Analyzing effects of surface roughness, voids, and particle–matrix interfacial bonding on the failure response of a heterogeneous adhesive

Bowen Liang\textsuperscript{a}, Anand Nagarajan\textsuperscript{a}, Hossein Ahmadian\textsuperscript{b}, Soheil Soghrati\textsuperscript{a,c,*}

\textsuperscript{a} Department of Mechanical and Aerospace Engineering, The Ohio State University, USA
\textsuperscript{b} Department of Integrated Systems Engineering, The Ohio State University, USA
\textsuperscript{c} Department of Materials Science and Engineering, The Ohio State University, USA

Received 24 April 2018; received in revised form 2 November 2018; accepted 3 December 2018
Available online 13 December 2018

Highlights
\begin{itemize}
  \item Integrated computational framework to create realistic FE models of adhesive microstructure.
  \item Employed ductile damage and cohesive–contact models to simulate mechanical behavior.
  \item Performed a comprehensive study to determine appropriate size of statistical RVE.
  \item Analyzed effects of various microstructural features on failure response.
  \item Outcome: adherend surface roughness and voids have negligible impact on strength and toughness.
\end{itemize}

Abstract

An automated computational framework is introduced to link the microstructure to the micromechanical behavior of a heterogeneous adhesive composed of an epoxy matrix and embedded silica particles. A new reconstruction algorithm is employed to synthesize 3D periodic microstructural models of this materials system based on morphological and statistical data extracted from micro-computed tomography images. A non-iterative mesh generation algorithm is implemented in parallel to transform resulting virtual microstructures into finite element (FE) models. The continuum ductile and cohesive damage models used for simulating the adhesive’s failure response are calibrated with experimental data and the statistical representative volume element (SRVE) is identified. We have then carried out several high-fidelity FE simulations to investigate the effects of pre-existing voids in the adhesive layer, surface roughness of adherends, and the bonding strength along particles–matrix interfaces on the failure response of the adhesive subject to tensile, compressive, and shear loads.

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Keywords: Heterogeneous adhesive; Finite element; Microstructure reconstruction; Cohesive damage; Surface roughness
1. Introduction

Adhesive bonding is a viable alternative to conventional joining techniques such as welding and riveting, which provides advantages such as reduced stress concentration, high corrosion resistance, water tightness, and the ability to join thin and dissimilar materials [1–3]. The latter feature is essential for the design of light-wight structures, which has promoted the use of adhesive joints in the automotive and aerospace industry in the past few decades [4,5]. For example, adhesive-bonded joints have been employed in manufacturing car body shells, as well as assembling windows, windscreens, and indoor cladding [6,7]. Structural adhesives have also been used to bond stringers to the skin of aircraft fuselage and wing structures [8]. Heterogeneities such as hard particles (e.g., glass and silica) [9], rubber [10], and carbon nanotubes [11] can be embedded in such adhesives to improve their fracture toughness, and provide multi-functionality (e.g., electrical conductivity [12] and self-healing capacity [13]). Note that the mechanical performance of the adhesive layer is highly dependent on its microstructural features, including the type, shape, volume fraction, and spatial arrangement of heterogeneities, as well as their interfacial bonding strength with the surrounding polymeric matrix [9]. Further, the preparation of adherend surfaces and their roughness could have a notable impact on the failure response of adhesive-bonded joints [14].

One of the challenges toward simulating the failure response of adhesive-bonded joints is the small thickness of an adhesive layer (often a few hundred microns [15]) compared to other characteristic length scales of the structure. This mismatch between characteristic length scales, which prohibits the use of direct numerical simulation (DNS), necessitates replacing the adhesive layer with zero-thickness elements and employing the cohesive zone modeling (CZM) to carry out a finite element (FE) simulation at the macroscale [16,17]. However, the implementation of CZM requires identifying an appropriate traction–separation law that characterizes the failure response of the adhesive layer. Several phenomenological and mathematically simplified cohesive models such as the polynomial [18], bilinear [17], exponential [18], and trapezoidal [19] models can be used for this purpose. However, such models do not guarantee the consistency of the constitutive relationship subject to mixed-mode loadings [20]. In order to address this issue, one can implement a potential-based cohesive model, in which the traction–separation law is evaluated based on an energy potential function [21–23]. A critical review of advantages and limitations of various CZMs is provided in [20].

Several experimental studies have been carried out to determine the effect of microstructure on the traction–separation law that characterizes the failure response of structural adhesives in CZM. Among these efforts, we can mention the study on the impacts of the joint thickness, type of loading, and curing temperature on the fracture toughness of polyethylene-based epoxy adhesives with embedded glass fibers [24]. Kawaguchi and Pearson [25,26] investigated the effects of moisture content and particle–matrix adhesion strength on the mechanical behavior of an epoxy-based adhesive reinforced with glass particles. A study on the failure response of a silver-filled conductive adhesive subject to an elevated temperature and high humidity is presented in [12]. Also, efforts have been made to experimentally quantify the effect of different design and manufacturing parameters such as the introduction of heterogeneities [27], operating environment [28], and surface treatment [29,30] on the failure response of adhesives.

Unlike experimental studies, only a limited number of computational works have investigated the relationship between the microstructure and the failure response of structural adhesives. Multiscale approaches [31,32] can be employed for such analyses by assigning a representative volume element (RVE) to each point of the macroscopic model and simulating the mechanical behavior at the microscale to evaluate the corresponding constitutive behavior [33]. In the context of multiscale FE analysis, the FE$^2$ method [31,34] is one of the most popular techniques, in which the stress state at each quadrature point of elements discretizing the macroscopic model is evaluated by simulating the failure response of a corresponding RVE. Recently, the homogenization theorem is employed to derive computational cohesive laws for thin adhesive layers by linking the microstructure to the damage process [35–38]. Fully-coupled FE$^2$ multiscale analyses are also carried out to approximate the failure response of adhesive-bonded joints [39,40], although this approach could be highly computationally demanding when modeling the complex 3D microstructure of heterogeneous adhesives.

