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DOI 10.1016/j.polymer.2024.127463

Publication date 2024

Document Version Final published version

Published in Polymer

Citation (APA)

Soudmand, B. H., Biglari, H., Fotouhi, M., Seyedzavvar, M., & Choupani, N. (2024). A finite element approach for addressing the interphase modulus and size interdependency and its integration into micromechanical elastic modulus prediction in polystyrene/SiO nanocomposites. *Polymer*, *309*, Article 127463. https://doi.org/10.1016/j.polymer.2024.127463

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Polymer



journal homepage: www.elsevier.com/locate/polymer

A finite element approach for addressing the interphase modulus and size interdependency and its integration into micromechanical elastic modulus prediction in polystyrene/SiO₂ nanocomposites

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ARTICLE INFO

Keywords: Interphase modulus Interphase size Interdependency Finite element Polymer nanocomposite

ABSTRACT

The perturbed transitional area between the nanoparticle and matrix shapes the properties of polymer nanocomposites. Due to the stochastic nature of these interphase regions, their size and physical properties are intricately linked. For instance, a higher interphase modulus, E_{int}, might result from a thinner interphase, and vice versa. The inherent randomness can introduce variability in the interphase modulus with respect to interphase thickness, t_{int}. This challenges the practicality of conventional micromechanical approaches, which assume the interphase modulus to be either a constant or a function of filler and matrix properties when predicting the elastic modulus of polymer nanocomposites. Unlike conventional approaches, which simply used interphase quantification to predict global stiffness and treated the interphase modulus independently of its thickness, this study aims, for the first time, to consider the stochastic nature of the interphase, seeking to exclusively explore the interdependencies within the $E_{int} - t_{int}$ relationship in polystyrene/SiO₂ nanocomposites. Simulations were conducted using finite element analysis, FEA, providing high accuracy and flexibility. To manage the large number of simulations, FEA was streamlined with a customized Python scripting, generating a spectrum of (E_{int}, E_{int}) t_{int}) solutions for varying SiO₂ contents based on experimental measurements and a rigorous methodology. Subsequently, empirical equations were formulated, unveiling the relationship between E_{int} and t_{int} per composition. The FEA-driven interphase intercorrelation scheme was compared to the results obtained from a modified three-phase Halpin-Tsai model. Additionally, the FEA scheme was utilized to modulate the HT model by adjusting its relevant interphase terms.

1. Introduction

Nano-reinforced polymer composites are garnering much interest due to their exceptional mechanical properties such as tensile modulus and strength, attained even with addition of small volume fraction of dispersed nano-phases [1–5]. To fully maximize the potential of nano-additives in bolstering the mechanical properties of the base polymer, it is crucial to ensure two primary factors: the even distribution of nanoparticles and the constitution of robust interactions between the nanoparticles and the host matrix [6]. Uniform dispersion amplifies the surface area of nanoparticles within the polymer, while sturdy bonding enables efficient load transfer from the matrix to these rigid nanoparticles [7]. Engineering the polymer-filler interaction hence holds significant importance in augmenting the overall properties [8]. The goal is to design an efficient interphase that modulates the interaction between nanoparticles and the matrix by increasing the attraction between filler and polymer while surpassing interparticle forces [9]. Evaluating the strength of particle bonding involves inspecting the characteristics of the interphase area where the nanoparticle engages with the surrounding material [10]. Interphase quantification can serve as a robust indicator of the bonding effectiveness, indicating how efficiently the composite system attains its intended performance [11].

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https://doi.org/10.1016/j.polymer.2024.127463

Received 28 April 2024; Received in revised form 18 July 2024; Accepted 1 August 2024 Available online 3 August 2024 0032-3861 /@ 2024 Elsevier Ltd. All rights are reserved including those for text and data mining. All tra

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Direct assessment of mechanical properties within an interphase region requires specialized techniques like instrumented nanoindentation testing (IIT) and atomic force microscopy (AFM). However, the limitations imposed by the close proximity of the nanoparticle restrict both elastic and plastic deformation during indentations, making it challenging to solely analyze interphase effects in isolation [12]. The influence of compatibilizers and surface modifications can also lead to uncertainty in defining interphase domains and present challenges in accurately characterizing the interphase characteristics [13]. Moreover, these techniques predominantly rely on localized measurements, lacking the ability to provide effective quantities that reflect the overall physical properties of the interphase [14].

One pragmatic method for determining the physical traits of the polymer-filler interaction involves conducting an inverse analysis by correlating the macro-level experimental measurements to the interphase properties at the microscale via homogenization techniques such as analytical schemes (e.g., Mori-Tanaka and Halpin-Tsai), finite element analysis (FEA), or combination of the two [15-18]. In the framework of analytical formulations, Zare et al. [19] modified the Maxwell model by incorporating t_{int} and E_{int} terms to the model, aiming to predict the tensile modulus and strength of nanocomposites with spherical particles. Through systematic assessments at various t_{int} and E_{int} values, they observed variations in the tensile properties predicted by the developed model. Their findings suggested that the integration of a thick interphase, characterized by a high elastic modulus, significantly contributes to the improvement of the composite material's tensile properties. Jamali et al. [20] adapted the Pukanszky model to determine the interphase thickness and strength within polymer/diamond nanocomposites. While explicit and closed-form solutions offer computational advantages by simplifying calculations and assuming ideal conditions for heterogeneous media, this inherent simplicity constrains computational efficiency and prediction accuracy, thus limiting the accurate description of diverse features.

