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Micromechanics model for predicting effective elastic moduli of porous ceramic matrices with randomly oriented carbon nanotube reinforcements

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Multi-step micromechanics-based models are developed to predict the overall effective elastic moduli of porous ceramic with randomly oriented carbon nanotube (CNT) reinforcements. The presence of porosity in the ceramic matrix that has been previously neglected in the literature is considered in present analysis. The ceramic matrix with porosity is first homogenized using a classical Mori-Tanaka model. Then, the homogenized porous ceramic matrix with randomly oriented CNTs is analysed using two micromechanics models. The results predicted by the present models are compared with experimental and analytical results that have been reported in literature. The comparison shows that the discrepancies between the present analytical results and experimental data are about 10% for 4 wt% of CNTs and about 0.5% for 8 wt% CNTs, both substantially lower than the discrepancies currently reported in the literature. [http://dx.doi.org/10.1063/1.4931453]

I. INTRODUCTION

There has been a growing interest in utilizing CNTs as nanoscale structural reinforcement in the ceramic matrix.[1–4] However reports show only moderate improvements of elastic moduli in the CNT reinforced composites[5–7] and ceramic nanocomposites.[8,9] Several researchers have attributed this disparity to the waviness and agglomeration of CNTs and also the weakened interface and partial debonding between the CNTs and the matrix. Fisher *et al.*[10] have investigated the effects of CNT waviness and 2D/3D random orientation on the effective elastic modulus by using finite element method to simplify the wavy CNT as straight CNT to be integrated into the Mori-Tanaka model. Anumandla and Gibson[11] have proposed a similar micromechanical model with the use of representative volume element (RVE) to account for the waviness of CNTs and random orientation of the nanotubes. Furthermore, Shi *et al.*[12] have examined the effects of waviness and agglomeration of CNTs on effective stiffness of CNT-reinforced composites. To analyze the effect of agglomeration and interface condition on the plasticity of CNT-metal composites, Barai and Weng[13] developed a two scale micromechanical model. This method has considered randomly oriented and transversely isotropic CNTs.

Using a modified Eshelby model,[14] Chen*et al.*[8] studied plasma sprayed aluminum oxide coating reinforced CNT. The longitudinal elastic modulus was predicted for aligned CNT reinforced aluminum oxide nanocomposites and porous aluminum oxide. The study compared the experimental results of plasma sprayed aluminum oxide reinforced CNT nanocomposite with computed results. A conclusion was that the modified Eshelby tensor model[14] was able to predict the elastic modulus of CNT reinforced nanocomposite. However, the research did not consider presence of

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porosity in the matrix even if porosities have been found to be present in the ceramic. The orientation of the CNT was also not considered in their model.

II. POROSITY IN THE CERAMIC MATRIX

In this paper, two multi-step micromechanics-based models are to be developed to examine the effects of randomly oriented CNTs and matrix porosity on the overall elastic properties of ceramic nanocomposites. The presence of porosity in the ceramic matrices has been neglected in the studies so far, including those by Chen *et al.*[8] The results of the present models are then compared with the experimental and analytical results reported in Chen *et al.*[8] to be able to validate the accuracy of the present model.

Porosity is always measured in relation to the theoretical density of composite even though porosity exists only in the matrix and not the CNTs. Therefore when examining the effect of porosity in overall elastic modulus of a nanocomposite, it has to be re-evaluated as porosity in the matrix. This will result in lower effective elastic moduli of the matrix and also a change in the volume fraction between the CNTs and porous matrix.

