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Effect of Interface Energy on Size-Dependent Effective Dynamic Properties of Nanocomposites with Coated Nano-Fibers

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Abstract: In nanocomposites, coated nano-fibers are often used to obtain good performance, and the high interface-to-volume ratio shows great effect on the macroscopic effective properties of nanocomposites. In this study, the effect of interface energy around the unidirectional coated nanofibers on the effective dynamic effective properties is explicitly addressed by effective medium method and wave function expansion method. The multiple scattering resulting from the series coating nano-fibers is reduced to the problem of one typical nano-fiber in the effective medium. The dynamic effective shear modulus is obtained on the basis of the derived imperfect interface conditions. Analyses show that the effect of the interface properties on the dynamic effective shear modulus is significantly related to the coating layer, the nano-fiber, and the wave frequency. Due to the existence of softer coating layers, the effect of the interfaces around the layers increases greatly. Comparison with the existing results is also illustrated in the numerical examples.

Keywords: Nano composites, Interfacial strength, Effective medium method, Dynamic effective properties

1 Introduction

Since the pioneer work of Hill (1964), the prediction of the effective material properties of heterogeneous composites has been the subject of considerable scientific and engineering interests in the following years (Benveniste, 1996; Gibiansky and Torquato, 1997; Chen and Dvorak, 2006; Yuan and Zhang, 2010). In real heterogeneous materials, imperfect interfacial bonding between the matrix and the

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second-phase inhomogeneities may exist during the process of manufacture. Two models of imperfect interface, namely, the linear spring model (LSM) and the interface stress model (ISM), were widely used to simulate the discontinuities of field quantities at the interfaces and predict the overall properties of heterogeneous materials (Hashin, 1990; Tan et al., 2005; Duan et al., 2005, Gornet, Marguet, and Marckmann, 2006).

In nanocomposites, surface stress in solids is defined as a configurational force that is work-conjugate to surface strain with respect to the free surface energy. Due to the high ratio of the surface/interface area to the volume of the bulk and the fraction of energy stored in the surfaces/interfaces, the properties of the nano-systems exhibit obvious size dependent behavior. A further optimization of the interphase behavior can lead to a better overall composite performance. To illustrate the surface/interface effect, a generic continuum model incorporating surface effects was first elaborated by Gurtin and Murdoch (1975). Based on this theory, the size-dependent elastic behavior of nano-bars and nano-plate (Miller and Shenoy, 2000), nanosized inhomogeneties (Sharma et al, 2003; Duan et al, 2005), nano-filims (He et al, 2004) and spherical inhomogeneities at nano-meter length scale (Dingreville et al, 2005) were studied. All these analyses came to the same conclusion that surface elasticity around the nano-inhomogeneities plays an important role in the mechanical response of systems in the nano-meter range.

Propagation of elastic waves in composite materials is one of the key issues regarding their dynamic performance of material structures. Using this method, the dynamic effective material properties of particulate (Wei and Huang, 2006; Wei, 2007) or fiber-reinforced composite materials (Kanaun and Levin, 2003; Kim, 2003) have been investigated by numerous researchers. Compared with the conventional composites, the higher interface-to-volume ratio and the interface energy may show great effect on the propagating wave number and effective material properties of nanocomposites. Just recently, the size-dependent dynamic effective properties of unidirectional nanocomposites subjected to plane elastic waves were studied by Hasheminejad and Avazmohammadi (2009), and the interface energy effects were analyzed.

In the course of designing nanocomposites, coated fibers are often used to obtain lots of good performance of composites. In nanocomposite with coated nano-fibers, the ratio of interfaces to volume is higher, and the effective material properties are more dependent on the two interfaces around the coating layers of each nano-fiber.

The primary purpose of the current work is to study the dynamic effective properties resulting from the multiple scattering of plane elastic waves around the cylindrical coated nano-fibers. The effective medium method is used to simplify the complex scattering phenomenon. The effective propagating wave number and dynamic effective properties are obtained by solving the typical coated nano-fiber problem. The effect of surfaces/interfaces energy around the coated nano-fibers on the dynamic effective properties is analyzed in detail. This dynamic nano-model is particularly useful in understanding the physical phenomena relevant to the effect of coated fiber size and interfacial elastic properties of coating layer on the effective dynamic properties of nanostructure solids.