FEA is a versatile computational tool that surpasses traditional analytical and semi-analytical solutions, offering enhanced design flexibility in terms of geometry, phase quantities, and constitutive equations. FEA provides a balance between precise micromechanical accuracy and adaptable geometry, enabling a realistic representation of composite materials without excessive computational demands. FEA has been used to simulate interphase properties across various nanoparticlereinforced polymer systems [21–24]. For instance, Baneriee et al. [25] employed FEA to examine the impact of interphase modulus and thickness on the mechanical properties of single-walled carbon nanotube/polymer matrix composites by modeling a three-phase representative volume element (RVE). Furthermore, FEA has been extensively combined with molecular dynamics (MD) and continuum mechanics to characterize the mechanical properties of polymer nanocomposites using a multi-scale approach, particularly focusing on the particle-matrix interface [26-28]. Odegard et al. [29] proposed an equivalent-continuum approach to model the nanotube/polymer interface by defining an effective continuum fiber. This method facilitates constitutive modeling of nanotube-reinforced polymer composites, allowing for the prediction of bulk mechanical properties through micromechanical analysis. Yang et al. [30] employed a scale-bridging method to quantify mechanical properties. Initially, they utilized micromechanics to incorporate the particle-matrix interface. Subsequently, they estimated the interface thickness and elastic properties through atomistic structure using MD simulations. Cho et al. [31] introduced an effective interface concept that incorporated particle-size effects on the elastic modulus of nanocomposites. This model accurately predicted interfacial properties influenced by particle size, outperforming micromechanics-based scale bridging methods. Choi et al. [32] integrated MD with 3D FEA to predict global stiffness and the polymer-to-particle load transfer. This was achieved by incorporating an interphase identification procedure that aligned the energy density of the interphase with values predicted by MD simulations. The model

effectively represented stress distribution within the interphase and matrix regions at the atomistic level, while also accounting for the particle-size dependent stiffness of the nanocomposite. Shin et al. [33] employed an equivalent cluster-based homogenization method, characterizing the percolated interphase region near particle clusters with MD and FEA. This approach achieved high computational efficiency without compromising accuracy. Baek et al. [34] introduced a modified interphase model using a multiscale bridging technique, which accounted for interfacial overlapped zones with properties distinct from those of non-overlapped regions. This new model effectively incorporated the effects of particle agglomeration, producing results that qualitatively aligned with experimental trends observed in epoxy/SiC nanocomposites. All the aforementioned studies have aimed to predict the elastic modulus of polymer nanocomposites by incorporating interphase characteristics, such as interphase thickness and modulus, utilizing multiscale techniques based on FEA, MD, and continuum mechanics. While interfacial quantification was majorly utilized for predicting mechanical properties, none of these scholarly works exclusively explored the intrinsic interrelationship between interphase modulus and thickness independently within the interphase region. Instead, these interfacial features were majorly treated independently of each other. Moreover, the potential impact of this interdependence on previously proposed micromechanical models has not been thoroughly investigated.

To fill this research gap, this study addresses the inherent interdependence of interfacial features resulting from the stochastic nature of the interphase. Specifically, the variability of the interphase modulus was investigated with respect to its thickness in polystyrene (PS)/SiO₂ polymer nanocomposite using an extensive series of FEAs, marking the first attempt to account for these complex interactions. This interdependent behavior was then factored into the micromechanical predictive modeling of the elastic modulus in the nanocomposites. FEAs were performed on an RVE containing a single particle. The weight fractions of particles were set to match those found in the tested compositions, aiming to ascertain the effective modulus. The simulations determined the elastic modulus across various interphase thicknesses and moduli. An automated Python scripting procedure was developed to facilitate the execution of numerous FEAs. Based on experimental findings and FEA results, a methodology was devised to establish the correlation between interphase modulus and thickness across various compositions. The FEA-based interdependency relations served as reference guides to adjust the interphase terms in a modified three-phase Halpin-Tsai model. Additional coefficients were integrated into the model using this reference, offering a potent method to enhance the precision of interphase quantities within micromechanical models through FEA assistance.

2. Methodology

2.1. FEA

To ascertain the tensile modulus of the nanocomposite, an RVE was constructed. This RVE comprised a single spherical particle positioned at its center, encased by the interphase material, which, in turn, was surrounded by the matrix. A general-purpose linear brick element with reduced integration (C3D8R) was employed to mesh all three components—matrix, particles, and the interphase—featuring three translational degrees of freedom per node. Surface-based tie constraint connection type was introduced at the particle-interphase and interphase-matrix interphase regions to establish links between the RVE constituents and inhibit inter-component decoherence under tension, simulating prefect bonding conditions. To facilitate the conduction of numerous FE simulations, an automated Python script was developed and integrated into the Abaqus software to generate the RVE models and conduct the simulations.

The elastic moduli and Poisson's ratios for both the polymer

(polystyrene - PS) and fillers (SiO₂ nanoparticles) were established at specific, fixed values: (3.12 GPa, 0.34) for PS and (80 GPa, 0.17) for SiO₂ nanoparticles with a radius of 8 nm, as defined in Ref. [35]. Table 1 outlines the weight fractions of nanoparticles in PS/SiO₂ nanocomposites alongside their respective measured elastic moduli under uniaxial tensile test for each concentration, as derived from the referenced paper. To align with each concentration, the RVE size was calibrated accordingly to match the particular loading fraction. The simulations entailed a predefined range of interphase modulus and thickness values, determined through the analytical approach outputs, as elucidated in subsequent sections.

Considering that both the matrix and the rigid spherical inclusion exhibit isotropic characteristics—their properties are not dependent on direction or specific planes of symmetry—the resulting microstructure can demonstrate an isotropic behavior, which can be represented by the elasticity tensor as expressed below [36]:

$$C = \begin{bmatrix} C_{11} & C_{12} & C_{12} & 0 & 0 & 0 \\ C_{12} & C_{11} & C_{12} & 0 & 0 & 0 \\ C_{12} & C_{12} & C_{11} & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{(C_{11} - C_{12})}{2} & 0 & 0 \\ 0 & 0 & 0 & 0 & \frac{(C_{11} - C_{12})}{2} & 0 \\ 0 & 0 & 0 & 0 & 0 & \frac{(C_{11} - C_{12})}{2} \end{bmatrix}$$
(1)