The density of composite is given as,[15]

$$\rho_c = \rho_f v_f + \rho_m v_m \tag{1}$$

where ρ_c, ρ_f and ρ_m are the density of the composite, fiber and matrix, respectively. v_f and v_m are the volume fraction of fiber and matrix. By considering the presence of porosity in the matrix, the density a composite with porosity can be written as,

$$RD_c \rho_c = \rho_f v_f + RD_m \rho_m v_m \tag{2}$$

where RD_c and RD_m are the relative densities of the composite and the matrix, respectively. By substituting Eq. (1) into (2) and making RD_m the subject, the relative density of the matrix can be written as,

$$RD_m = \frac{RD_c \left(\rho_f v_f + \rho_m v_m\right) - \rho_f v_f}{\rho_m v_m} \tag{3}$$

Density of porous matrix, $\rho_{m(\phi)}$, can be calculated using the following equation,

$$\rho_{m(\phi)} = RD_m \rho_m \tag{4}$$

The volume fraction of fiber in a composite is given by,[15]

$$v_f = \frac{1}{1 + \frac{\rho_f}{\rho_m}\left(\frac{1}{w_f} - 1\right)} \tag{5}$$

where w_f is the weight fraction of the fiber. Based on Eq. (4) and (5), the volume fraction of fiber in relation to porous matrix can be defined as,

$$v_{f(\phi)} = \frac{1}{1 + \frac{\rho_f}{\rho_{m(\phi)}}\left(\frac{1}{w_f} - 1\right)} \tag{6}$$

With the parameters determined in Eq. (3) and (6), the effective elastic moduli of a porous matrix with randomly oriented CNT-reinforcement can be predicted by using a multi-step micromechanics model, which will be shown in the succeeding section.

III. MULTI-STEP MICROMECHANICS MODEL

Two multi-step micromechanics-based models are developed to evaluate the effective elastic moduli of porous matrix with randomly oriented CNT-reinforcement, as shown in FIG.1(a). In the first step shown in FIG.1(b), the classical Mori-Tanaka method is used to evaluate the effective

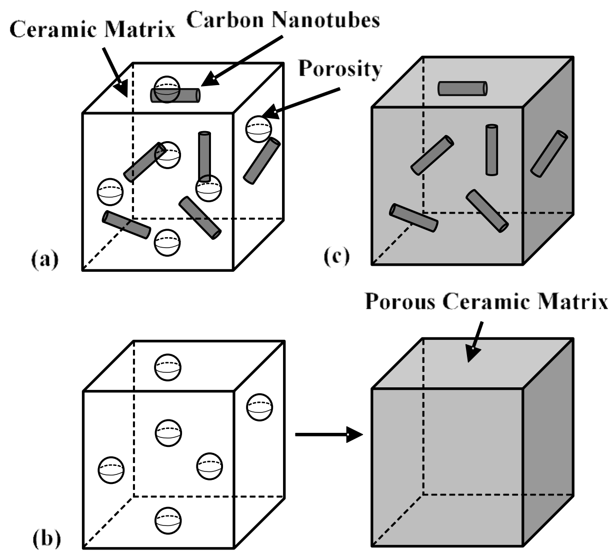

FIG. 1. Multi-step micromechanical model. a) Schematic of ceramic matrix with porosity and CNT inclusions. b) Transformation of ceramic matrix with spherical void inclusions into porous ceramic. c) Porous ceramic matrix with randomly oriented CNT inclusions.

elastic modulus of porous ceramic by considering ceramic matrix with spherical void inclusions. The details for this method can be found in elsewhere[16,17] and will not be presented here for the sake of brevity.

To evaluate the overall elastic properties of porous ceramic matrix with randomly oriented CNT-reinforcement, we assume the porous ceramic matrix to be homogenous and isotropic. In the second step shown in FIG. 1(c), the Mori-Tanaka method is again used to evaluate the effective elastic modulus of porous ceramic with CNT-reinforcement by considering the porous ceramic matrix with randomly oriented CNT inclusions.

Both micromechanics models in this study use the Mori–Tanaka mean-field approach. The first model uses the Mori-Tanaka method with orientation averaging by Huang[18] and the second model uses a Mori-Tanaka method presented by Tandon and Weng.[19]