2 Problem formulation

Consider an unbounded matrix with a large number of coated nano-fibers, as depicted in Fig. 1, where (oxy) is the Cartesian coordinate system. The long and unidirectional nano-fibers with identical properties are randomly distributed in the matrix. The inner and outer radii are denoted as a and b, respectively. The composite medium is considered to be statistically homogeneous. Both the nano-fibers and matrix phases are assumed to be transversely isotropic, with the symmetry along the nano-fiber axis ($z$axis). The volume fraction of the nano-fibers is denoted as n_0.

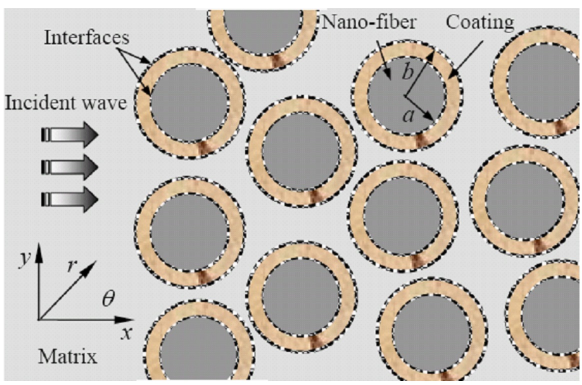

Figure 1: Geometry and coordinate systems of the problem

Let μ_f and ρ_f be the shear modulus and mass density of the fiber μ_c and ρ_c those of the coating layer, and μ_m and ρ_m those of the matrix. The transversely isotropic fibers and matrix make the overall behavior of the composite transversely isotropic, characterized by two effective elastic constants μ_* and ρ_*. An anti-plane shear wave with frequency ω propagates in the effective medium.

For nanocomposites, the effective properties are governed by the classical bulk elastic strain energy. Following the work of Gurtin and Murdoch (1975), the surface/interface region is regarded as a layer of vanishing thickness adhering to the solid without slipping. The elastic field within the bulk solid is described by the differential equations of classical elasticity, while the interface has its own elastic

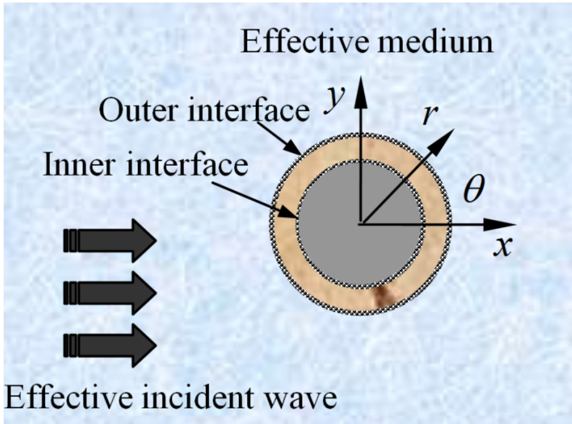

Figure 2: One typical coated nano-fiber in the effective medium

constants and is characterized by an additional constitutive law. It is assumed that the interface stresses exist at the inner interface Ω_1 and outer interface Ω_2 of the coating layer. The shear modulus and mass density of the inner interface Ω_1 are denoted as μ_{s1}, ρ_{s1}, and those of outer interface Ω_2 are μ_{s2}, ρ_{s2}.