It must be noted that the concept of RVE is introduced in [41–43] as a small representative volume, for which the volume average of material properties are similar to those of the macroscopic body. However, due to perturbations in local morphologies and material properties, especially for localized properties such as plasticity and damage [44], the underlying assumption that the microstructure is composed of periodic repetitive arrays of heterogeneities in aforementioned references is violated. Thus, the concept of statistical RVE (SRVE) has been used in several works (e.g., [44–47]) to evaluate homogenized properties for the macroscopic analysis. Statistical approaches involving the
construction of multiple realizations of the microstructure with various sizes and volume fractions of heterogeneities are employed in this context to determine the existence of SRVE and identify its smallest characteristic length scales.

Accurate prediction of the traction–separation law used in CZM requires incorporating realistic microstructural features of the adhesive layer in the FE model. The adhesive microstructure can be characterized using digital data such as focused ion beam scanning electron microscope (FIB-SEM) [48] and micro-computed tomography (micro-CT) images [49]. Such data can be directly converted into an FE model after performing required image processing steps (noise filtration, smoothing, segmentation, etc.), followed by the construction of a conforming mesh. However, for adhesives with intricate microstructures involving arbitrary-shaped particles and pre-existing defects (e.g., voids), identifying all material interfaces in the imaging data to build a realistic microstructural model is often a major challenge [50]. Even in cases that such challenges are successfully overcome, this laborious and expensive process only provides a single FE model, which would not be suitable for applications such as Integrated Computational Materials Engineering (ICME) that require performing multiple simulations on distinct microstructures.

An alternative to direct image-based modeling is to utilize a reconstruction algorithm to virtually build realistic microstructures based on particles’ morphologies and statistical features (e.g., volume fraction and spatial arrangement) extracted from digital data. Several algorithms are introduced for the reconstruction of such heterogeneous microstructures, such as descriptor-based methods [51,52], pixel switching [53,54], mass–spring mutation operator [55,56], random field algorithms [57], and Voronoi Tessellation based techniques [58]. During the reconstruction process, the microstructure is characterized using a set of probability descriptor functions (PDFs), e.g., N-point correlation [59], lineal-path [60], chord-length distribution [61] functions, and Fourier-shape-descriptors to characterize realistic morphologies [62]. Stochastic optimization techniques such as the genetic algorithm (GA) [56] and simulated annealing (SA) [63] are often employed to replicate these PDFs in the reconstructed microstructure. Note that eliminating overlapping particles while quantifying their optimized spatial arrangement in such algorithms could be a computationally demanding process. It is also worth mentioning that some of the aforementioned methods are not capable of reconstructing a periodic microstructure, which is crucial for assigning periodic boundary conditions in the FE model to minimize damage localization along domain boundaries in micromechanical simulations [64]. A comprehensive review of microstructure characterization techniques and reconstruction algorithms is provided in [65].

After reconstructing the microstructure, the next step is to build the FE model by generating an appropriate conforming mesh with small element aspect ratios and a negligible discretization error. This can be achieved using sophisticated mesh generation algorithms such as the Delaunay refinement [66], Octree-based methods [67], or the advancing front [68]. In all these algorithms, an iterative optimization/smoothing phase is employed to improve elements’ shapes and aspect ratios, and eliminate those with extremely high or low dihedral angles. For 3D problems with complex geometries, such as a heterogeneous adhesive microstructure, this could be a challenging and time-consuming process that may require user intervention to manually change parameters or locally modify the mesh structure. Enriched FE-based techniques such as the eXtended/Generalized FEM (X/GFEM) [69–71] and hierarchical interface-enriched FEM (HIFEM) [72,73] can be employed to obviate the need for the construction of conforming meshes. However, the implementation of such methods often involves additional challenges such as ill-conditioning of the stiffness matrix and the loss of accuracy in recovering stresses along material interfaces [74,75]. Further, the lack of control on aspect ratios of integration sub-elements practically prohibits the application of explicit time integration methods for solving nonlinear transient problems in such methods, as the stable time increment is a function of characteristic length scales of elements [76].

Recently, Soghrati et al. [77–79] have introduced a new mesh generation algorithm named Conforming to Interface Structured Adaptive Mesh Refinement (CISAMR) that can resolve the implementation issues associated with both conventional meshing and enriched FE-based algorithms. Similar to the latter type of methods, CISAMR allows the use of a simple structured mesh for discretizing the domain. However, instead of using enrichment functions to approximate field discontinuities, this non-iterative algorithm locally transforms the background elements cut by material interfaces into adaptively-refined conforming elements with proper aspect ratios. More details regarding the CISAMR algorithm, which is employed for creating FE models of the adhesive microstructure in the current manuscript, are presented in Section 4.

Appropriate constitutive models must then be incorporated in the FE model to simulate the failure response in the adhesive layer [80]. Two major damage mechanisms can be identified in a heterogeneous adhesive: delamination along particle–matrix interfaces and damage within the matrix [81]. The former can be simulated using cohesive elements with a given traction–separation law to quantify the interfacial bonding strength, as well as a contact model
to avoid particle/matrix interpenetration under compression [82]. The polymer matrix of structural adhesives often shows brittle failure behavior subject to tensile loads, which can be simulated using a brittle continuum damage model [35,38]. However, this materials system experiences a significant plastic deformation before damage under shear and compression [83], which must be simulated using an appropriate ductile damage model [84]. In this work, a phenomenological ductile damage model, relating the damage criterion to stress triaxiality, is employed to simulate both brittle and ductile failure modes [83].

In this manuscript we employ a multiscale computational cohesive model to study the impact of various microstructural features, namely pre-existing voids, particle–matrix interfacial bonding, and adherend surface roughness, on the failure response of a heterogeneous adhesive. The formulation of this model, together with cohesive–contact and continuum ductile damage models used for simulating the failure response of adhesive, is presented in Section 2. A stochastic microstructure reconstruction algorithm is introduced in Section 3 for synthesizing realistic microstructural models of the adhesive, followed by a brief overview of the CISAMR mesh generation algorithm in Section 4. The automated computational framework formed by integrating these algorithms enables constructing realistic 3D microstructural models and subsequently high fidelity FE models of the adhesive layer, which is one of the key differences of this modeling effort compared to previous studies on this subject. The challenging process of constructing such models has led to either using over-simplified geometric models for adhesives or overlooking phenomena involved in their failure process (e.g., ductile damage in the matrix or contact forces along particle–matrix interfaces) in majority of existing works. After calibrating damage models with experimental data, we also investigate the existence and appropriate size of SRVE in Section 5. A comprehensive study on the impact of microstructural features such as pre-existing voids, adherent surface roughness, and particle–matrix interfacial bonding strength on the failure response of the adhesive layer is presented in Section 6. Final concluding remarks are provided in Section 7.