In the first model (Model 1), the overall elastic stiffness tensor of a porous ceramic is given by,[16]

$$C^{m(\phi)} = C^{m} + \phi_m \left\{ \left(C^{\phi} - C^{m}\right) A^{dil(\phi)} \right\} \left((1-\phi_m)\,\mathbf{I} + \phi_m \left\{ A^{dil(\phi)} \right\} \right) \tag{7}$$

where $\mathbf{I}$ is the identity tensor, C^{ϕ} and C^{m} are the stiffness tensor of the pore and matrix. The amount of porosity in the matrix can be shown as $\phi_m = (1 - RD_m)$. The elastic modulus of pores is zero where $E^{\phi} = 0$ therefore $C^{\phi} = 0$. The bracket pair { } represents the average value of the term over all possible orientation. The shape of the void inclusion considered is spherical hence there is no need to consider the orientation of the inclusion. The dilute strain concentration tensor $A^{dil(\phi)}$ for pore inclusions is given by,

$$A^{dil(\phi)} = \left[\mathbf{I} + \mathrm{S}^{s}\left(C^{\phi}\right)^{-1}\left(C^{m} - C^{\phi}\right)\right]^{-1} \tag{8}$$

where S^{s} is the Eshelby tensor and its components can be found in Mura.[20] The overall elastic stiffness tensor of porous ceramic matrix with randomly oriented CNT-reinforcement is given by,[16]

$$C^{c(\phi)} = C^{m(\phi)} + v_{f(\phi)} \left\{ \left(C^{f} - C^{m(\phi)}\right) A^{dil(f)} \right\} \left(\left(1 - v_{f(\phi)}\right)\mathbf{I} + v_{f(\phi)} \left\{ A^{dil(f)} \right\} \right) \tag{9}$$

where C^{f}, $C^{m(\phi)}$ and $C^{c(\phi)}$ are the stiffness tensor of the fiber, porous matrix and composite with porous matrix, respectively, and $v_{f(\phi)}$ is found in Eq. (6).

The dilute strain concentration tensor $A^{dil(\phi)}$ for fiber inclusions is given by,

$$A^{dil(f)} = \left[\mathbf{I} + S^f (C^f)^{-1} \left(C^{m(\phi)} - C^f\right)\right]^{-1} \tag{10}$$

where S^f is the Eshelby tensor for shape of fiber. In this case the shape considered for the fiber inclusions are prolate and cylindrical. Prolate ellipsoid is defined by aspect ratio and cylindrical shape is universal and the equivalent Eshelby tensor for the shapes considered can be found in Mura.[20] Since the CNTs are randomly oriented, orientation averaging has to be taken into account. Based on Huang[18] the orientational average is given as

$$\{A^{dil}\} = \frac{1}{4\pi} \int_{-\pi}^{\pi} \int_{0}^{\pi} A^{dil}_{ijkl}(\theta_1, \theta_2) \sin\theta_2 d\theta_2 d\theta_1 \tag{11}$$

where the transformation of the tensor from local to global coordinate is written as

$$A^{dil}_{ijkl} = \alpha_{ip}\alpha_{jq}\alpha_{kr}\alpha_{ls}A^{dil}_{pqrs} \tag{12}$$

The coordinate transformation matrix can be found in Huang.[18] As the transformation in Eq. (12) is a tensor transformation, in order for transformation to be valid, specific tensors expressed using contracted notation must first be converted to their appropriate tensorial components.

In the second model (Model 2), the effective elastic modulus of the composite with randomly oriented inclusions is evaluated from a general equation with the bulk and shear modulus given as

$$E = \frac{9k\mu}{3k + \mu} \tag{13}$$

The effective bulk and shear moduli of the composite are given by:

$$\frac{k}{k_0} = \frac{1}{1 + cp} \tag{14}$$

$$\frac{\mu}{\mu_0} = \frac{1}{1 + cq} \tag{15}$$

where c is the volume concentration/fraction, k_0 and μ_0 are the bulk and shear modulus of the matrix, k and μ are the bulk and shear modulus of the composite.

$$p = \frac{p_2}{p_1} \tag{16}$$

$$q = \frac{q_2}{q_1} \tag{17}$$

where p_1, p_2, q_1 and q_2 are explicit expressions and these expressions are different for void inclusions and fiber inclusions. Details of the expression can be found in Zhao *et al.*[17] for void inclusions and Tandon and Weng[19] for fiber inclusions. The explicit expressions requires the bulk and shear modulus of the matrix and inclusions to evaluate the effective bulk and shear moduli of a porous matrix or composite.