Suppose that an anti-plane shear wave of frequency ω with polarization parallel to the nano-fibers propagates in the composite material. For this case, the wave field within the $x-y$ plane can be formulated in the framework of scalar wave propagation, and only the component of the amplitude of the displacement field in the z direction exists, i.e.,

$$u_x = u_y = 0, \quad u_z = W(x,y). \tag{1}$$

When the shear wave propagates in the matrix material, the interaction between the nano-fibers gives rise to the dispersion relations for shear waves, and the propagating wave number will change. The propagating wave number is denoted as the effective wave number. The dispersion relation can be written as (Kanaun and Levin, 2003)

$$\mu_*(k_*)k_*^2 - \rho_*(k_*)\omega^2 = 0, \tag{2}$$

where k_* is the effective wave number, $\mu_*(k_*)$ and $\rho_*(k_*)$ are the effective material properties related to the effective wave number. The relation between them can be expressed as

$$\mu_* = \mu_m(\rho_*/\rho_m)\left[Re(k_m/k_*)\right]^2, \tag{3}$$

in which $k_m = \omega\sqrt{\mu_m/\rho_m}$. In the following, it is convenient to assume that, for a small density contrast, the effective density is the mean (frequency-independent) density. So, the simple rule of mixture is used in the numerical examples, i.e.,

$$\rho_* = n_0\rho_f + (1-n_0)\bar{\rho}_m, \tag{4}$$

where $\bar{\rho}_m = c\rho_f + (1-c)\rho_c$ with $c = b/a$.

According to the hypotheses of Effective Medium Method (EMM), the interaction between many nano-fibers is reduced to a one-fiber problem (Kanaun and Levin, 2003). This problem is the diffraction of a monochromatic plane shear wave on an isolated nano-fiber embedded in the effective medium with the effective shear modulus and mass density.

3 A typical coated nano-fiber in the effective medium

When the multiple scattering among the nano-fibers is reduced to the typical coated one-fiber problem in the effective medium, the scattering of effective waves resulting from the typical nano-fiber comes into being. The wave fields around the typical nano-fiber can be described in the following forms.

a. Effective incident waves

The effective incident wave in the effective medium propagates in the x direction with frequency ω. Using wave function expansion method, it can be expressed as (Pao and Mow, 1973)

$$W^{*(in)}(r,\theta) = W_0 e^{-\mathrm{i}(k_* x - \omega t)} = W_0 \sum_{n=-\infty}^{\infty} i^n J_n(k_* r) e^{\mathrm{i}n\theta}, \tag{5}$$

where w_0 is the amplitude of the incident waves, $k_* = \omega/\sqrt{\mu_*/\rho_*}$is the effective incident wave number in the effective medium, and $J_n(\cdot)$ is the nth Bessel function of the first kind.

b. Scattered field around the typical coating nano-fiber

When the effective incident waves propagate in the effective medium with a coated nano-fiber, the scattered waves around the nano-fiber come into being. The displacement fields of the scattered waves in the effective medium can be expressed as

$$W^{*(sc)}(r,\theta) = \sum_{n=-\infty}^{\infty} a_n H_n^{(1)}(k_* r) e^{\mathrm{i}n\theta}, \tag{6}$$

where a_n are the mode coefficients of scattered waves around the typical nano-fiber, $H_n^{(1)}(\cdot)$ is the nth Hankel function of the first kind, and denotes the outgoing propagating waves.

The wave field in the effective medium is the superposition of effective incident waves and the scattered waves.

c. Wave field in the coating layer of the typical nano-fiber

The wave field in the coating layer may be described by the sum of the two components (outgoing and ingoing waves), and are expressed in the following form

$$W^{*(c)} = W_0 \left[\sum_{n=-\infty}^{\infty} b_n H_n^{(1)}(k_c r) e^{\mathrm{i}n\theta} + \sum_{n=-\infty}^{\infty} c_n H_n^{(2)}(k_c r) e^{\mathrm{i}n\theta} \right], \tag{7}$$

where $k_c = \omega / \sqrt{\mu_c / \rho_c}$, $H_n^{(2)}(\cdot)$ are the nth Hankel functions of the second kind, and denote the ingoing waves. b_n and c_n are the mode coefficients in the coating layer. It is noted that the superscript (c) denotes the wave field in the coating layer.

d. Refracted waves inside the typical nano-fiber

The refracted waves inside the typical nano-fibers are standing waves, and can be expressed as

$$W^{*(r)} = W_0 \sum_{n=-\infty}^{\infty} d_n J_n(k_f r) e^{\mathrm{i}n\theta}, \tag{8}$$

where $k_f = \omega / \sqrt{\mu_f / \rho_f}$, the superscript (r) stands for the refracted waves, and d_n are the mode coefficients of refracted waves.

