PARTICLE-TO-PARTICLE INTERACTIONS IN SYNTACTIC FOAMS

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Summary

This work studies the elastic interaction between a pair of hollow particles embedded in a dissimilar medium and subjected to a remote uniaxial tensile loading. The Boussinesq-Papkovich stress function approach is integrated with a multi-pole series expansion to establish a tractable semi-analytical solution of the Navier-Cauchy equation. Results specialized to glass-vinyl ester syntactic foams show that neglecting interactions among particles in the modeling scheme generally result into underestimation of the stiffness and overestimation of the strength. Moreoever, it is found that particle wall thickness can be used to control the intensity of particle-to-particle interactions and their effects on the response of the composite.

1 Introduction

Hollow particle filled composites are a special class of closed cell foams [1] where porosity appears in the form of air enclosed inside thin shells that are embedded in a matrix material. Their porous microstructure is used in marine applications to achieve low density [2], low moisture absorption [3], and high damage tolerance [4]. These systems are generally referred to as syntactic foams and a large spectrum of material compositions is explored in the technical literature, including metal and polymer matrix foams filled with carbon and glass inclusions [5].

Particle wall thickness and volume fraction can be jointly used to tailor the mechanical properties of syntactic foams [6]. Modeling efforts generally assume particles to be of same size and wall thickness and neglect particle-to-particle interactions. However, commercially available microballoons show significant polydispersions in diameter and wall thickness [7] and syntactic foams are generally synthesized with particle volume fractions in the range of $30-60\%$ where particle-to-particle interactions are expected to be important. In [8], an analytical treatment of the effect of particle polydispersivity on syntactic foam elastic properties is presented by using a differential scheme [9]; however, particle-to-particle interactions are therein neglected.

The interaction between two spherical elastic regions in an infinite medium is originally addressed in [10], where a multi-pole expansion technique is developed to analyze the interactions between two cavities. This approach is extended to analyze the interaction between two solid particles for axisymmetric loading conditions in [11]. In [12] and [13], a micromechanics-based elastic model is developed for two-phase functionally graded materials. Locally pair-wise interactions are taken into account by extending the Eshelby's equivalent inclusion method to the case of two equal spherical solid particles embedded in an infinite matrix domain. These studies are not directly applicable to syntactic foams as they focus on homogeneous dispersions of solid inclusions.

In this work, the multi-pole expansion technique presented in [11] is adapted to study the interactions between two hollow particles embedded in an elastic medium that is subjected to remote uniaxial tensile loading. A semi-analytical formulation for the stress and displacement fields is obtained by using the Boussinesq-Papkovich stress function approach. By applying suitable continuity conditions at the particle-matrix interfaces, the problem reduces to an algebraic linear system, whose dimension depends on the targeted solution accuracy. Interfacial stress fields are correlated to the overall elastic properties of the composite by using a generalization of the Eshelby's formula [14]. A parametric study is performed to describe the role

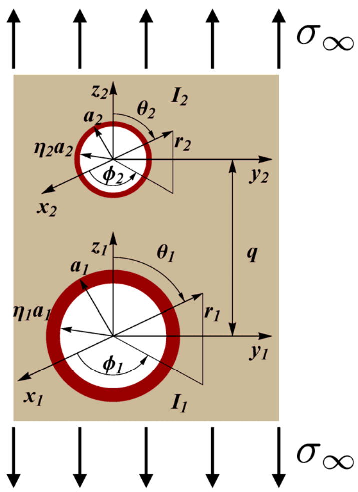

Fig. 1. Schematic of the problem.

of particle wall thicknesses and relative inter-particle distances on effective elastic behavior of the composite. Results are specialized to glass-vinyl ester syntactic foams.