IV. RESULTS AND DISCUSSIONS

Chen *et al.*[8] presented experimental data of plasma sprayed Al_2O_3-CNT nanocomposite and two micromechanical model results that used aligned CNT and aligned pores with exact dimensions as the CNTs. In this paper, two multi-step micromechanical models are employed in this paper to predict the overall elastic moduli of Al_2O_3-CNT nanocomposite using the parameters used in Chen *et al.*[8] The recorded relative density in the study[8] for 4 and 8 wt% CNT reinforced aluminum oxide nanocomposite are 90.2% and 93.9%, respectively. The elastic modulus, density and Poisson's ratio of aluminum oxide and CNT that are used in the present study are $E_{AL_2O_3} = 390$ GPa, $E_{CNT} = 600 - 1000$ GPa, $\rho_{AL_2O_3} = 3.96$ g/cm^3, $\rho_{CNT} = 2.25$ g/cm^3, $\upsilon_{AL_2O_3} = 0.22$ and $\upsilon_{CNT} = 0.18$.

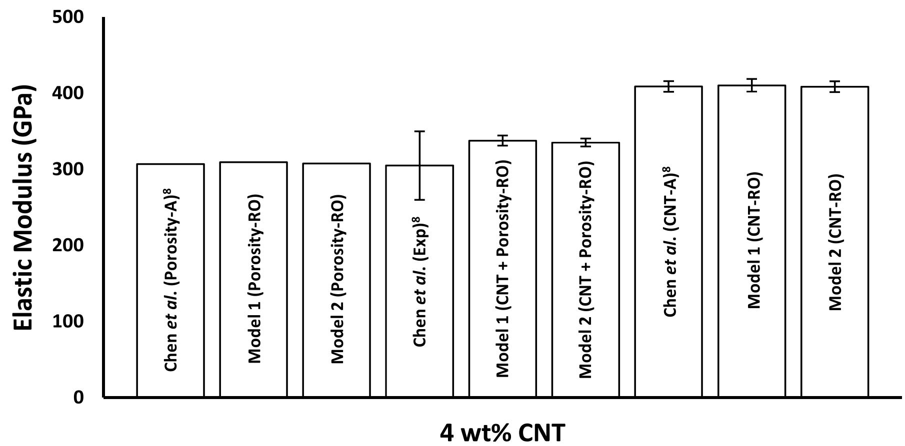

FIG. 2. Comparison between Chen *et al.*[8] analytical and experimental results with the results of the present models for 4 wt% of CNT. (A: aligned and RO: randomly oriented).

The shape of CNT in the study by Chen *et al.*[8] is based on a modified Eshelby tensor by Paul and Lee[14] which are cylindrical inclusion in two different sizes of CNT, R_{CNT} = 40 nm, L_{CNT} = 500 nm and R_{CNT} = 70 nm and L_{CNT} = 2000 nm where R is diameter and L is length. Whereas, the CNT shapes considered here are prolate and cylindrical.

The two micromechanics-based models each presents three results comprising of: (a) ceramic matrix with randomly oriented CNT, (b) ceramic matrix with randomly oriented pores with exact dimensions as the CNT inclusions, and (c) porous ceramic matrix with randomly oriented CNTs reinforcements. The validities of the Model 1 and 2 are tested by comparing results of ceramic matrix with randomly oriented CNT and ceramic matrix with randomly oriented pores with exact dimensions as the CNT inclusions. Results from the present analytical models are in good agreement with each other and the model used in Chen *et al.*[8] as shown in FIG. 2 and FIG. 3.