4 Boundary conditions around the typical coated nano-fiber

Following the work of Gurtin and Murdoch (1975), and considering the anti-plane nature in this study, the boundary conditions at the inner interface Ω_1 and outer interface Ω_2 of the coating layer are expresses as follows.

a. At the inner interface, i.e., $r = a$

$$W^{*(r)} = W^{*(c)}, \tag{9}$$

$$\tau_{rz}^{*(r)} - \tau_{rz}^{*(c)} = (\mu_{s1} - \tau_{01}) \frac{\partial \tau_{\theta z}^{*(c)}}{a \partial \theta}, \tag{10}$$

b. At the outer interface, i.e., $r = b$

$$W^{*(c)} = W^{*(m)}, \tag{11}$$

$$\tau_{rz}^{*(c)} - \tau_{rz}^{*(m)} = (\mu_{s2} - \tau_{02}) \frac{\partial \tau_{\theta z}^{*(m)}}{b \partial \theta}. \tag{12}$$

It should be noted that the deformations resulting from the residual surface stresses τ_{01} and τ_{02} are always independent of the external loading, and it is assumed that the two residual surface stresses are zero, i.e., $\tau_{01} = \tau_{02} = 0$.

Substituting Eqs.(5-8) into Eqs.(9-12), and making use of the orthogonality relation of $e^{\mathrm{i}n\theta}$, a set of algebraic equations can be obtained

$$a_n H_n^{(1)}(k_* b) - b_n H_n^{(1)}(k_c b) - c_n H_n^{(2)}(k_c b) = -W_0 i^n J_n(k_* b), \tag{13}$$

$$\begin{aligned} & a_n \left\{ \mu_* \left[n H_n^{(1)}(k_* b) - k_* b H_{n+1}^{(1)}(k_* b) \right] - \frac{n^2 \mu_{1s}}{b} H_n^{(1)}(k_* b) \right\} \\ & - b_n \left\{ \mu_c \left[n H_n^{(1)}(k_c b) - k_c b H_{n+1}^{(1)}(k_c b) \right] \right\} \\ & - c_n \left\{ \mu_c \left[n H_n^{(2)}(k_c b) - k_c b H_{n+1}^{(2)}(k_c b) \right] \right\} \\ & = -W_0 i^n \left\{ \mu_* \left[n J_n(k_* b) - k_* b J_{n+1}(k_* b) \right] - \frac{n^2 \mu_{1s}}{b} J_n(k_* b) \right\}, \end{aligned} \tag{14}$$

$$b_n H_n^{(1)}(k_c a) + c_n H_n^{(2)}(k_c a) - d_n J_n(k_f a) = 0, \tag{15}$$

$$\begin{aligned} & b_n \left\{ \mu_c \left[n H_n^{(1)}(k_c a) - k_c a H_{n+1}^{(1)}(k_c a) \right] - \frac{n^2 \mu_{s2}}{a} H_n^{(1)}(k_c a) \right\} \\ & + c_n \left\{ \mu_c \left[n H_n^{(2)}(k_c a) - k_c a H_{n+1}^{(2)}(k_c a) \right] - \frac{n^2 \mu_{s2}}{a} H_n^{(2)}(k_c a) \right\} \\ & - d_n \left\{ \mu_f \left[n J_n(k_f a) - k_f a J_{n+1}(k_f a) \right] \right\} = 0, \end{aligned} \tag{16}$$

From the above equations, the mode coefficients can be solved.