Following the computation of the stiffness matrix stated above, the elastic characteristics of the material—such as the tensile modulus (E) and Poisson's ratio (v)—can be derived using the following procedure:

$$E = C_{11} - \frac{2C_{12}^2}{C_{11} + C_{12}} \tag{2}$$

$$v = \frac{C_{12}}{C_{11} + C_{12}} \tag{3}$$

To calculate the stiffness tensor components, the RVE undergoes a uniform average strain $\bar{\epsilon}_{ij}$ (i, j = 1, 2, 3). The external strain applied, $\hat{\epsilon}_{ij}$, consists of six components derived from enforcing specific boundary conditions on the displacement vector as follows [37]:

$$\begin{bmatrix} u_1(L, x_2, x_3) \\ u_2(L, x_2, x_3) \\ u_3(L, x_2, x_3) \end{bmatrix} - \begin{bmatrix} u_1(-L, x_2, x_3) \\ u_2(-L, x_2, x_3) \\ u_3(-L, x_2, x_3) \end{bmatrix} = 2L \begin{bmatrix} \widehat{\varepsilon}_{11} \\ \widehat{\varepsilon}_{21} \\ \widehat{\varepsilon}_{31} \end{bmatrix}, 0 \le x_2, x_3 \le L$$
(4)

$$\begin{bmatrix} u_1(x_1, L, x_3) \\ u_2(x_1, L, x_3) \\ u_3(x_1, L, x_3) \end{bmatrix} - \begin{bmatrix} u_1(x_1, -L, x_3) \\ u_2(x_1, -L, x_3) \\ u_3(x_1, -L, x_3) \end{bmatrix} = 2L \begin{bmatrix} \widehat{\varepsilon}_{12} \\ \widehat{\varepsilon}_{22} \\ \widehat{\varepsilon}_{32} \end{bmatrix}, 0 \le x_1, x_3 \le L$$
(5)

$$\begin{bmatrix} u_1(\mathbf{x}_1, \mathbf{x}_2, L) \\ u_2(\mathbf{x}_1, \mathbf{x}_2, L) \\ u_3(\mathbf{x}_1, \mathbf{x}_2, L) \end{bmatrix} - \begin{bmatrix} u_1(\mathbf{x}_1, \mathbf{x}_2, -L) \\ u_2(\mathbf{x}_1, \mathbf{x}_2, -L) \\ u_3(\mathbf{x}_1, \mathbf{x}_2, -L) \end{bmatrix} = 2L \begin{bmatrix} \widehat{\varepsilon}_{13} \\ \widehat{\varepsilon}_{23} \\ \widehat{\varepsilon}_{33} \end{bmatrix}, 0 \le \mathbf{x}_1, \mathbf{x}_2 \le L$$
 (6)

Due to the intricate strain field formed within the RVE under the applied external strain, $\hat{\varepsilon}_{ij}$ in Eqs. (4)–(6), a volume-based averaging scheme was used to derive the average strain developed within the RVE according to the subsequent formulation [38]:

Table 1 SiO₂ nanoparticle weight fractions in PS/SiO₂ nanocomposites and corresponding elastic moduli measured via uniaxial tensile tests (Ref. [35]).

SiO ₂ content (wt.	0.1	0.2	0.75	2	5	10
%) Elastic modulus (GPa)	3.1437	3.1503	3.1830	3.2540	3.4900	3.6040

$$\overline{\varepsilon}_{ij} = \frac{1}{|\omega|} \int \varepsilon_{ij} \, d\omega = \widehat{\varepsilon}_{ij} \, (i, j = 1, 2, 3) \tag{7}$$

In Eq. (7), ω stands for the RVE volume in microscopic domain. Computing the average strain, $\overline{\epsilon}_{ij}$, the constitutive relation between average stress, for a homogenous composite material can be expressed as below:

$$\overline{\sigma}_{p} = \overline{C}_{pq} \overline{\varepsilon}_{q} \ (p, q = 1, 2, ..., 6)$$
(8)

Here, $\bar{\sigma}_p$ represents the average macroscopic stress exerted on the RVE, while \overline{C}_{pq} stands for the microscopic constitutive tensor as defined in Eq. (1) for the isotropic state. Using the displacement-based approach, the elements in each column of tensor \overline{C}_{pq} can be obtained by solving one of six distinct elastic models for the RVE. Each model corresponds to specific columns of the stiffness tensor \overline{C}_{pq} , derived under particular boundary conditions outlined in Eqs. (4)–(6). To compute the components of each column, only a single non-zero strain component, \hat{e}_q is considered, with the remaining elements intentionally set to zero. By assigning a unit value to the non-zero strain term and after implementing the specified boundary conditions in Eqs. (4)–(6), the resulting computed average stress tensor field, $\overline{\sigma}_p$, will align with a single column of the elastic matrix. This column corresponds to the index of the nonzero strain component as stated below:

$$\overline{C}_{pq} = \overline{\sigma}_p = \frac{1}{|\omega|} \int \sigma_p(\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3) \, d\omega, \, \widehat{\varepsilon}_q = 1 \tag{9}$$

The integration process outlined in Eq. (9) is numerically executed at every integration point of an element using the Gauss-Legendre quadrature approach in FE simulation.

Due to the isotropic nature of the examined RVE, obtaining the elastic components in the initial column of the elastic tensor (i.e., C_{11} and C_{12}) can allow for the computation of E and ν , based on Eqs. (2) and (3). Accordingly, a value of 1 was assigned to the external strain field in the x_1 direction ($\hat{\varepsilon}_1 = 1$), corresponding to the first column of the elastic tensor (\overline{C}_{p1} in Eq. (9)), while the remaining components were set to zero. These values were subsequently applied to the boundary conditions outlined in Eqs. (4)–(6). To reduce the computational cost and due to the symmetrical features of the RVE in terms of geometry and constraints, the analysis focused on the top-right-back section of the RVE, constituting one-eighth of the structure, as shown in Fig. 1. Consequently, the boundary conditions in Eqs. (4)-(6) were modified as per Table 2, wherein L represents half the length of the RVE along all three axes $(x_1,$ x_2, x_3), depicted in Fig. 1. The stress-based displacement conditions, stated in Table 2, do not require direct implementation in the FE software. This is because the displacement-based formulation on cube faces inherently satisfies crucial stress-based boundary conditions on the



Fig. 1. The configuration of the RVE model defined within the FE simulation.

Polymer 309 (2024) 127463

Table 2

Boundary conditions set at various faces of the cubic RVE to enforce specific displacement and stress fields.