2. Problem formulation

2.1. Micro–macro formulation

Consider an adhesive-bonded structure \( \Omega \subset \mathbb{R}^3 \) with boundary \( \partial \Omega \) described in the macroscopic coordinate system \( x_M \), as schematically shown in Fig. 1. The domain boundary \( \partial \Omega \) is composed of two disjoint subsets \( \partial \Omega_u \) and \( \partial \Omega_t \), where Dirichlet (prescribed displacement \( \bar{u} \)) and Neumann (traction vector \( \bar{t} \)) boundary conditions are applied, respectively. Also, \( \Omega \) is partitioned into three sub-domains: \( \Omega = \Omega^+ \cup \Omega^A \cup \Omega^- \), which correspond to the upper adherend, adhesive layer, and lower adherend, respectively. Since the thickness of the adhesive layer \( l_c \) is significantly smaller than other characteristic length scales of the structure, \( \Omega^A \) can be approximated as a 3D surface \( \Gamma_c \) with unit normal vector \( n \) in this model.

Corresponding to a macroscopic point \( x_M \) on \( \Gamma_c \), consider microscopic domain \( \Theta \) of the adhesive with thickness \( l_c \) and in-plane dimension \( l \) defined in the microscopic coordinate system \( x_m \) (Fig. 1). It is assumed that \( \Theta \) is locally periodic in \( x_m \) and \( x_m \) directions, which is crucial for the use of periodic boundary conditions later in the FE analysis. Relating the macroscopic domain \( \Omega \) and microscopic domain \( \Theta \) using a first-order homogenization approach, the displacement field \( \mathbf{u}(x_M, x_m) \) is decomposed into macroscopic \( \mathbf{u}_M \) and microscopic \( \mathbf{u}_m \) terms, which can be written as

\[
\mathbf{u}(x_M, x_m) = \mathbf{u}_M(x_M) + \mathbf{u}_m(x_m).
\]
The strain tensor \( \mathbf{\varepsilon} \) in the adhesive layer is then given by
\[
\mathbf{\varepsilon}(x_M, x_m) = \mathbf{\varepsilon}_M(x_M) + \mathbf{\varepsilon}_m(x_m),
\] (2)
where \( \mathbf{\varepsilon}_M = \nabla \mathbf{u}_M \) and \( \mathbf{\varepsilon}_m = \nabla \mathbf{u}_m \) are macroscopic and microscopic strain tensors, respectively. The strong form of governing equations for evaluating the deformation response of the bonded structure is then given by: Find \( \mathbf{u}_M \) and \( \mathbf{u}_m \) such that
\[
\begin{align*}
\nabla \mathbf{\sigma}_M + \mathbf{f} &= 0 \quad \text{in } \Omega^\pm \\
\nabla \mathbf{\sigma}_m &= 0 \quad \text{in } \Theta \\
\mathbf{\sigma}_M \cdot \mathbf{n} \mid_{\Gamma_c} + \mathbf{\sigma}_m \cdot \mathbf{n} \mid_{\Gamma_c} &= 0 \quad \text{on } \Gamma_c \\
\mathbf{u}_M &= \mathbf{\bar{u}} \quad \text{on } \partial \Omega_u \\
\mathbf{\sigma}_M \cdot \mathbf{n} &= \mathbf{t} \quad \text{on } \partial \Omega_t,
\end{align*}
\] (3)
where \( \mathbf{f} \) represents macroscopic body force, and \( \mathbf{\sigma}_M \) and \( \mathbf{\sigma}_m \) are macroscopic and microscopic stress tensors, respectively.

Using the linear kinematics formulation introduced in [35], the macroscopic strain tensor \( \mathbf{\varepsilon}_M \) in the adhesive layer can be approximated as
\[
\mathbf{\varepsilon}_M \approx \frac{1}{2l_c} \begin{bmatrix}
2l_c \nabla \mathbf{X}_1 \mathbf{u}_{M_1} & l_c \left( \nabla \mathbf{X}_2 \mathbf{u}_{M_1} \right) \\
l_c \left( \nabla \mathbf{X}_2 \mathbf{u}_{M_1} \right) & 2l_c \nabla \mathbf{X}_2 \mathbf{u}_{M_2}
\end{bmatrix}
\begin{bmatrix}
\mathbf{\varepsilon}_{M_1} \\
\mathbf{\varepsilon}_{M_2}
\end{bmatrix},
\] (4)
where \( \mathbf{\varepsilon}_{uM} = (\mathbf{\bar{u}}^M - \mathbf{u}_M) \) denotes the macroscopic displacement jump across the adhesive’s thickness. Note that due to the small value of \( l_c \), in-plane strain components in (4), i.e., \( \varepsilon_{M_{11}}, \varepsilon_{M_{12}}, \varepsilon_{M_{11}}, \) and \( \varepsilon_{M_{22}} \), are negligible compared to out-of-plane components. An in-depth study on the effect of in-plane strains on the failure response of adhesives is presented in [85].