Using Eq. (3), (5) and (6), volume fraction of CNT in ceramic matrix, relative density of ceramic matrix and volume fraction of CNT in porous matrix for 4 wt% of CNT are determined as $v_f \approx 0.0683$, $RD_m \approx 89.79\%$ and $v_{f(\phi)} \approx 0.0618$. As observed in FIG. 2 and Table I, the results obtained from the two models presented here are in good agreement with the analytical and experimental results presented by Chen *et al.*[8] The difference between the experimental and computed elastic modulus for porous matrix with CNT-reinforcement is ~10.7% and ~9.9% for Models 1 and 2, respectively. The lower limit of the experimental results (260 GPa) are well below the

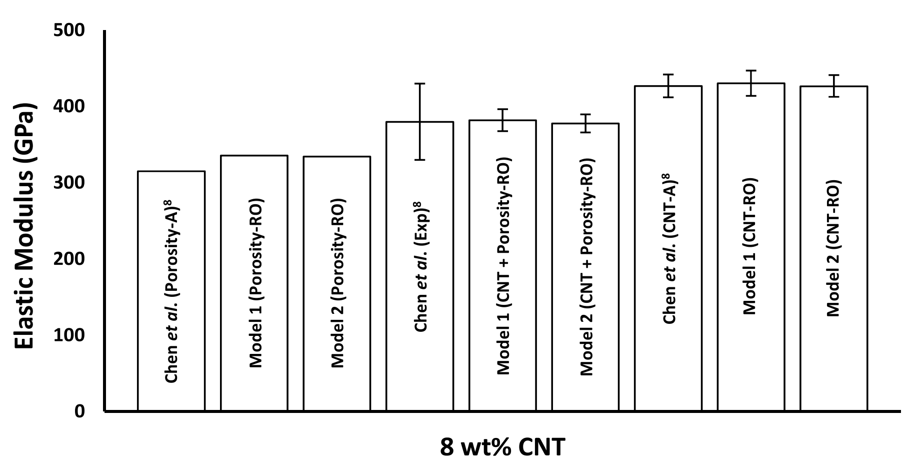

FIG. 3. Comparison between Chen *et al.*[8] computed and experimental results with present models for 8 wt% of CNT. (A: aligned and RO: randomly oriented).

TABLE I. Effective elastic moduli of analytical and experimental results.

	Chen *et al.*			Present non-porous matrix				Present porous matrix	
				Model 1		Model 2		Model 1	Model 2
Weightage of CNT	CNT	Pores	Exp.	CNT	Pores	CNT	Pores	CNT	CNT
E (GPa) 4 wt%	409	307	260-350 (305)	410	310	408	308	338	335
E (GPa) 8 wt%	427	315	330-430 (380)	430	336	427	334	382	378

analytical results of ceramic matrix with porosity and the upper limit of the experimental results (350 GPa) are well below the analytical results of ceramic matrix with fully bonded CNT inclusions as well. Chen *et al.*[8] concluded that the low CNT content in the 4 wt% nanocomposite causes more splat sliding to occur and resulted in lower experimental elastic modulus.

Using Eq. (3), (5) and (6), volume fraction of CNT in ceramic matrix, relative density of ceramic matrix and volume fraction of CNT in porous matrix for 8 wt% of CNT are determined as $v_f \approx 0.1327$, $RD_m \approx 93.37\%$ and $v_{f(\phi)} \approx 0.125$. As observed in FIG. 3 and Table I, the analytical results from the two models for porous matrix with CNT-reinforcement is in good agreement with the experimental result present by Chen *et al.*[8] The difference between the experimental and computed elastic modulus for porous matrix with CNT-reinforcement is ~0.5% and ~0.6% for Models 1 and 2, respectively. The lower limit and upper limit of the experimental results (330 to 430 GPa), this is in good agreement with analytical results of ceramic matrix with porosity and fully bonded CNT inclusions.

V. CONCLUSIONS

In summary, two multi-step micromechanics models have been developed to take into account the presence of porosity in the ceramic matrix as well as orientation of CNTs. These models can be used to predict the effective elastic properties of ceramic matrices with CNT-reinforcement. On average Model 1 predicts effective elastic moduli at 0.7 percentage point higher than Model 2 among the three results. Both models show improved agreements with the experimental and analytical results over the existing model reported by Chen *et al.*[8] The results confirmed that it is important to consider the presence of porosity in the ceramic matrix. Future works can look at effective ranges of the porosity and volume fraction of CNT in the porous ceramic matrix with CNT-reinforcement on the multi-step micromechanics model which would require quantitative experimental data.

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