2 Problem statement

A schematic of the problem is illustrated in Fig. 1. The problem consists of two hollow spherical particles, S_1 and S_2, embedded in a matrix under a remote uniaxial loading σ_∞ along the $z = z_1 = z_2$-axis. Two spherical coordinates systems (r_1, θ_1, ϕ_1) and (r_2, θ_2, ϕ_2) are selected to identify displacement and stress fields with respect to each particle center. The particle and the matrix materials are assumed to be isotropic, linear elastic, and homogeneous. Particle-matrix interfaces are assumed perfect and particles are aligned with the loading direction at a distance q between their centers. Dimensionless radial coordinate $\tilde{r}_i = r/q$ are used when possible. The particles have in general different outer radius and wall thickness; the outer radius of the i-th particle is referred to as a_i and η_i defines its radius ratio, that is, the ratio between the inner and the outer radii. The ratio of the particle radius a_i to the distance q is labeled as $\tilde{a}_i$ for $i = 1, 2$. The parameter $\Gamma = q/(a_1 + a_2) = 1/(\tilde{a}_1 + \tilde{a}_2)$ is used to describe the inter-particle distance $q - (a_1 + a_2)$, while the ratio between the outer radii of the two particles is called A. Note that particles tend to be closer as Γ decreases.

Particle geometry and loading conditions allow for reducing the three-dimensional problem in Fig. 1 to a two-dimensional $(2D)$ scenario using axisymmetry. In what follows, subscripts m and p and superscripts m, S_1, and S_2, are used to identify properties, stress fields, and displacements fields of the matrix and particle materials, respectively.

3 Method of solution

By following the work in [11], the solution of the Navier-Cauchy equation set for the stress and displacement fields within the matrix and the particles are solved in the matrix and particle materials in terms of Legendre polynomials and their derivatives. The general solution form is here omitted for brevity. Notably, fields within the matrix region require the knowledge of four sequences of scalar numbers along with the values of the remote fields. In addition, the determination of fields within each particle is controlled by four other sequences of scalar numbers. Thus, a total of twelve sequences of real numbers are needed to thoroughly describe the stress and displacement fields within the considered syntactic foams.

The remote stress and displacement fields scaled with respect to q are conveniently written in terms of solid spherical harmonics, that is,

$$\sigma_{rr}^{\infty}(\tilde{r}_i, \theta_i) = \sum_{n=0}^{\infty} \sigma_n^i P_n(\cos\theta_i), \quad \sigma_{r\theta}^{\infty}(\tilde{r}_i, \theta_i) = \sin\theta_i \sum_{n=0}^{\infty} \tau_n^i P_n'(\cos\theta_i) \tag{1a}$$

$$\tilde{u}_r^{\infty}(\tilde{r}_i, \theta_i) = \sum_{n=0}^{\infty} \varrho_n^i P_n(\cos\theta_i), \quad \tilde{u}_\theta^{\infty}(\tilde{r}_i, \theta_i) = \sin\theta_i \sum_{n=0}^{\infty} \epsilon_n^i P_n'(\cos\theta_i) \tag{1b}$$

Here, σ_n^i, τ_n^i, ϱ_n^i, and ϵ_n^i are known coefficients, and the notation refers to the spherical coordinate systems defined in Fig. 1, where the index i varies between 1 and 2 to span the two particles. For the considered uniaxial loading along the z-axis, the only non-zero coefficients in equation set (1) are

$$\sigma_0^i = \frac{\sigma_\infty}{3}, \sigma_2^i = \frac{2}{3}\sigma_\infty, \tau_2^i = -\frac{\sigma_\infty}{3} \tag{2a}$$

$$\varrho_0^i = \frac{\tilde{r}_i(1-2\nu_m)}{6\mu_m(1+\nu_m)}\sigma_\infty, \varrho_2^i = \frac{\tilde{r}_i}{3\mu_m}\sigma_\infty, \epsilon_2^i = -\frac{\tilde{r}_i}{6\mu_m}\sigma_\infty \tag{2b}$$

where μ_m and μ_p are the shear moduli of the matrix and particle materials, respectively, and, ν_m and ν_p identify their Poisson's ratios.

Stress and displacements fields in the matrix and particle are determined by imposing the following twelve continuity conditions

$$\sigma_{rr}^{m}(\tilde{a}_i,\theta_i)=\sigma_{rr}^{S_i}(\tilde{a}_i,\theta_i),\ \tau_{r\theta}^{m}(\tilde{a}_i,\theta_i)=\tau_{r\theta}^{S_i}(\tilde{a}_i,\theta_i) \tag{3a}$$

$$\tilde{u}_{r}^{m}(\tilde{a}_i,\theta_i)=\tilde{u}_{r}^{S_i}(\tilde{a}_i,\theta_i),\ \tilde{u}_{\theta}^{m}(\tilde{a}_i,\theta_i)=\tilde{u}_{\theta}^{S_i}(\tilde{a}_i,\theta_i) \tag{3b}$$

$$\sigma_{rr}^{S_i}(\eta_i\tilde{a}_i,\theta_i)=0,\ \tau_{r\theta}^{S_i}(\eta_i\tilde{a}_i,\theta_i)=0 \tag{3c}$$

