 7]. These materials have the same epoxy matrix (EPOF-828) and differ in the type of carbon reinforcement. The following reinforcements are considered: R1 – commercial carbon microfibre Thornel T-300 with the diameter of 8 micron; R2 – graphite microwhiskers with the diameter 1 micron; R3 – zigzag carbon nanotubes with the average tube diameter of 10 nm; R4 – chiral carbon nanotubes with the average diameter of 10 nm. Physical properties of the matrix and four carbon reinforcements are given in the Table 1. Two fibre volume fractions are considered: $c^f = 0.1$ (very small) and $c^f = 0.2$ (sufficiently small), with resulting composite materials denoted as CM1 and CM2, respectively.

Table 1: Physical properties of the constituent materials

Constituent materials	Density ρ (kPa s^2/m^2)	Young's modulus E (GPa)	Lame modulus λ (GPa)	Shear modulus μ (GPa)	Poisson's ratio ν
Epoxy resin (EPOF-828)	1.21	2.68	3.83	0.96	0.4
R1	1.75	228	131.5	88	0.3
R2	2.25	1000	576.9	385	0.3
R3	1.33	648	472.9	221	0.33
R4	1.40	1240	715.4	477	0.3

3 COMPUTER SIMULATION OF THE WAVE PATTERN. DEBONDING MECHANISM IN FIBRE-REINFORCED COMPOSITES

In this section, the transverse plane wave propagating along the fibres (along the applicate axis) and polarised perpendicularly to them (along the abscise axis) is examined. Seven physical constants are required for the analysis of this wave using the theory of structural mixtures, which can be obtained from the experiments or calculated, i.e. three elastic constants $C_{1313}^{(1)}, C_{1313}^{(2)}, C_{1313}^{(3)}$; two partial densities ρ_{11}, ρ_{22}; shear interaction constant β_1 and inertial interaction constant $\rho_{12}^{(1)}$. In this study, the values of physical constants were calculated using formulae of Bedford et al [6], Bedford and Drumheller [7], McNiven and Mengi [8] and Yeh [11]. They are given in Table 2. Once the physical constants are known, the coefficients of amplitude re-distribution, eqn (5), are calculated using *Mathematica 5.1* software.

Table 2: Physical constants required in the analysis

Composite material	$C_{1313}^{(1)}$ (GPa)	$C_{1313}^{(2)}$ (GPa)	$C_{1313}^{(3)}$ (GPa)	β_1 (10^{22} Pa/m^2)	$\rho_{12}^{(1)}$ (10^3 Pa·s^2/m^2)
CM1R1	0.0938	0.9461	0.0128	0.0115	1.006
CM1R2	0.1037	0.9568	0.0029	0.7726	1.109
CM1R3	0.1015	0.9544	0.0051	7672.3	1.094
CM1R4	0.1043	0.9574	0.0024	7748.9	1.115
CM2R1	0.2237	0.9417	0.0157	0.0164	0.7317
CM2R2	0.2363	0.9558	0.0036	1.081	0.7833
CM2R3	0.2335	0.9527	0.0062	10809	0.7756
CM2R4	0.2370	0.9566	0.0029	10885	0.7865

Figures 1 and 2 show the coefficients of amplitude re-distribution $l\left(k_1^{(1)},\omega\right)$ and $l\left(k_1^{(2)},\omega\right)$, respectively, as functions of frequency. The results are given for two composite materials with 0.2 volume fraction of Thornel T-300 microfibres (CM2R1) and zigzag nanotubes (CM2R3), but other composites from Table 2 behave in a similar way.