In order to derive the weak form of (3), the function space \( \mathcal{V}_\Omega \) of admissible variations \( \delta \mathbf{u}_M \) is defined as
\[
\mathcal{V}_\Omega = \{ \delta \mathbf{u}_M(x_M) \mid \delta \mathbf{u}_M \subset H^1(\Omega), \delta \mathbf{u}_M = 0 \text{ on } \partial \Omega_u \},
\] (5)
where \( H^1(\cdot) \) is the first-order Hilbertian Sobolev space. Using the principle of virtual work, the weak form of (3) is then expressed as [35]
\[
\int_{\Omega^\pm} (\mathbf{\sigma}_M : \nabla \delta \mathbf{u}_M - \mathbf{f} : \delta \mathbf{u}_M) \, d\Omega + \int_{\Gamma_c} \mathbf{t} : \delta \mathbf{u}_M \, dA - \int_{\partial \Omega_t} \mathbf{t} \cdot \delta \mathbf{u}_M \, dA = 0,
\] (6)
where \( \mathbf{t} \) is the macroscopic traction in the adhesive layer. Using Hill–Mandel micro-homogeneity principle [86], stating that the potential energy density of a macroscopic point is equal to the average energy density at its corresponding microscopic domain, the macroscopic and microscopic scales can be linked together as
\[
\inf_{\mathbf{u}_M} \Phi_M(\mathbf{u}_M) = \inf_{\mathbf{\varepsilon}_M, \mathbf{\varepsilon}_m} \frac{1}{|\Theta|} \int_{\Theta} \Phi_m(\mathbf{\varepsilon}_M + \mathbf{\varepsilon}_m) \, d\Theta,
\] (7)
where \( \Phi_M \) and \( \Phi_m \) are free energy densities at each scale. The microscopic free energy density can be evaluated as
\[
\Phi_m(\mathbf{\varepsilon}) = \frac{1}{2}(\mathbf{\varepsilon}_M + \mathbf{\varepsilon}_m) : \mathbf{\sigma}_m.
\] (8)
Also, the microscopic stress tensor \( \mathbf{\sigma}_m \) is given by
\[
\mathbf{\sigma}_m = \mathbf{C} : (\mathbf{\varepsilon}_M + \mathbf{\varepsilon}_m)
\] (9)
where \( \mathbf{C} \) is the secant stiffness tensor.

Since macroscopic and microscopic displacements are independent, applying standard variational principles to (7) yields the weak form of the problem at the microscale, which can be written as: Find \( \mathbf{u}_m \) such that
\[
\frac{1}{|\Theta|} \int_{\Theta} \mathbf{\sigma}_m : \nabla \mathbf{u}_m \, d\Theta = 0, \quad \forall \mathbf{u}_m \in \mathcal{V}_\Theta,
\] (10)
where \( |\Theta| \) is the measure (volume) of \( \Theta \). Therefore, the macroscopic traction vector \( \mathbf{t} \) that results from applying the macroscopic displacement jump \( \mathbf{\varepsilon}_{uM} \) to the microscopic domain can be evaluated as
\[
\mathbf{t} = \frac{1}{|\Theta|} \int_{\Theta} \mathbf{C} \left( \frac{1}{l_c} \mathbf{\varepsilon}_{uM} + \nabla \mathbf{u}_m \right) \, d\Theta.
\] (11)
According to (11), evaluating $t$ requires solving both (6) and (10) for the macroscopic and microscopic displacement fields, which establishes the interconnection between two scales. This equation also needs to evaluate the constitutive relationship of each material phase in the microscopic domain, where the elastoplastic behavior and damage process within the epoxy matrix, and the debonding along particle–matrix interfaces will be discussed in Sections 2.2 and 2.3, respectively.

2.2. Ductile damage model for the matrix

While epoxy shows a brittle fracture behavior subject to tensile loads, it experiences a considerable plastic deformation before damage under compression or pure shear [87,88]. In order to simulate the failure response of the matrix, we implement a ductile phenomenological damage model [84,89], which is based on the concept of void nucleation, growth and further coalescence. In this model, the von Mises yield criterion and an isotropic hardening law are employed to characterize the elastoplastic behavior of the epoxy. The yield surface $f_{\sigma_{ij}}$ can be written as

$$f_{\sigma_{ij}} = q - \sigma_Y(\varepsilon_{eq}^{pl}) = 0,$$

where $q$ is the von Mises stress, $\varepsilon_{eq}^{pl}$ is the equivalent plastic strain, and $\sigma_Y(\varepsilon_{eq}^{pl})$ is the yield function.

Using the strain equivalence principle [90], the microscopic stress tensor $\sigma_m$ in the damaged material is related to the effective (undamaged) stress tensor $\sigma_{eff}$ as

$$\sigma_m = (1 - \omega)\sigma_{eff},$$

where $\omega$ is a scalar damage parameter ranging from 0 (undamaged) to 1 (fully damaged). The damage initiation occurs when the equivalent plastic strain $\varepsilon_{eq}^{pl}$ reaches a threshold value $\varepsilon_0^{pl}$, which is a function of the stress triaxiality $\eta = -p/q$ ($p$: hydrostatic stress) and equivalent plastic strain rate $\dot{\varepsilon}_0^{pl}$ as

$$\varepsilon_0^{pl} = \varepsilon_0^{pl}(\eta, \dot{\varepsilon}_0^{eq}).$$

An internal state variable $\omega_D$, which increases monotonically with the plastic deformation, is employed to track the magnitude of equivalent plastic strain $\varepsilon_{eq}^{pl}$ and determine the damage initiation when the following criterion is satisfied:

$$\omega_D = \int \frac{d\varepsilon_{eq}^{pl}}{\varepsilon_0^{pl}(\eta, \dot{\varepsilon}_{eq}^{pl})} = 1.$$ (15)

After the initiation of damage ($\omega_D = 1$), it propagates in the matrix and the material starts to lose its load carrying capacity until being fully damaged at $\omega = 1$. During this process, a stress–displacement relationship [91] is employed to mitigate the mesh dependency effect associated with the use of this continuum damage model by incorporating a characteristic length parameter $L$ in the fracture energy $G_f$ as

$$G_f = \int_{\varepsilon_0^{eq}}^{\varepsilon_{eq}^{pl}} L\sigma_Y d\varepsilon_{eq}^{pl} = \int_0^{\dot{\varepsilon}_{pl}} \sigma_Y d\dot{\varepsilon}_{pl},$$

where the effective plastic displacement $\ddot{\varepsilon}_{pl}$ is given by

$$\ddot{\varepsilon}_{pl} = L\varepsilon_{eq}^{pl}.$$ (17)

The evolution of damage parameter $\omega = \omega(\ddot{\varepsilon}_{pl})$ as a function of the effective plastic displacement is defined as an exponential relationship, which must be calibrated with experimental data. For more implementation details and the calibration procedure, refer to [84,92].