$0 \le x_2, x_3 \le L$	$0 \le x_1, x_3 \le L$	$0 \le x_1, x_2 \le L$
$u_1(L,x_2,x_3) = L$	$u_2(x_1,L,x_3) = 0$	$u_3(x_1,x_2,L) = 0$
$u_1(0,x_2,x_3) = 0$	$u_2(x_1,0,x_3) = 0$	$u_3(x_1, x_2, 0) = 0$
$\sigma_{12}(L,x_2,x_3) = 0$	$\sigma_{21}(x_1,L,x_3) = 0$	$\sigma_{31}(x_1, x_2, L) = 0$
$\sigma_{12}(0,x_2,x_3) = 0$	$\sigma_{21}(x_1,0,x_3) = 0$	$\sigma_{31}(x_1, x_2, 0) = 0$
$\sigma_{13}(L,x_2,x_3)=0$	$\sigma_{23}(x_1,L,x_3) = 0$	$\sigma_{32}(x_1,x_2,L) = 0$
$\sigma_{13}(0,x_2,x_3)=0$	$\sigma_{23}(x_1, 0, x_3) = 0$	$\sigma_{32}(x_1, x_2, 0) = 0$

surface boundaries [37].

2.2. Analytical approach

Halpin and Tsai [39] presented a mathematical framework for predicting the effective modulus of polymer composites with diverse geometries. For spherical inclusions, this model can be represented as below [40]:

$$E_c = E_m \left[\frac{1 + 2\eta \phi_f}{1 - 2\eta \phi_f} \right] \tag{10}$$

$$\eta = \frac{\frac{E_f}{E_m} - 1}{\frac{E_f}{E_m} + 2} \tag{11}$$

In Eq. (10), the indices *m* and *f* pertain to the matrix and filler, respectively. *E* stands for the tensile modulus, and ϕ indicates the volume fraction. To incorporate the interphase between the particle surface and the adjacent matrix, it is essential to adapt Eq. (10) by introducing the relevant interphase characteristics. The primary factor to consider is the volume fraction of the interphase, ϕ_{int} , which can be computed as described below, assuming a spherical structure [41]:

$$\phi_{int} = \left[\left(\frac{t_{int} + r_f}{r_f} \right)^3 - 1 \right] \phi_f \tag{12}$$

Here, t_{int} represents the thickness of the interphase, and r_f stands for the radius of the particle. Adding the interphase elastic modulus, denoted as, E_{int} , to Eq. (10) leads to the following expression [41]:



Fig. 2. (a) A comparative analysis between experimental [35] and predicted elastic modulus values for *PS/SiO*₂ nanocomposites, (b) illustration of the interphase modulus dependency on the polymer-filler interphase thickness, and (c) dependency of interphase volume fraction, \emptyset_{int} , on the interphase thickness, t_{int} .

$$E_{c} = E_{m} \left[\frac{1 + 2\eta\phi_{f} + 2\eta_{int}\phi_{int}}{1 - \eta\phi_{f} - \eta_{int}\phi_{int}} \right]$$
(13)

$$\eta_{int} = \frac{\frac{E_{int}}{E_m} - 1}{\frac{E_{int}}{E_m} + 2} \tag{14}$$

3. Results and discussion

3.1. Correlation between interphase thickness and modulus based on modified Halpin-Tsai model

The experimental elastic modulus data, as documented in Ref. [35], were compared with the predictive results generated by the modified Halpin-Tsai analytical model (Eqs. 12–14) for polystyrene (*PS*)/SiO₂ nanocomposites, as depicted in Fig. 2a. The adopted micromechanical model introduces two adjustable parameters, namely interphase modulus, E_{int} , and interphase thickness, t_{int} . By finely tuning these two variables, a remarkable alignment between the predicted and measured elastic modulus results was achieved across all particle concentrations, as evident in Fig. 2a. Notably, a range of (E_{int} , t_{int}) solutions exists for each composite, illustrating a continuous variation as shown in Fig. 2b for different nano-SiO₂ contents.

In Fig. 2b, it is evident that a range of solutions exist per sample corresponding to its specific E_c , across various loading fractions. This is presented in terms of the variation in the relative interphase modulus, E_{int}/E_m , against t_{int} . As observed in Fig. 2b, a noteworthy exponential decrease in interphase modulus occurs as the interphase thickness increases, particularly at lower particle loadings. This phenomenon can be explained by considering that, at greater interphase thicknesses, a larger number of polymer chains are available to contribute to the overall stiffness (E_c), reducing the degree of chain suppression and resulting in a lower E_{int} within the interphase region.

Generally, as the filler loading increases, there is a consistent reduction in the developed Eint, as shown in Fig. 2b. This trend is attributed to the higher volume fraction of the interphase with increased particle loading, which distributes a fixed degree of polymer chain immobilization, represented by a given E_c per sample, over a wider interphase region. This trend is clearly visible in Fig. 2c, where higher particle loading corresponds to higher interphase volume fractions. Furthermore, based on Fig. 2b, it is evident that above 0.75 wt% SiO₂, the interphase modulus, E_{int} , remains relatively constant and insensitive to interphase thickness. These samples exhibit relatively similar values for higher particle concentrations, approaching a minimum E_{int} . This threshold indicates that there exists a minimum level of chain densification within the interphase that is necessary for participating in load transfer from the matrix to the particle, especially at higher interphase volume fractions. Importantly, this requirement remains consistent regardless of variations in interphase thickness (Fig. 2b and c).

3.2. Derivation of interphase modulus and thickness via the automated FEA

To investigate the impact of interphase parameters—specifically modulus and thickness—on the effective modulus of the nanocomposite, simulations were carried out across a range of E_{int} and t_{int} values. The minimum interphase layer thickness was set at 2 nm, while the maximum thickness was determined as 8 nm. This upper limit was opted specifically for comparing the FEA results against those derived from the analytical model. The interphase modulus values ranged from the minimum possible magnitude of E_m to the maximum achievable value, which equaled E_f . Each incremental step for t_{int} and E_{int} was set at 0.5 nm and $E_m/2$, respectively, resulting in a total of 650 simulation runs for each composite. Fig. 3 presents the simulation results derived from the FEA applied to the single-particle RVE that represents the nanocomposite, detailed in section 2.1. The results portray the effective modulus variation in relation to E_{int} , where each curve corresponds to distinct interphase thicknesses.