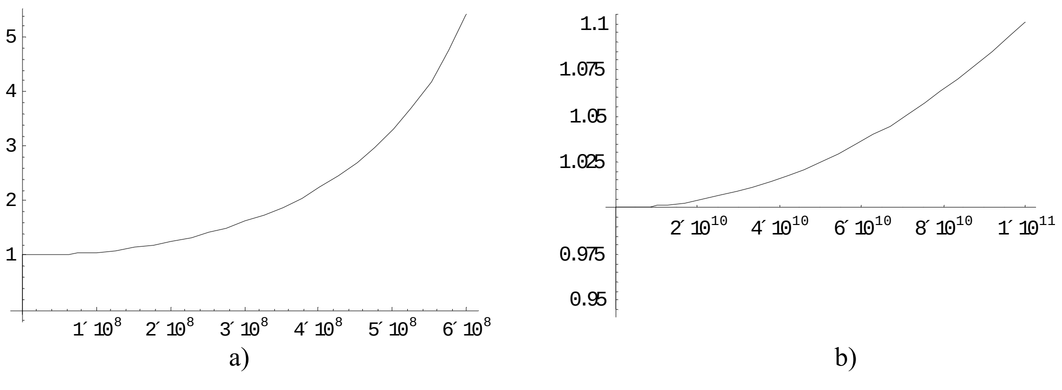
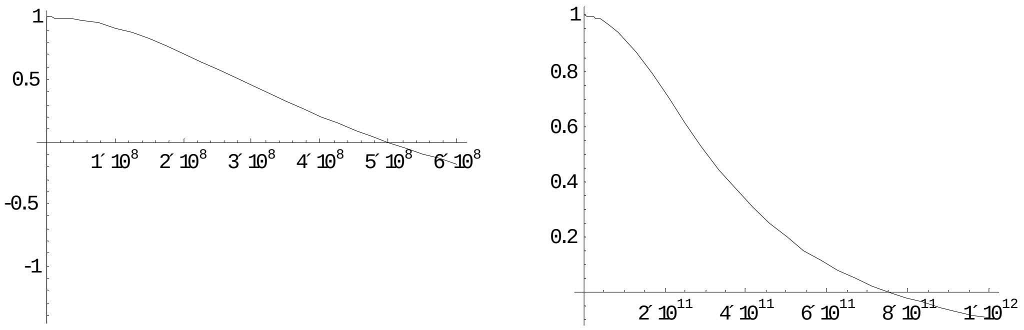


Figure 1: Coefficient of the amplitude re-distribution $l\left(k_1^{(1)},\omega\right)$ as a function of frequency for CM2R1 (a) and CM2R4 (b).



a) b)

Figure 2: Coefficient of the amplitude re-distribution $l\left(k_1^{(2)},\omega\right)$ as a function of frequency for CM2R1 (a) and CM2R4 (b).

The first mode (the slow mode – with the typical phase velocity) will propagate independently in both constituents, having the amplitude $u_1^{(1)o}$ in the fibres and $l\left(k_1^{(1)},\omega\right)u_1^{(1)o}$ in the matrix. The second mode (the fast mode – with significantly higher phase velocity) will propagate simultaneously with the first, independently in both constituents of the materials, having the amplitude $l\left(k_1^{(1)},\omega\right)u_1^{(2)o}$ in the fibres and $u_1^{(2)o}$ in the matrix.

As one can see from Fig. 1, the value of $l\left(k_1^{(1)},\omega\right)$ is always positive. It means that the fibres and the matrix always vibrate in-phase. In the same time, the value of $l\left(k_1^{(2)},\omega\right)$, Fig. 2, is positive only in the range $\omega_{cut} \le \omega \le \omega_{crit}$, where ω_{cut} is the cutting frequency of the material and ω_{crit} is some critical frequency, after which it becomes negative. The cutting frequency is $\omega_{cut} = 354$ MHz for CM2R1 and $\omega_{cut} = 290.3$ GHz for CM2R3; the critical frequency is $\omega_{crit} = 500$ MHz for CM2R1 and $\omega_{crit} = 750$ GHz for CM2R3. Therefore, for all frequencies higher than ω_{crit}, the fibres and the matrix vibrate in anti-phase. This confirms an assumption about the existence of anti-phase vibrations.