2.3. Cohesive–contact damage model

The compressive and cohesive interfacial behavior between the silica particles and epoxy matrix are simulated using a surface-based contact model and a cohesive damage model, respectively [93,94]. When material interfaces sustain compressive tractions, a hard overclosure relationship is employed in the contact model to avoid interpenetration between surfaces. This is achieved by introducing Lagrange multiplier $\lambda$ as the surface contact pressure, and
enforcing additional constraint equations [95,96], which can be expressed as
\[ \delta_n \lambda = 0, \]  
(18)
where \( \delta_n \geq 0 \) is the enforced normal separation along contact surfaces.

A cohesive damage model is used to simulate the bonding and delamination behavior of material interfaces subject to tensile tractions. In the undamaged state, the cohesive behavior along material interfaces is modeled using a linear elastic traction–separation law as
\[
\begin{bmatrix}
  t_s \\
  t_t \\
  t_n
\end{bmatrix}
= 
\begin{bmatrix}
  K_s & 0 & 0 \\
  0 & K_t & 0 \\
  0 & 0 & K_n
\end{bmatrix}
\begin{bmatrix}
  \delta_s \\
  \delta_t \\
  \delta_n
\end{bmatrix}
= K \delta,
\]  
(19)
where \( t_{\text{surf}} \) is the surface traction, \( K \) is the cohesive stiffness, and \( \delta \) is the contact separation. Here, subscripts \( s/t \) and \( n \) represent, respectively, the first/second orthogonal in-plane shear and normal components of the traction, stiffness and separation vectors.

The onset of damage along the material interface is defined as
\[
\max \left\{ t_s^0, t_t^0, \langle t_n^0 \rangle \right\} = 1,
\]  
(20)
where \( t_i^0 \) are the cohesive strength values corresponding to the damage initiation. The Macaulay brackets \( \langle \cdot \rangle \) indicate that the compressive traction normal to the contact surface does not affect the cohesive behavior. After reaching the initiation threshold given in (20), the propagation of damage along the cohesive surfaces is simulated using a scalar damage variable \( D \), which characterizes the decay of the surface traction vector as
\[
\begin{bmatrix}
  t_s \\
  t_t \\
  t_n
\end{bmatrix}
= (1 - D)
\begin{bmatrix}
  t_s \\
  t_t \\
  t_n
\end{bmatrix}.
\]  
(21)

In order to simulate the propagation of damage, we first define the effective separation \( \delta_m \) as [82]
\[
\delta_m = \sqrt{\delta_s^2 + \delta_t^2 + \langle \delta_n \rangle^2}.
\]  
(22)
Assuming a linear softening behavior during the debonding process, the evolution of the damage parameter \( D \) can be then expressed as
\[
D = \frac{\delta_m^f (\delta_m - \delta_m^0)}{\delta_m (\delta_m^f - \delta_m^0)},
\]  
(23)
where \( \delta_m^f \) and \( \delta_m^0 \) are effective separations at the complete failure and at the damage initiation, respectively.

It is worth mentioning that Park et al. [97] have shown that non-potential-based cohesive models, including the current model, could yield non-physical traction–separation paths (positive stiffness during unloading) under specific loading conditions. It has been noted in this reference that even when the traction–separation response is realistic, its oscillations could cause numerical instability and lack of convergence. While potential-based cohesive models such as the PPR model introduced in [23] can be utilized to address such drawbacks in the presence of cyclic loads (e.g., fatigue loading), we did not encounter these challenges during the implementation of the current cohesive–contact model. Here, no positive stiffness was observed in traction–separation responses of cohesive elements under monotonically increasing tension, shear, and compressive loads. Further, we were able to use this model, together with the continuum damage model described in Section 2.2 to simulate highly nonlinear failure responses of massive FE models of the adhesive microstructure with tens of millions of degrees of freedom (cf. Section 5.2).

3. Image-based microstructural modeling

3.1. Image processing and characterization

The high-fidelity simulation of the damage process in the structural adhesive studied in this work (epoxy matrix, silica particles) requires incorporating realistic microstructural features of this particulate composite in the FE model. Such features include statistical microstructural descriptors such as the volume fraction, size distribution, and spatial
Fig. 2. (a) Image processing and segmentation of the micro-CT imaging data to extract particles’ geometries as patches of NURBS functions; (b) difficulty in identifying material interfaces in a small portion of one of the 2D slices of the micro-CT data; (c) a subset of particles with acceptable morphologies stored in the shape library.

arrangement of embedded particles, as well as their morphologies, which can be extracted from 3D imaging data. Fig. 2a illustrates the high-resolution micro-CT data (voxel size: 700 nm) acquired to characterize the adhesive microstructure studied in this work. Image processing steps, including noise filtration and segmentation [98,99], were conducted on 2D slices of these imaging data to identify material interfaces after the binary representation of microstructure. The unit pixel separation approach was then employed to reconstruct a 3D voxelated image by connecting adjacent 2D slice images, followed by applying a smoothing filter to extract 3D particle morphologies.

After performing image processing, due to the complexity of the adhesive microstructure and in particular local clustering of particles, it was not feasible to properly identify several of material interfaces and thereby actual shapes of particles in micro-CT data (cf. Fig. 2a). For example, the inclusion marked as unrealistic particle in this figure is in fact composed of multiple particles, where interfaces between them have not been distinguished due to close proximity. Fig. 2b better elucidates this issue in a small portion of one of the 2D slices of the micro-CT data. Note that higher resolution imaging techniques such as nano-CT, which enables reducing the voxel size to 50 nm or less, could be employed to more realistically identify shapes of all particles. However, an increased resolution means that
only a smaller volume of the microstructure can be visualized, which may not be sufficient to reconstruct (virtually or directly) an SRVE for this material.