Fig. 3 exhibits a consistent rise in the elastic modulus of the nanocomposites as the concentration of nano-SiO₂ particles rises, when contrasted with lower levels of SiO₂ incorporation. The pattern is ascribed to the higher rigidity of the inorganic particles in contrast to the polymer matrix, introducing further stiffness to the nanocomposite system as the concentration of nano-SiO₂ rises. According to Fig. 3, increasing the interphase thickness resulted in a consistent rise in effective modulus across all compounds, given a constant Eint. This relationship is rooted in thicker interphases accommodating a greater portion of entangled polymer chains, thereby enhancing the interphase area's ability to bear loads and significantly contributing to stress redistribution within the nanocomposite. In Fig. 3, it's clear that across all compositions, when the interphase size is extremely small (e.g., $t_{int} =$ 2 nm), the change in elastic modulus concerning the interphase modulus appears to be minimal compared to thicker interphases. This suggests that in these narrow transition zones from particle to host polymer, the impact of chain immobilization on the load-bearing capacity seems limited, regardless of the densification degree. Conversely, within larger interphase, stress redistribution occurs more readily under external force, allowing for a more efficient response.

Moreover, in a set interphase layer thickness, a higher interphase modulus leads to an increased effective modulus across all compositions. This occurs due to a greater chain densification that reinforces the interphase load-bearing capacity and augments overall stiffness. At lower E_{int} values, a noticeable rise in effective modulus occurs, especially with greater interphase thicknesses. However, beyond a specific threshold value ($E_{int} \approx 17 - 18$ GPa), a more gradual trend emerges, indicating the limited impact of E_{int} on the composite modulus once this threshold is surpassed. At this point, the interphase thickness takes precedence in altering E_c . This suggests that while the entanglement of polymer chains within the interphase can elevate load-bearing capacity to a certain extent, the size of the interphase area imposes no limit on enhancing E_c .

To investigate load-bearing characteristics within interphase regions, von Mises stress values were computed at individual integration points under the specified displacement loads. Fig. 4 illustrates stress distribution within an octant of a typical interphase material. Analysis across various compounds unveiled two primary areas of maximum stress concentration within each interphase material: the central region and the edges, as shown in Fig. 4a and b respectively. To precisely map stress distribution, specific symmetric paths were defined within these high-stress regions, aligning with the positions of maximum stress elements, as visualized in Fig. 4.

Fig. 5 illustrates von Mises stress distribution diagrams acquired along predefined paths based on maximum stress, for different compounds and interphase thicknesses, while maintaining a constant interphase modulus ($E_{int} = 30$ GPa). As depicted in Fig. 5, nanocomposites, containing 0.1 and 0.2 wt% SiO₂, exhibited notably sharp stress peaks across all interphase layer thicknesses, displaying a maximum stress profile configuration similar to the one depicted in Fig. 4a, corresponding to type-I. The type-I stress peaks were more pronounced in thinner interphases compared to thicker ones. Typically, thinner interphases experience heightened stress concentration due to more localized stress distribution and prominent high-stress zones at their peripheral regions.

With the increase in layer thickness, a reduction in stress localization was observed in Fig. 5a and b in the high-stress areas, indicated by a stress peak depression for thicker interphases. In the 0.75 wt% sample (Fig. 5c), however, thicker interphases exhibited much broader and smoother stress profiles within high-stress regions, denoted as type-II in Fig. 4b. This contradicts the patterns observed in thinner interphases of identical composition or samples with lower amounts of SiO₂ (Fig. 5a and b). The suppression of the stress peak can be ascribed to heightened stress-bearing capacity within thick interphases. This sustains elevated



Fig. 3. Relationship between effective modulus and interphase modulus across various interphase layer thicknesses in PS/SiO₂ nanocomposites with differing nano-SiO₂ contents: (a) 0.1, (b) 0.2, (c) 0.75, (d) 2, (e) 5, and (f) 10 wt%.

stress levels across a greater number of elements located within the thicker regions, resulting in a broader distribution of high-stress elements rather than a distinct peak (Fig. 5c). The alteration in stress distribution led to the occurrence of maximum stress throughout the thickness and near the RVE edge, as shown in Fig. 4b.

Based on Fig. 5, a critical interphase thickness, $t_{int,c}$, can be defined beyond which, the stress distribution profile shifts from type-I with a

sharp, localized peak at the interphase periphery (Fig. 4a) to type-II, characterized by a broader and smoother peak developed around its thickness (Fig. 4b). This transition correlates with the increased interphase volume fraction (\emptyset_{int}) described in Eq. (12), stemming from a rise in the particle volume fraction (\emptyset_f). A larger interphase radius, with a constant thickness, demonstrates better resistance to higher stress levels by spreading stress across higher number of elements. Improved

B.H. Soudmand et al.



Fig. 4. Symmetric path delineation to illustrate maximum stress contours within an octant of the interphase material, highlighting distinct occurrences at two key positions: (a) Center (type-I) and (b) edge (type-II).

resilience of the interphase facilitates more effective stress redistribution within the material, preventing localized concentration in specific regions. Higher stress intensity is noticeable in the type-II stress profile when compared to type-I (excluding those at the peak area). Type-I exhibits a sharp peak with the remaining elements experience significantly lower stress intensities (Fig. 5c–e). As \emptyset_f increases further, this trend persists, revealing a consistent decrease in $t_{int,c}$, reaching values equal to or less than 2 nm ($t_{int,c} \leq 2$ nm), as depicted in Fig. 5e and f, respectively.