It is important to check whether or not the critical frequency ω_{crit} is the limiting frequency, and if not, to establish the frequency range, in which the components of the mixture vibrate in anti-phase. The simplest way to do this is to calculate the frequency for the wavelength, equal to the fibre diameter. This frequency can be determined from the phase velocity V and the wavelength λ as $\omega = 2\pi V/\lambda$. For the considered composite materials these quantities are as follows: $V^{eff} = 0.711$ km/s for CM2R1 and $V^{eff} = 0.768$ km/s for CM2R3; the minimal phase velocities for the second mode are $V_{ph}^{(2)} = 1.671$ km/s for CM2R1 and $V_{ph}^{(2)} = 1.876$ km/s for CM2R3; $\lambda = 8 \cdot 10^{-6}$ m for CM2R1 and $\lambda = 10 \cdot 10^{-9}$ m for CM2R3. Then the upper limit is calculated as $\omega^{upper} = 1.312$ GHz for CM2R1 and $\omega^{upper} = 1.179$ THz for CM2R3. Therefore, the frequency range for anti-phase vibrations from 500 MHz to 1.312 GHz for CM2R1 and from 750 GHz to 1.179THz for CM2R3.

Thus, based on the theoretical analysis in the framework of the theory of structural mixtures and the numerical results for concrete unidirectional fibre-reinforced composite materials, it is established that the second mode of the transverse wave, propagation along the fibres and polarised perpendicularly to the fibre direction, can create (in the range of high frequencies) a kinematical pattern that could be critical to the strength of the material. Propagation of this wave generates anti-phase vibrations in the components of the composite, which in their turn create forces, capable of breaking the interfacial adhesion between the fibres and the matrix. The phenomenon of anti-phase vibrations of the fibres and the matrix can be classified as a new debonding mechanism for fibre-reinforced composite materials.

REFERENCES

[1] *Composite materials.* Eds. Broutman L.J., Krock R.H.: Vol.1. *Interfaces in metal matrix composites*. (Ed. Metcalfe A.G.); Vol.6. *Interfaces in polymer matrix composites*. (Ed. Plueddemann E.D.) Academic Press, New York and London, 1974.

[2] Guz I.A., Rushchitsky J.J. Comparison of mechanical properties and effects in micro- and nanocomposites with carbon fillers (carbon microfibers, graphite microwhiskers and carbon nanotubes. *Mechanics of Composite Materials,* 40(3), pp.23-40, 2004.

[3] Rushchitsky J.J., Guz I.A. Comparison of characteristics of wave evolution in nonlinearly elastic micro- and nanocomposites with carbon fillers. *International Applied Mechanics,* 40(8), pp.88-98, 2004.

[4] Guz A.N., Rushchitsky J.J. Nanomaterials. On mechanics of nanomaterials. *International Applied Mechanics,* 39(11), pp.36-69, 2003.

[5] Rushchitsky J.J. Elements of the mixture theory. Kiev: Naukova dumka, 1991. (In Russian)

[6] Bedford A., Drumheller G.S., Sutherland H.J. On modeling the dynamics of composite materials. In: S.Nemat-Nasser, Mechanics Today. Vol. 3, New York: Pergamon Press, pp.1-54, 1976.

[7] Bedford A., Drumheller G.S. Theories of immiscible and structured mixtures. *International Journal of Engineering Sciences*, 21(8), pp.863-960, 1983.

[8] McNiven H.D., Mengi Y. A mathematical model for the linear dynamic behaviour of two-phase periodic materials. *International Journal of Solids and Structures*, 15(4), pp.271-280, 1979.

[9] Rushchitsky J.J. Extension the microstructural theory of two-phase mixtures to composite materials. *International Applied Mechanics*, 36(5), pp.586-614, 2000.

[10] Rushchitsky J.J. Interaction of waves in solid mixtures. *Applied Mechanics Reviews,* 52(2), pp.35-74, 1999.

[11] Yeh R.H.T. Variational bounds of unidirectional fiber reinforced composites. *Journal of Applied Physics,* 42(2), pp.419-428, 1973.