Then, by exploiting the orthogonality of the Legendre polynomials and their derivatives in the interval $[0,\pi]$ and by accounting for the remote fields (1) and (2), equation set (3) can be converted into a linear system the coefficients of the series expansion and pertaining to both the inner and outer surfaces of S_1 and S_2. These expressions are here omitted for brevity but allow for finding the generic term of all the unknown sets of coefficients. A numerical approximation of the solution is obtained by truncating the summations in the series solutions at the first T summands.

4 Effective properties

The effect of particle-to-particle interaction on the overall mechanical response of the system is studied by analyzing the change in the relative effective elastic compliance of the composite per unit volume fraction of particles. The Eshelby's formula allows for decomposing the strain energy stored in the composite as follows

$$U=U_0+U_{\mathrm{INT}} \tag{4}$$

Here, U_0 is the strain energy stored in a configuration where the hollow inclusions are replaced with matrix material and U_{INT} is the strain energy stored in a configuration where the external remote loading is replaced by the system of interfacial tractions due to particle-to-particle interactions. The energy U can be expressed as a function of particle outer radii and volume fractions as follows:

$$U=\frac{1}{2}V\frac{\sigma_\infty^2}{E_{\mathrm{eff}}}=\frac{2\pi\left(a_1^3+a_2^3\right)\sigma_\infty^2}{3\Phi E_{\mathrm{eff}}} \tag{5}$$

where V is the volume of the system, E_{eff} is the effective elastic modulus of the composite, and Φ is the volume fraction of particles. Correspondingly, U_0 has the same form as Eq. (5) with the only difference that E_{eff} is replaced by E_m that is the matrix Young's modulus. In the considered two particle system, U_{INT} can be expressed as follows:

$$\begin{aligned}U_{\mathrm{INT}}=&q^3\pi\sum_{i=1}^{2}\Bigg\{\int_0^\pi\left[\sigma_{rr}^{\infty}(\tilde{a}_i,\theta_i)\tilde{u}_r^{S_i}(\tilde{a}_i,\theta_i)+\right.\\&\left.\sigma_{r\theta}^{\infty}(\tilde{a}_i,\theta_i)\tilde{u}_\theta^{S_i}(\tilde{a}_i,\theta_i)\right]\tilde{a}_i^2\sin\theta_i\mathrm{d}\theta_i-\\&\int_0^\pi\left[\sigma_{rr}^{S_i}(\tilde{a}_i,\theta_i)\tilde{u}_r^{\infty}(\tilde{a}_i,\theta_i)+\right.\\&\left.\tau_{r\theta}^{S_i}(\tilde{a}_i,\theta_i)\tilde{u}_\theta^{\infty}(\tilde{a}_i,\theta_i)\right]\tilde{a}_i^2\sin\theta_i\mathrm{d}\theta_i\Bigg\}\end{aligned} \tag{6}$$

By combining Eq. (4), Eq. (5), and Eq. (6) the change in the relative effective elastic compliance per unit volume fraction of particles $\mathcal{S}$ is calculated as

$$\mathcal{S}=\left(\frac{E_m}{E_{\mathrm{eff}}}-1\right)\frac{1}{\Phi} \tag{7}$$

5 Results

In this section, the cases of equal sized particles are studied, that is, A=1. Inter-particle distance is varied by selecting the values 5 and 1.25 for Γ. For each case, both η_1 and η_2 span the range $[0.01,0.99]$. All results presented in this section are obtained by selecting $E_m=3.21\,\mathrm{GPa}$, $\nu_m=0.3$, $\nu_p=0.21$, and the particle Young's modulus $E_p=60\,\mathrm{GPa}$ corresponding to a glass-vinyl ester system, see for example [6].

When the particles have the same outer radius, the problem geometry and the loading conditions allow for reducing the total number of cases to be numerically addressed since the solution for a generic pair of radius ratio values (η_1,η_2) can be used to study the corresponding case when η_1 and η_2 are inverted. In this analysis, the results for the problem of two hollow particles are compared with those obtained for the case of a single inclusion embedded in the same matrix material and undergoing the same tensile loading. This comparison is aimed at clarifying the effect of particle-to-particle interactions and refers to features defined per unit volume fraction of inclusions.