In Fig. 5, the type-I profile displays a decrease in stress intensity with increasing t_{int} at lower SiO₂ concentrations (Fig. 5a–c). Conversely, the type-II stress distribution consistently rises between 2 and 10 wt% SiO₂ (Fig. 5d-f) concerning t_{int}, showing a sharper peak at thicker interphases. The heightened stress intensity, particularly evident in the thicker interphases of the type-II profile (seen in Fig. 5d-f), is due to a notable increase in the $\mathcal{Q}_f/\mathcal{Q}_m$ ratio. This disparity significantly amplifies stress between the matrix and filler due to substantial differences in their elastic moduli. Consequently, this intensified stress concentration at the filler-polymer boundary directly impacts the interphase, leading to more pronounced high-stress regions. The higher E_c at higher nanoparticle fractions (Fig. 3e and f) necessitates a more intense stress redistribution process within the interphase, resulting in more severe stress localization and intensity among the high-stress-bearing elements. This phenomenon is particularly emphasized with thicker interphases, as depicted in the diagrams of Fig. 5d-f.

A methodology was employed to derive a spectrum of potential (E_{int} , t_{int}) solutions for various compounds, leveraging the FEA outputs

showcased in Fig. 3. Using the PS/0.75SiO₂ compound as an example, this methodology illustrates the process in Fig. 6. For each composition, the intersection between the $y = E_{exp}$ line (representing the experimental elastic modulus) and the $E_c - E_{int}$ curves offer multiple potential (E_{int} , t_{int}) solutions. The intersection points form a 2D curve per sample, exemplified in Fig. 6. To enhance precision in determining the intersection point, the $E_c - E_{int}$ curves for each interphase layer thickness underwent interpolation, resulting in a dataset of 10⁶ data points within its range.

Fig. 7 displays the resulting outputs obtained from the methodology outlined in Fig. 6, displaying the variation of E_{int}/E_m in relation to the interphase thickness for various compounds. To ensure clarity and coherence within the diagram, an initial value of t_{int} at 3.5 nm was chosen. This decision aimed to enhance visualization, as certain samples failed to yield a viable (E_{int} , t_{int}) solution below this threshold.

In Fig. 7, it's apparent that thinner interphase layers demand a higher interphase modulus to achieve the desired elastic modulus in the nanocomposite. A higher interphase modulus, at a constant interphase layer thickness, signifies the superior performance of the nanocomposite in forming a robust interphase, shown by higher E_{int}/E_m ratios. Notably, increasing SiO₂ content consistently enhances interphase bonding in nanocomposites across various interphase layer thicknesses, especially up to a 5 wt% SiO₂ loading. The consistent increase in interphase modulus due to the addition of nanoparticles, maintaining higher E_{int} values even at elevated concentrations, highlights the effective integration of nanoparticles into the matrix. This integration, in terms of bonding and distribution, suggests an absence of significant negative



Fig. 5. Von Mises stress profiles along predefined symmetric paths, delineated in Fig. 4, including diverse interphase thicknesses while maintaining a constant interphase modulus of $E_{int} = 30$ GPa, for different nano-SiO₂ contents: (a) 0.1, (b) 0.2, (c) 0.75, (d) 2, (e) 5, and (f) 10 wt%.

effects typically caused by particles, like poor interfacial adhesion or dispersion, both of which can degrade the quality of the polymer-filler interphase.

However, in the case of PS/10SiO₂, the effectiveness diminishes, likely due to potential nanoparticle dispersion deterioration at the 10 wt % SiO₂ concentration. This aggregation weakens the polymer-filler adhesion, leading to lower E_{int}/E_m ratios and hindering the $E_c \mid_{exp}$ from increasing in line with the rise in nanoparticle loading. Based on Fig. 7, when t_{int} values are extremely low ($t_{int} \leq 4nm$), the required interphase

modulus to achieve the specified $E_c |_{exp}$ increases exponentially, surpassing the matrix values significantly. This becomes impractical in realworld scenarios. Hence, a viable solution is to ensure that the interphase thickness exceeds a minimum t_{int} (e.g., 4 nm). This compensates for the necessity of excessively high interphase modulus.

To establish empirical relations between interphase modulus and thickness across various compositions, FEA was employed, leveraging the curves presented in Fig. 7. Eqs. 15–20 represent the derived empirical relationships $E_{int} - t_{int}$ for distinct compounds. These



Fig. 6. Derivation of the $E_{int} - t_{int}$ plots for each sample, obtained from the $E_c - E_{int}$ plots alongside the experimentally determined elastic modulus, $E_c |_{exp}$, specific to each composition.

equations were derived by fitting high-order (degree 8) polynomials to each curve, yielding coefficients that characterize the relationships. The units utilized for the interphase modulus and thickness were selected as GPa and nm, respectively, aligning with those specified in Fig. 7.

$$E_{int}^{0.1 wt.\%} = E_m \left(9.2 t_{int}^6 - 100 t_{int}^5 + 667.9 t_{int}^4 - 2816 t_{int}^3 + 7311.8 t_{int}^2 - 10694.1 t_{int} + 6764.9\right)$$
(15)

$$E_{int}^{0.2 \text{ wt.\%}} = E_m \left(4.3 \text{ } t_{int}^6 - 49.4 \text{ } t_{int}^5 + 352.4 \text{ } t_{int}^4 - 1594.5 \text{ } t_{int}^3 + 4471.2 \text{ } t_{int}^2 - 7110.8 \text{ } t_{int} + 4931.9 \right)$$

$$(16)$$

$$\begin{split} E_{int}^{0.75 \text{ wt.\%}} = & E_m \Big(-2.1 \ t_{int}^7 + 42.5 \ t_{int}^6 - 483.6 \ t_{int}^5 + 3406.2 \ t_{int}^4 - 15200 \ t_{int}^3 \\ &+ 41963.5 \ t_{int}^2 - 65536.3 \ t_{int} + 44358.2 \Big) \end{split}$$
(17)

$$E_{int}^{2 \text{ wt.}\%} = E_m \left(-1.1 \text{ } t_{int}^7 + 23.5 \text{ } t_{int}^6 - 279.1 \text{ } t_{int}^5 + 2059.7 \text{ } t_{int}^4 - 9662.4 \text{ } t_{int}^3 + 28141.5 \text{ } t_{int}^2 - 46536.6 \text{ } t_{int} + 33485.7 \right)$$
(18)

$$\begin{split} E_{int}^{5 wt.\%} = & E_m \Big(-1.8 \ t_{int}^7 + 37.7 \ t_{int}^6 - 450.1 \ t_{int}^5 + 3338.6 \ t_{int}^4 - 15744.9 \ t_{int}^3 \\ & + 46105.5 \ t_{int}^2 - 76660.2 \ t_{int} + 55448.6 \Big) \end{split}$$

$$E_{int}^{10 \ wt.\%} = E_m \left(-0.13 \ t_{int}^7 + 2.4 \ t_{int}^6 - 24.9 \ t_{int}^5 + 156.9 \ t_{int}^4 - 620.1 \ t_{int}^3 + 1502.5 \ t_{int}^2 - 2046.2 \ t_{int} + 1215.4 \right)$$
(20)