5 Effective propagating wave number

Once the scattered field due to a single coated nano-fiber is known, the phase velocities and attenuations of the effective propagating waves in the composite can be easily calculated by means of an iterative process based on the following formula

$$\left(k_{SH}^{(j+1)} / k_{SH}^{(j)} \right)^2 = \left[1 + 2\pi n_0 f^{(j)}(0) / \left(k_{SH}^{(j)} \right)^2 \right]^2 - \left[2\pi n_0 f^{(j)}(\pi) / \left(k_{SH}^{(j)} \right)^2 \right]^2, \tag{17}$$

where

$$f^{(j)}(0) = \sum_{n=-\infty}^{\infty} (-i)^n a_n^{(j)}, \quad f^{(j)}(\pi) = \sum_{n=-\infty}^{\infty} i^n a_{-n}^{(j)}. \tag{18}$$

It is noted that $f^{(j)}(0)$ is the forward scattering amplitude of the typical nano-fiber, and$f^{(j)}(\pi)$ is the backward scattering amplitude of the typical nano-fiber

The iteration is started by taking effective properties $k_{SH}^{(0)}$ with $k_{SH}^{(0)} = k_{SH}^{(m)}$. Then, the forward and backward scattered amplitudes are calculated. Next, the obtained effective wave numbers are used as the new effective wave numbers in the above equation and these procedures are repeated until the convergence is obtained. Consequently, the wave number equations take the final (converged) form

$$1 = \left[1 + 2\pi n_0 f^*(0) / (k_{SH}^*)^2\right]^2 - \left[2\pi n_0 f^*(\pi) / (k_{SH}^*)^2\right]^2. \tag{19}$$

6 Numerical examples and analysis

Specially designed coating layer and the surfaces/interfaces can improve the dynamic effective properties of nanocomposites. In order to illustrate the surfaces/ interfaces effect at the coating layer on the dynamic elastic modulus, some numerical examples are given in this section.

Figure 3 illustrates the interface effect on the dynamic effective properties in the region of low frequency. In Fig. 3, the nano-fiber is stiffer than the matrix. It can be seen that if the dimension of the fiber greater than 4.0nm, the effective shear modulus nearly shows no variation with surface/interface properties. The existence of surface/interface results in the increase of effective shear modulus. The smaller the nano-fiber is, the greater the surface/ interface effect is. The interface effect increases greatly with the decrease of the radius of the nano-fiber. These results are consistent with those in Hasheminejad and Avazmohammadi (2009).

Figure 4 illustrates the interface effect around the coating layer on the dynamic effective properties in the region of low frequency. In Fig. 4, the nano-fiber is stiffer than the matrix. It can be seen that due to the existence of coating layer, the dimension of the fiber shows less effect on the effective shear modulus. Compared with the results in Fig. 3, it is clear that the surface/interface effect on the dynamic effective properties decreases when the coating layer is softer than the fiber and matrix. The effect of the outer surface/interface is greater than that of the inner surface/interface.

Figure 5 illustrates the interface effect around the coating layer on the dynamic effective properties in the region of low frequency. In Fig. 5, the nano-fiber is softer than the matrix. By comparing with the results in Fig. 4, it is clear that the dimension of the nano-fiber shows greater effect on the dynamic effective shear modulus. In this case, the effect of the interfaces around the coating layer on the dynamic effective shear modulus also increases.

Figure 6 illustrates the interface effect around the coating layer on the dynamic

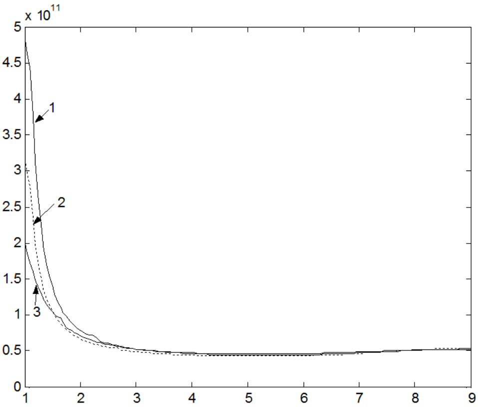

Figure 3: Dynamic effective properties without the effect of coating layer $k = 0.5, \mu_m = 34.7 \times 10^9 N/m^2, \mu_f = 69.1 \times 10^9 N/m^2, \rho_m = 2.7 \times 10^3 kg/m^3, b/a = 1.0, n_0 = 0.2$
$1.\mu_{s1} = 52 \times 10^9 N/m^2; 2.\mu_{s1} = 25 \times 10^9 N/m^2; 3.\mu_{s1} = 0$