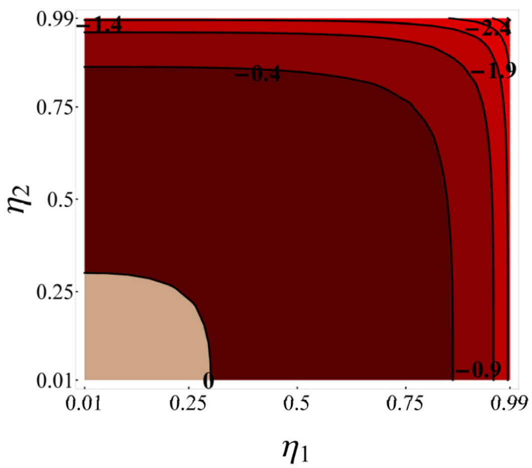

Fig. 2. $\mathcal{S}_{\text{solid}}$- $\mathcal{S}_5$ as functions of η_1 and η_2.

In Fig. 2, the difference between the change in the relative elastic compliance per unit volume fraction for the case of a single solid inclusion $\mathcal{S}_{\text{solid}}$ and $\mathcal{S}_5$ is plotted as a function of η_1 and η_2. The notation $\mathcal{S}_5$ refers to Γ=5 and a similar notation is used for other cases explored in what follows. The change in the relative compliance per unit volume fraction in case of a single solid sphere can be computed by specializing the findings of [9] to the considered material system to obtain -1.83; this means that a solid glass particle produces an increase in the elastic modulus of the neat resin of per unit particle volume fraction. In Fig. 2, two regions are identified by two color scales, brown and red. The brown region identifies configurations where the interaction between the particles yields a higher effective stiffness than the case of a single solid particle. Nevertheless, due to the considerable distance between the particles such stiffening effect is not significative as it causes a maximum value for of 0.006. As η_1 and η_2 increase, this result is inverted. In particular, such change is negative when the particle radius ratios are greater than approximately 0.3, that is, when the particle wall thickness is still considerably large. Thus, very thin interacting particles produce a weaker stiffening effect as compared to a single solid particle.

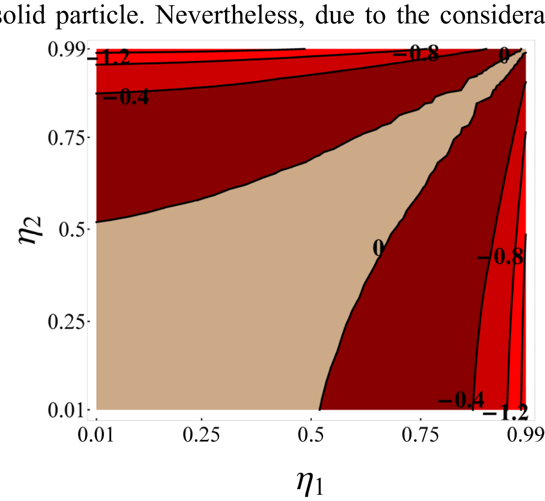

Fig. 3. $\mathcal{S}_{\text{ave}}$- $\mathcal{S}_5$ as functions of η_1 and η_2.

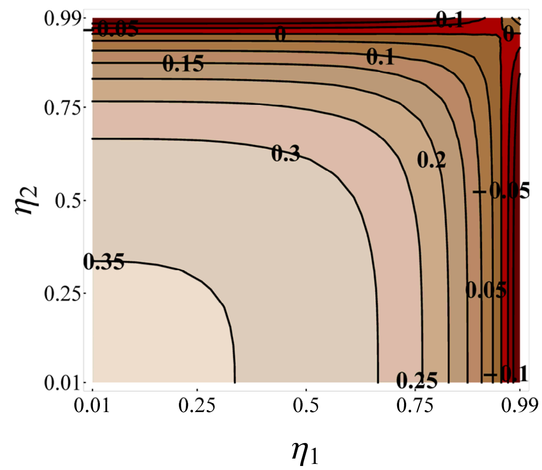

Fig. 4. $\mathcal{S}_5$- $\mathcal{S}_{1.25}$ as functions of η_1 and η_2.