Despite the aforementioned challenges, the micro-CT data illustrated in Fig. 2a can still provide invaluable morphological and statistical information for the virtual reconstruction of realistic microstructural models. As shown in this figure, although several identified inclusions have unrealistic complex morphologies after the image segmentation, some of them are marked as realistic particles. Given the low volume fraction of particles in this microstructure ($V_f \approx 10\%$), such particles are away from other embedded inclusions and thereby their interfaces with the surrounding matrix can be properly identified. After selecting all realistic particles by visual inspection, their morphologies are stored in a shape library, a small subset of which is depicted in Fig. 2c. In this work, the geometry of each particle is described using multiple patches of Non-Uniform Rational B-Splines (NURBS) parametric functions [100]. A NURBS surface $C(v)$ is composed of $n$ B-splines $M^d_i(v)$ of order $d$ that are functions of parametric coordinate $v$. The shape of $C(v)$ is interpolated using a set of control points $x_i$ with weights $w_i$, which can be written as

$$C(v) = \sum_{i=1}^{n} \frac{x_i w_i M^d_i(v)}{\sum_{j=1}^{n} w_j M^d_j(v)}. \quad (24)$$

### 3.2. Statistical microstructural descriptors

In addition to the particles’ shapes, reconstructing a realistic adhesive microstructure requires taking into account statistical microstructural descriptors such as the volume fraction, size distribution, and spatial arrangement of particles. Similar to the shape library, the probability distribution functions (PDFs) associated with these statistical descriptors can be extracted from the imaging data. In order to characterize the particles’ size distribution, we employ a log-normal distribution function for radii $R_i$ of their circumventing ellipsoids as

$$f(R_i) = \frac{1}{\sqrt{2\pi} S_R} \exp\left[-\frac{(R_i - N_R)^2}{2S_R^2}\right] \quad (25)$$

where $N_R$ is the mean value and $S_R$ is the standard deviation. A two-point correlation function $S_2(r)$ is selected as PDF to characterize the spatial arrangement of particles. $S_2(r)$ yields the probability of finding two particles separated by distance $r$ in the microstructure [59,101], which statistically quantifies both the near-field and far-field spatial distributions. In order to evaluate this function for a two-phase microstructure, one must first define the indicator function $I(x)$ associated with the particle phase $\Theta_p$ as

$$I(x) = \begin{cases} 
1 & x \subset \Theta_p \\
0 & \text{otherwise}, 
\end{cases} \quad (26)$$

The two-point correlation function can then be expressed as

$$S_2(\mathbf{x}_1, \mathbf{x}_2) = \left[I(\mathbf{x}_1)I(\mathbf{x}_2)\right] \quad (27)$$

where $\mathbf{x}_1$ and $\mathbf{x}_2$ are two arbitrary points in the microstructure, and the square brackets $[\cdot]$ indicate a linear expectation operator. For a material’s microstructure that satisfies the ergodicity, statistical homogeneity, and isotropy conditions, (27) is simplified to

$$S_2(\mathbf{x}_1, \mathbf{x}_2) = S_2(|\mathbf{x}_1 - \mathbf{x}_2|) = S_2(r). \quad (28)$$

The sampling template algorithm presented in [63] is implemented for the evaluation of the two-point correlation function during the microstructure reconstruction process. In this approach, instead of tossing multiple line segments, a spherical template with $N_{tp}$ radial points and $N_{cp}$ circumferential points is randomly thrown $M$ times into the domain and the number of successful hits is recorded. Appropriate values for these sampling parameters are selected based on a convergence study to ensure that $S_2(r)$ is not affected by the values chosen for $N_{tp}$, $N_{cp}$, and $M$. It is worth mentioning that the microstructure reconstruction algorithm described next can accommodate other statistical descriptors for the spatial arrangement of particles, such as the nearest neighbor distribution [102], lineal-path [60] and Ripley’s K function [103]. Here, for the sake of computational efficiency, we have only chosen the two-point correlation function, as it statistically characterizes both the local and global arrangements of particles in the microstructure.
3.3. Microstructure reconstruction algorithm

Three main phases of the reconstruction algorithm used for synthesizing periodic repeating unit cells (RUCs) of the adhesive microstructure are schematically shown in Fig. 3. As the first step, the Random Sequential Absorption (RSA) algorithm is employed to reconstruct an initial (raw) periodic microstructure with ellipsoid-shaped inclusions. Radii of ellipsoids in this raw microstructure are selected based on $f(R_i)$, meaning each inclusion circumscribes the morphology of an actual particle stored in the shape library. In order to ensure the periodicity of the microstructure during the packing process, when a particle intersects with one of the lateral faces of RUC, a mirrored copy of that is created on the parallel face. In this work, the raw RUC is reconstructed with a volume fraction of $V_{\text{raw}} = 40\%$, which is 4 times higher than that of the final microstructure.

The next step is to replace each ellipsoid-shaped inclusion with the actual morphology of its corresponding particle from the shape library, as shown in Fig. 3b. The resulting microstructural model, which is referred as microstructure template hereafter, contains particles with realistic shapes but a higher volume fraction than the target value. Given the fact that the shapes of particles are parameterized in terms of NURBS patches, a simple orthogonal transformation is applied to control points of each NURBS function to locate the particle within its corresponding circumscribing ellipsoid.

Finally, a multi-objective GA is employed to transform the microstructure template ($V_{\text{raw}} = 40\%$) into the final microstructural model ($V_f = 10\%$) by selective elimination of certain particles. In addition to replicating the desired volume fraction, the remaining particles must replicate the target PDFs associated with their size distribution and spatial arrangement. Thus, the objective functions to be minimized in this optimization phase are defined as $L_2$-norms of the error between target and simulated (\tilde{)} PDFs, \textit{i.e.},

$$
E_1 = \|V_f - \tilde{V}_f\|_{L_2}, \quad E_2 = \|f(R_i) - \tilde{f}(R_i)\|_{L_2}, \quad E_3 = \|S_2(r) - \tilde{S}_2(r)\|_{L_2}.
$$

(29)

The GA optimization is carried out on a randomly generated initial population $\mathcal{P}_0$ of $N_p$ candidate solutions (individuals), which are evolved into a Pareto-optimal front. Each individual in $\mathcal{P}_0$ is a binary chromosome $C$ encoding
Fig. 4. Three periodic RUCs with a thickness of $l_c = 300 \, \mu m$ and different lengths of (a) $l = 300 \, \mu m$, (b) $l = 600 \, \mu m$ and (c) $l = 900 \, \mu m$ synthesized using the RSA-NURBS-GA-based microstructure reconstruction algorithm.

the particles of the microstructure template, in which 0’s and 1’s refer to their absence and presence in the final microstructure, respectively. Amongst periodic pairs of particles intersecting with lateral faces of RUC, only one of the particles is encoded in $C$ and the other one pursues its status (presence or absence) to maintain the periodicity. After the completion of the optimization phase, one of the chromosomes from the resulting Pareto-optimal front is decoded to reconstruct the final periodic RUC replicating target $V_f$, $f(R_i)$, and $S_2(r)$.