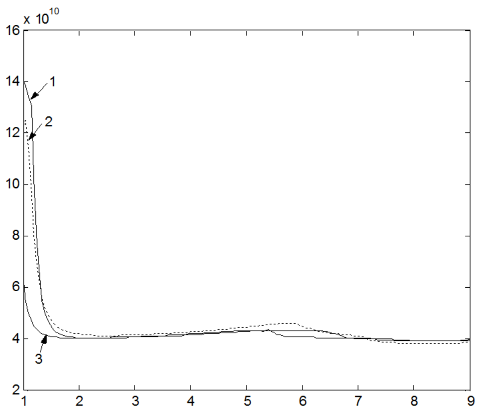

Figure 4: Dynamic effective properties without the effect of coating layer $k = 0.5, \mu_m = 34.7 \times 10^9 N/m^2, \mu_f = 69.1 \times 10^9 N/m^2, \mu_c = 12 \times 10^9 N/m^2, \rho_m = 2.7 \times 10^3 kg/m^3, b/a = 1.2, n_0 = 0.2$
$1.\mu_{s1} = 52 \times 10^9 N/m^2, \mu_{s2} = 52 \times 10^9 N/m^2; 2.\mu_{s1} = 52 \times 10^9 N/m^2, \mu_{s2} = 0; 3.\mu_{s1} = 0, \mu_{s2} = 52 \times 10^9 N/m^2;$

effective properties in the region of high frequency. In Fig. 6, the nano-fiber and the coating layer are both stiffer than the matrix. By comparing with the results in

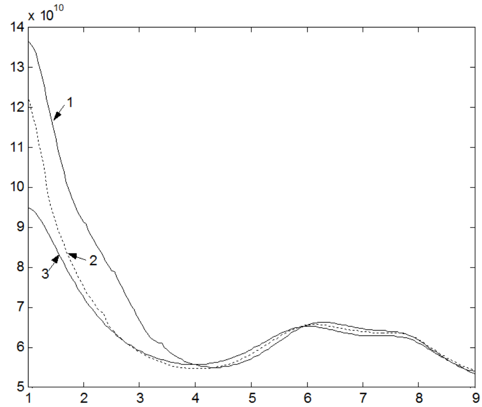

Figure 5: Dynamic effective properties without the effect of coating layer $k = 0.5, \mu_m = 34.7 \times 10^9 N/m^2, \mu_f = 17.3 \times 10^9 N/m^2, \mu_c = 12 \times 10^9 N/m^2, \rho_m = \rho_c = \rho_f = 2.7 \times 10^3 kg/m^3, b/a = 1.2, n_0 = 0.2$
$1.\mu_{s1} = 52 \times 10^9 N/m^2, \mu_{s2} = 52 \times 10^9 N/m^2; 2.\mu_{s1} = 52 \times 10^9 N/m^2, \mu_{s2} = 0; 3.\mu_{s1} = 0, \mu_{s2} = 52 \times 10^9 N/m^2$

Fig. 4, it can be seen that only when the nano-fiber is small than 2.5nm, the surface/interface shows effect on the dynamic effective properties. However, the surfaces/interfaces effects around the coating layer increase with the wave frequency.

In Fig. 7, the coating layer is softer than the matrix, and the nano-fiber is stiffer than the matrix. By comparing with the results in Fig. 6, it can be seen that the dimension of the nano-fibers increases due to the existence of the softer coating layer. When the coating layer becomes softer, the outer surface shows greater effect on the dynamic effective shear modulus.