Fig. 3 shows the difference between $\mathcal{S}_{\text{ave}}$ and $\mathcal{S}_5$ as a function of η_1 and η_2. For each pair (η_1, η_2), $\mathcal{S}_{\text{ave}}$ is the change in the relative effective elastic compliance per unit volume fraction for the case of a single hollow particle which has a wall thickness equal to the average of the two particles' wall thicknesses. Such quantity can also be computed by adapting results reported in [9]. The brown region in the plot identifies geometric configurations such that $\mathcal{S}_{\text{ave}}$>$\mathcal{S}_5$. When $\eta_1 = \eta_2 = \eta_{\text{ave}}$, $\mathcal{S}_{\text{ave}}$ is always greater than $\mathcal{S}_5$. Thus, particle-to-particle interaction always provides a stiffening effect as compared to the single hollow particle problem. However, such effect is minimal due to the considerable distance between the particles; therefore, $\Gamma = 5$ can be considered a good approximation of a single hollow particle problem. As the value of $|\eta_1 - \eta_2|$ increases, a transition from the brown region to the red one illustrates that a single hollow particle is a better reinforcement than two particles with a large difference in their wall thicknesses.

The role of the inter-particle distance is described in Fig. 4. This contour plots illustrate the difference between $\mathcal{S}_5$ and $\mathcal{S}_{1.25}$ as a function of η_1 and η_2. Fig. 4 demonstrates that particle wall thicknesses play an important role in defining the

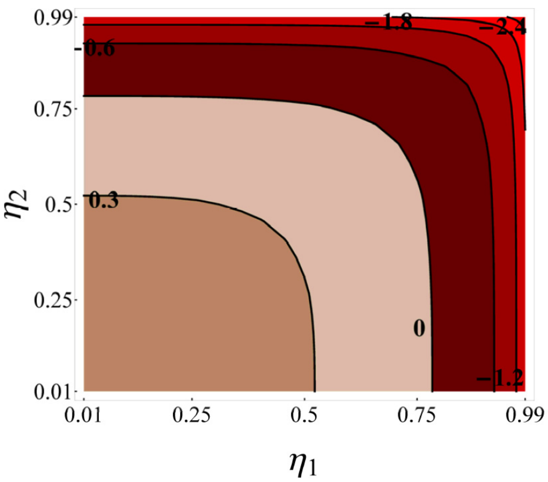

Fig. 5. $\mathcal{S}_{\text{solid}}$- $\mathcal{S}_{1.25}$ as functions of η_1 and η_2.

effect of inter-particle distance on the effective elastic compliance. Generally, as Γ decreases, the composite become stiffer for the majority of the analyzed geometric configurations, see the brown region in Fig. 4. However, as η_1 or η_2 is increased, the stiffening effect reduces until vanishing for η_i greater than 0.94, see the narrow red region in Fig. 4 which only excludes the case of particles with approximately equal wall thicknesses. This finding suggests that particle clustering weakens the elastic properties of glass-vinyl ester syntactic foams when very thin-walled particles are used. The maximum value in Fig. 4 is 0.35 which corresponds to a positive change of 19% with respect to $\mathcal{S}_5$.

Further insight into the effect of inter-particle distance can be garnered from Fig. 5 that duplicates the analysis in Fig. 2 for the case of closely spaced particles. By comparing Figs. 2 and 5, it can be noticed that that elastic interaction between inclusions may overcome the detrimental effect of the entrapped voids and actually yield stiffer composites.

6 Conclusions

In this paper, the effect of particle-to-particle interaction on the global mechanical properties of syntactic foams is studied. More specifically, the problem of two hollow particles embedded in an infinite medium subjected to uniaxial tensile loading is analyzed.

Change in the elastic properties of the composite due to particle-to particle interaction of hollow particles is investigated. Semi-analytical results specialized to glass-vinyl ester systems show that particle-to-particle interaction has a prominent effect on the composite stiffness. This effect is beneficial if the particle wall thickness is sufficiently high; on the other hand, it is detrimental for thin walled particles. The magnitude of this effect is amplified as the inter-particle distance is decreased. Therefore, modeling efforts based on single inclusion schemes are expected to overestimate the effective elastic modulus of the composite for thin walled particles and underestimate for quasi-solid inclusions. This discrepancy is expected to be more pronounced at high filler volume fractions where particles are closely spaced.

Acknowledgments

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