Fig. 4 shows three periodic RUCs of the adhesive layer with a thickness of $l_c = 300 \, \mu m$ and in-plane lengths of $l = 300 \, \mu m$, $600 \, \mu m$, and $900 \, \mu m$ reconstructed using this algorithm. These statistically equivalent RUCs will be analyzed in Section 5.2 to determine the optimal size of SRVE for this materials system. A comparison between simulated and distribution $f(R_i)$ and two-point correlation $S_2(r)$ functions of these microstructures and target PDFs extracted from the imaging data is provided in Fig. 5. Except for the smallest RUC ($l = 300 \, \mu m$), the small volume of which does not allow a perfect replication of particles’ size distribution and spatial arrangement, PDFs associated with the larger RUCs show an excellent agreement with the target distribution functions. It must be noted that one of the key advantages of the microstructure reconstruction algorithm introduced in this section is the computational efficiency, as the RSA-NURBS-based packing is an inexpensive process and the GA optimization phase does not require re-checking overlapping between particles. Thus, even the largest virtual RUC shown in Fig. 4 is reconstructed sequentially (no parallel implementation) on a regular iMac with a 2.7 GHz Intel Core i7 processor and 16 GB RAM.
Fig. 5. Target and synthesized statistical descriptors associated with the RUCs shown in Fig. 4: (a) size distribution $f(R_i)$, and (b) two-point correlation function $S_2(r)$.

4. Automated FE modeling

4.1. Mesh generation algorithm

Simulating the micromechanical behavior of the adhesive layer requires converting the virtual microstructures reconstructed using the RSA-NURBS-GA algorithm into appropriate FE models. In this work, CISAMR [79] is implemented in parallel to build high-quality, periodic conforming meshes for discretizing these RUCs. This non-iterative algorithm transforms a structured mesh composed of tetrahedral elements into an adaptively-refined conforming mesh in four consecutive steps, as schematically shown in Fig. 6. For completeness, a brief overview of each step is provided below:

1. **h-adaptive refinement.** A customized Structured Adaptive Mesh Refinement (SAMR) algorithm is employed to locally refine the background elements intersecting with particle–matrix interfaces and some of their neighboring elements, as shown in Fig. 6b. Main objectives of this phase are twofold: to reduce the geometric discretization error and to more accurately approximate stress concentrations that lead to damage nucleation along material interfaces. Thereby, the number of refinement levels for each particle is selected based on its size, curvature, and distance to adjacent particles. Note that applying similar levels of refinement and subdivision patterns to periodic pairs of particles intersecting with lateral faces of RUC during this process automatically guarantees the construction of a periodic mesh.

2. **r-adaptivity.** In this phase, nodes of all elements cut by material interfaces are visited to determine if they must be relocated to the interface or maintain their position. The new location of each node can be determined independently of adjacent nodes based on its proximity to the interface. As illustrated in Fig. 6c, after the completion of this non-iterative process, $\approx 50\%$ of the elements originally intersecting the material interface are transformed into conforming tetrahedrons.

3. **Face-swap.** A small fraction of conforming tetrahedrons formed along material interfaces after the completion of the $r$-adaptivity phase may have exceedingly high aspect ratios (sliver or cap shaped) due to specific patterns of relocating their nodes. Given the fixed arrangement of elements in the initial structured mesh, such degenerate tetrahedrons can be easily eliminated by swapping one of their faces with a neighboring element. Note that only the elements of each cubic block (composed of five tetrahedrons) of the background mesh containing a sliver or cap shaped tetrahedron, and one of its adjacent blocks are rearranged during this process.

4. **Sub-terahedralization.** Lastly, as shown in Fig. 6d, all remaining nonconforming elements cut by material interfaces are subdivided to build the final conforming mesh. As discussed in [79], using certain patterns for subdividing elements in this phase ensures that aspect ratios of resulting subelements do not exceed 5. Note that
cohesive elements are also created along shared faces of these conforming elements lying on opposing sides of the material interface. As shown in Fig. 6b, performing the SAMR phase leads to the creation of hanging nodes on edges of neighboring elements, which are eliminated by sub-tetrahedralization.

Fig. 7 illustrates a small portion of the periodic conforming mesh generated using CISAMR for discretizing the adhesive RUC shown in Fig. 4b. A 180 × 180 × 90 initial structured mesh is used for the construction of this mesh, where either one or two levels of $h$-adaptivity is applied along material interfaces depending on the shape and size of particles. The inset of Fig. 7 shows proper aspect ratios of conforming tetrahedrons, as well as the cohesive elements created along particle–matrix interfaces to simulate their debonding under different loading conditions. The same background element size ($h = 3.33 \, \mu m$) is employed to generate FE models for the other two RUCs and then simulate their failure responses. A mesh convergence study is presented in Section 5 to show that resulting conforming elements are sufficiently refined and do not cause mesh-dependency effects.

For the two largest RUCs containing hundreds of embedded particles, using $h = 3.33 \, \mu m$ as the background element size leads to the construction of massive conforming meshes, which would not be feasible sequentially due to memory restrictions. For example, for the RUC with $l = 0.9$ mm, the final conforming mesh is composed of approximately 33 million tetrahedral elements, which corresponds to more than 27 million degrees of freedom (DOFs). To address this issue, we have implemented the CISAMR algorithm in parallel based on a structured partitioning (i.e., cuboid-shaped partitions) of each RUC. A layer of redundant (ghost) elements is then assigned to the sub-mesh associated with each partition, allowing minimized communication between processors by implementing different phases of the CISAMR algorithm independently and only merging resulting nodes across partition faces.
afterwards. For the largest RUC ($l = 0.9$ mm), using 64 processors (Intel® Xeon® x5650, Ohio Supercomputer Center) allowed the construction of conforming mesh in 61 min.