7 Conclusion

In this study, the effective medium method and the surface/interface model of Gurtin and Murdoch have been adopted to investigate the dynamic effective properties of nanocomposites with coating nano-fibers. The effect of interfacial properties around the coating layers on the dynamic effective shear modulus under different parameters is analyzed in detail. It has been found that the effect of surface/interface elasticity on the dynamic effective shear modulus is significantly related to the coating layer, the nano-fiber, and the wave frequency. The main findings of this work are as follows:

a. Due to the existence of softer coating layers, the dimension of the fiber shows

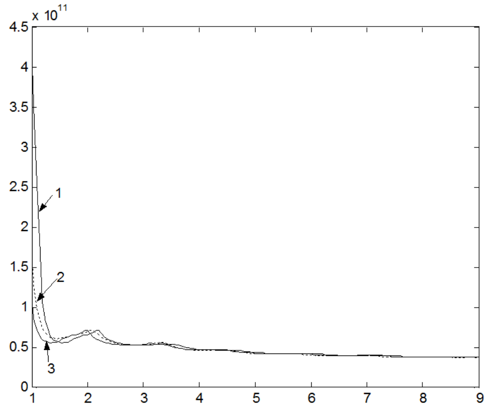

Figure 6: Dynamic effective properties without the effect of coating layer $k = 1.5, \mu_m = 34.7 \times 10^9 N/m^2, \mu_f = 17.3 \times 10^9 N/m^2, \mu_c = 48 \times 10^9 N/m^2, \rho_m = \rho_c = \rho_f = 2.7 \times 10^3 kg/m^3, b/a = 1.2, n_0 = 0.2$
$1.\mu_{s1} = 52 \times 10^9 N/m^2, \mu_{s2} = 52 \times 10^9 N/m^2; 2.\mu_{s1} = 52 \times 10^9 N/m^2, \mu_{s2} = 0; 3.\mu_{s1} = 0, \mu_{s2} = 52 \times 10^9 N/m^2$

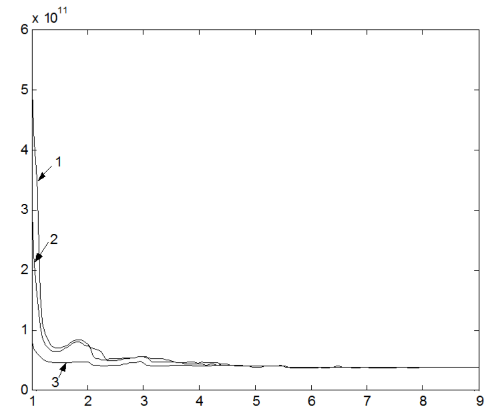

Figure 7: Dynamic effective properties without the effect of coating layer $k = 1.5, \mu_m = 34.7 \times 10^9 N/m^2, \mu_f = 17.3 \times 10^9 N/m^2, \mu_c = 12 \times 10^9 N/m^2, \rho_m = \rho_c = \rho_f = 2.7 \times 10^3 kg/m^3, b/a = 1.2, n_0 = 0.2$
$1.\mu_{s1} = 52 \times 10^9 N/m^2, \mu_{s2} = 52 \times 10^9 N/m^2; 2.\mu_{s1} = 52 \times 10^9 N/m^2, \mu_{s2} = 0; 3.\mu_{s1} = 0, \mu_{s2} = 52 \times 10^9 N/m^2$

less effect on the effective shear modulus in the region of low frequency.

b. If the coating layer is softer than the fiber and matrix, the surface/interface effect on the dynamic effective properties decreases. The effect of the outer surface/interface is greater than that of the inner surface/interface.

c. If the nano-fiber is softer than the matrix, the effect of the dimension of the nano-fiber on the dynamic effective shear modulus is greater, and the effect of the interfaces around the coating layer on the dynamic effective shear modulus also increases.

d. In the region of high frequency, the dimension of the nano-fiber shows less effect on the dynamic effective shear modulus; however, the effect of the interfaces around the coating layer increases.

The conclusion is of practical interest in the ultrasonic characterization and nondestructive evaluation of nanocomposites with coating nano-fibers.

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