Fig. 7. Variation of E_{int}/E_m ratio against interphase thickness derived from FEA results across different compounds.

Fig. 8 presents a comparison between the E_{int} - t_{int} curves obtained from FEA (Fig. 7) and the analytical Halpin-Tsai model (Fig. 2b) across various compositions. The findings clearly demonstrate that the FE model effectively addresses the limitations of the HT model by extending the E_{int} - t_{int} relation to lower interphase thickness values and higher magnitudes of interphase modulus, E_{int}. Based on Fig. 8, while there's a consistency in the relative alignment between FEA and HT results for lower concentrations at larger t_{int} values ($t_{int} \ge 7 nm$), the data, especially at higher filler loadings (≥ 2 wt%), lack proper alignment when compared against each other. The positioning of curves representing each sample's composition in relation to others varies notably between the two models. In the HT model, there's a trend of lower E_{int}/E_m ratios for higher levels of SiO₂ incorporation at a given *t_{int}*. Conversely, the FEA results provide an opposing trend, where higher concentrations generally lead to increased magnitudes of E_{int}/E_m for a fixed interphase thickness. Moreover, the E_{int}-t_{int} interconnection curves derived from the HT model indicate a relatively steady E_{int}/E_m ratio across various interphase thickness values, displaying a remarkable insensitivity to this parameter. In contrast, the FEA-based curves offer a more holistic and applicable (Eint-tint) solution curve for each composition, regardless of the nanoparticle concentration.



Fig. 8. Comparative plot depicting E_{int}-t_{int} interconnection curves derived from both analytical Halpin-Tsai and FEA models for different compounds.

The findings from Fig. 8 suggest that the modified Halpin-Tsai model, incorporating interphase terms such as the interphase modulus and thickness in Eqs. 12–14, exhibit significant limitations in effectively relating the interphase thickness to the resulting modulus, particularly when compared to the outcomes derived from FEA. To resolve the issue, the interphase term, η_{int} , outlined in Eq. (14) can be substituted with the term, η_{int}^* , as articulated below:

$$\eta_{int}^{*} = \frac{\alpha_{1} \left(\frac{E_{int}}{E_{m}}\right)^{\beta_{1}} + \gamma_{1}}{\alpha_{2} \left(\frac{E_{int}}{E_{m}}\right)^{\beta_{2}} + \gamma_{2}}$$
(21)

Here, the term $\frac{E_{ent}}{E_m}|_{FEM}$ denotes the y-axis values extracted from the $E_{int} - t_{int}$ curves generated through FEA as depicted in Fig. 5. To augment the density of data points within the curve range, 100 data points were interpolated for each curve in Fig. 7. The constants $(\alpha_1, \alpha_2, \beta_1, \beta_2, \gamma_1, \gamma_2)$ represent the adjustment parameters meticulously crafted to govern the following equation:

$$E_{c}|_{exp} = E_{m} \left[\frac{1 + 2\eta \phi_{f} + 2\eta_{int}^{*} \phi_{int}}{1 - \eta \phi_{f} - \eta_{int}^{*} \phi_{int}} \right]$$
(22)

To determine the unknown constants in the equation above, a process was utilized involving six randomly selected sample points per compound from the interpolated curves in Fig. 7. The t_{int} values for each point were substituted into the interphase volume fraction term, ϕ_{int} , as outlined in Eq. (12). Subsequently, the $\frac{E_{int}}{E_m}$ values corresponding to these points were inserted into Eq. (22) to derive the associated interphase term, η_{int}^* , as defined in Eq. (21). To uncover the unknown terms, the following minimization procedure was established:

Find: $\alpha_1, \alpha_2, \beta_1, \beta_2, \gamma_1, \gamma_2$.

$$\text{Minimize}: f\left(t_{int}, \frac{E_{int}}{E_m}|_{FEM}\right) = E_c \mid_{exp} - E_m \left[\frac{1 + 2\eta\phi_f + 2\eta^*_{int}\phi_{int}}{1 - \eta\phi_f - \eta^*_{int}\phi_{int}}\right]$$
(23)

 $\left(t_{int}, \frac{E_{int}}{E_m}|_{FEM}\right) =$ Six random data points extracted from the interpolated $E_{int} - t_{int}$ curves (Fig. 7)

After applying the minimization algorithm outlined in Eq. (23), six distinct formulations were derived in Eqs 24–29, each tailored to account for the pertinent η_{int}^* value to predict the elastic modulus of the

corresponding composition using the proposed HT-FEA approach, as described below:

$$\eta_{int}^{*}|_{0.1 \text{ wt.\%}} = \frac{2.3979 \left(\frac{E_{int}}{E_{m}}|_{FEM}\right)^{-0.1440_{1}} + 2.3129}{-2.3524 \left(\frac{E_{int}}{E_{m}}|_{FEM}\right)^{0.0066} + 2.7072}$$
(24)

$$u_{int}^{*}|_{0.2 \text{ wt.\%}} = \frac{0.6735 \left(\frac{E_{int}}{E_{m}}|_{FEM}\right)^{0.8926} + 0.8117}{0.5569 \left(\frac{E_{int}}{E_{m}}|_{FEM}\right)^{1.1036} + 3.2960}$$
(25)

$$h_{int}^{*}|_{0.75 \text{ wt.}\%} = \frac{0.4434 \left(\frac{E_{int}}{E_{m}}|_{FEM}\right)^{0.7964} + 0.4879}{0.0248 \left(\frac{E_{int}}{E_{m}}|_{FEM}\right)^{2.0967} + 2.5407}$$
(26)

$$\eta_{int}^{*}|_{2 \text{ wt.\%}} = \frac{1.0367 \left(\frac{E_{int}}{E_{m}}|_{FEM}\right)^{0.9141} + 1.2802}{0.0794 \left(\frac{E_{int}}{E_{m}}|_{FEM}\right)^{1.5586} + 1.3637}$$
(27)

$$\eta_{int}^{*}|_{5 \text{ wt.\%}} = \frac{1.0476 \left(\frac{E_{int}}{E_{m}}|_{FEM}\right)^{0.6843} + 1.2398}{1.3140 \left(\frac{E_{int}}{E_{m}}|_{FEM}\right)^{0.6629} + 1.2202}$$
(28)

$$\eta_{int}^{*}|_{10 \text{ wt},\%} = \frac{1.7439 \left(\frac{E_{int}}{E_{m}}|_{FEM}\right)^{0.4365} + 1.7856}{0.1065 \left(\frac{E_{int}}{E_{m}}|_{FEM}\right)^{1.3009} + 4.4386}$$
(29)

4. Conclusion

The inherent variability in interphase properties, depending on the polymer-filler interaction quality, induces inherent correlations between the elastic modulus and thickness of the transitional region. However, existing micromechanical models to date have not taken into account this interplay when predicting the elastic modulus in polymer nanocomposites. Instead, they largely treated the interphase modulus independently of its dimensions, disregarding the stochastic nature of the interphase. To address this concern, this study focuses on exploring the intrinsic variability in the $E_{int} - t_{int}$ relationship via a series of FEAs. Subsequently, this variability is incorporated into the micromechanical predictive modeling of the elastic modulus in polystyrene (PS)/SiO₂ nanocomposites. Python script was generated to automate the FEA, facilitating multiple simulations on the RVE model. These RVEs replicate particle fractions similar to those observed in experimental samples, varying in interphase thicknesses and elastic moduli. Numerous FEAs were carried out to generate $E_c - E_{int}$ diagrams including a range of interphase thicknesses for each composition. The diagrams depicted a steady rise in elastic modulus with the addition of particles. Additionally, thicker interphases with greater elastic moduli correlated with higher composite moduli. Analysis of stress distribution within the interphase revealed that interphase thickness and particle fraction significantly determined whether stress localized or distributed smoothly. Additionally, both interphase factors impacted the location of maximum stress occurrence. The $E_c - E_{int}$ diagrams were further employed to formulate $E_{int} - t_{int}$ curves using a practical methodology. This method offered a range of potential solutions (E_{int}, t_{int}) for different compounds by employing $E_c - E_{int}$ diagrams. For each composition, the solutions were derived by intersecting these curves with a horizontal line that represented the experimental elastic modulus of the respective sample. The interphase diagrams of $E_{int} - t_{int}$ indicated that thinner interphases demanded higher interphase moduli to develop the required modulus for a specific composition, while thicker interphases demonstrated the opposite trend. Additionally, following the interphase diagrams, the interphase modulus consistently increased with higher particle loadings for a specified thicker interphase layer. This trend signifies a consistent enhancement in interphase bonding within nanocomposites, aligning with the increased concentration of nanoparticles. Comparing interphase diagrams using FEA and the initial Halpin-Tsai model across different compositions showed that the FE model effectively overcame the limitations of the HT scheme. It extended the Eint-tint relation to smaller interphase thickness values and higher Eint, displaying a more coherent trend across various samples compared to the micromechanical model. Finally, the interphase diagrams (E_{int} - t_{int}) were employed to refine the HT model within the new FEA framework. The procedure involved establishing a modified interphase term, η_{int}^* , integrating the FE-centered interphase relationship specific to each composition.

5. Limitations, challenges, and future scope

The present paper introduces an innovative, rigorous, and precise approach to deriving the interfacial interrelationship using FEA, experimental data, and a micromechanical model. However, there are several issues that can be addressed in future studies, as follows:

- Despite the model's high accuracy, it relies on specific volume fractions of nanocomposite samples with known elastic moduli. Future work can enhance the model by considering the filler volume fraction, \emptyset_f , as a variable. This would allow for interfacial interrelation analysis as a function of nanofiller volume fraction, increasing the model's scalability and generalizability.
- A primary challenge in enhancing the generalizability of this hybrid model lies within the FEA process, which needs to be customized to the material model. This customization necessitates independent runs whenever material properties change. Developing a general model to predict interfacial interdependency in nanocomposite systems using a high-fidelity FEA approach requires addressing this limitation. This is particularly critical for systems lacking experimental data and having arbitrary volume fractions. Consequently, a comprehensive dataset is indispensable, including both FEA and experimental results across a wide range of nanocomposite systems.

Such a dataset would enable the construction of intricate multivariate surface response models using data-driven approaches, such as machine learning.

• The current methodology represents an initial step toward a more accurate demonstration of interfacial interdependency. Further work is needed to refine the model and extend its application to a broader range of nanocomposite systems. This includes considering various volume fractions, filler geometries (e.g., platelets, nanotubes, fibers), interfacial qualities, and other factors. Additionally, more intricate FE modeling can be employed to simulate complex interparticle interactions, particle distribution uniformity, and the presence of aggregates. These simulations can help investigate their role in altering interfacial interdependency in diverse polymer nanocomposite systems.

Funding

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

CRediT authorship contribution statement

Behzad Hashemi Soudmand: Writing – original draft, Validation, Software, Methodology, Investigation, Formal analysis, Conceptualization. Hasan Biglari: Writing – review & editing, Supervision, Methodology, Conceptualization. Mohammad Fotouhi: Writing – review & editing, Supervision, Methodology. Mirsadegh Seyedzavvar: Writing – review & editing, Supervision. Naghdali Choupani: Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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