### 4.2. Explicit dynamic FE analysis

Being a quasi-static problem in nature, the microscopic process of damage in the adhesive layer can be simulated using an implicit FE solver by incrementally applying the displacement jump $[u_M]$ on top and bottom faces of each RUC. However, the highly nonlinear behavior of this problem, involving the elastoplastic deformation and damage evolution in the epoxy matrix, as well as cohesive debonding and contact along particle–matrix interfaces, would practically prohibit the numerical convergence [104]. To address this issue, an explicit dynamic solver is utilized to simulate the failure response of RUCs, which could replicate results of a quasi-static simulation provided that the ratio of the kinetic to internal energy is negligible [105]. The dynamic equilibrium equations used in this approach can be written as

$$\ddot{u}_m(t) = M^{-1}(P^{(t)} - I^{(t)}),$$  \hspace{1cm} (30)

where $M$ is the mass matrix, and $P^{(t)}$ and $I^{(t)}$ are external and internal force at time $t$, respectively. Using the central-difference integration rule, the displacement at the next time step $u^{(t+\Delta t)}$ is given by

$$u^{(t+\Delta t)} = u^{(t-\Delta t/2)} + \frac{\Delta t}{2} \ddot{u}^{(t+\Delta t/2)} + \Delta t \dot{u}^{(t+\Delta t)},$$

$$u^{(t+\Delta t)} = u^{(t)} + \Delta t \dot{u}^{(t)} + \frac{\Delta t^2}{2} \ddot{u}^{(t)},$$  \hspace{1cm} (31)

where $\Delta t^{(t)}$ is time increment at time step $t$. Therefore, the strain and stress at every integration point can be evaluated as

$$e_m^{(t+\Delta t)} = \frac{u_m^{(t+\Delta t)} - u_m^{(t)}}{|\Theta|}, \quad \sigma_m^{(t+\Delta t)} = f(\sigma_m^{(t)}, e_m^{(t)}, e_m^{(t+\Delta t)}).$$  \hspace{1cm} (32)

### 5. Calibration and SRVE size study

#### 5.1. Calibration of continuum and cohesive damage models

Before using the reconstruction-CISAMR framework to investigate the effects of pre-existing voids, adherend surface roughness, and particle–matrix interfacial bonding on the failure response of the adhesive layer, the material parameters used in the FE model must be calibrated with experimental data. The heterogeneous adhesive consists of two phases: an epoxy matrix with elastic modulus $E_m = 3.9$ GPa and Poisson’s ratio $\nu_m = 0.39$, as well as embedded silica particles with $E_p = 71.7$ GPa and $\nu_p = 0.17$ [83,106].

The parameters used in the continuum ductile damage model characterizing the initiation and evolution of damage in the matrix are determined by simulating the stress–strain curves of a homogeneous 3D FE model with appropriate

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**Fig. 7.** Small portion of the periodic conforming mesh generated using CISAMR corresponding to the inbox of RUC shown in Fig. 4b.
Fig. 8. Comparison between experimentally measured [83] and numerically simulated stress–strain curves of the epoxy matrix subject to tensile, compressive, and shear loadings after the calibration of the ductile damage model.

Table 1
Calibrated plastic yield function used in the continuum ductile damage model. The yield stress $\sigma_Y$ values are given in MPa.

<table>
<thead>
<tr>
<th>$\sigma_Y$</th>
<th>29.0</th>
<th>37.0</th>
<th>52.2</th>
<th>84.8</th>
<th>95.3</th>
<th>96.3</th>
<th>97.8</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\bar{\varepsilon}_{pl}^0$</td>
<td>0.0</td>
<td>4.946e$-$4</td>
<td>1.496e$-$3</td>
<td>1.009e$-$2</td>
<td>2.274e$-$2</td>
<td>2.720e$-$2</td>
<td>1.417e$-$1</td>
</tr>
</tbody>
</table>

5.2. Determining the size of SRVE

In order to determine the appropriate SRVE size, failure responses of three synthesized RUCs of the adhesive layer with similar thicknesses of $l = 300$ µm but different in-plane dimensions of $l = 300$ µm, 600 µm, and 900 µm (cf. Fig. 4) are analyzed. After creating FE models using CISAMR, explicit dynamic simulations are conducted to evaluate the failure response under different types of loadings. Resulting macroscopic traction–separation curves of RUCs subject to tensile and compressive displacement jumps $\|u_{Mj}\|$ are illustrated in Figs. 9a and 9b, respectively. While the tensile results are nearly insensitive to the RUC size, for the compressive load, the smallest RUC with $l = 300$ µm clearly under-predicts the strength and ductility of the adhesive. The negligible difference between traction–separation responses of RUCs with $l = 600$ µm and 900 µm under compression in Fig. 9b suggests that the former can be selected as SRVE.

Simulated damage patterns after failure in RUCs with $l = 300$ µm and 900 µm subject to tensile and compressive loads are illustrated in Figs. 10 and 11, respectively. Note that the use of periodic meshes, together with periodic

boundary conditions to replicate the results of tension, compression, and shear tests presented in [83,107]. A comparison between these experimental results and corresponding FE simulations after the calibration of the damage model is depicted in Fig. 8, showing a good agreement for all three types of loadings. This has been achieved using thresholds $\bar{\varepsilon}_{pl}^0 = 0.033$, 0.13, and 0.27 for the equivalent plastic strain before damage initiation subject to tensile, compressive and shear loading, respectively. The effective plastic displacement $\bar{\varepsilon}_{pl}$ allowed before complete damage (mechanical failure) are 0.01 µm (tension), 0.1 µm (compression), and 0.2 µm (shear). Also, the plastic yield function $\sigma_Y(\bar{\varepsilon}_{pl}^0)$ used in the FE simulations is given in Table 1.

The surface-based cohesive damage model characterizing the debonding along silica–epoxy interfaces is calibrated based on experimental peel tests and molecular dynamics (MD) simulations presented in [108,109]. The calibrated material parameters include the cohesive stiffness $K_n = K_s = K_t = 4.54 \times 10^5$ GPa/m, adhesion strength $t^0_n = t^0_s = t^0_t = 33.5$ MPa, and effective separation after the damage initiation $\delta^f_m - \delta^0_m = 0.5$ µm. A small bulk viscosity ($b_1 = 0.06$ and $b_2 = 1.2$) is also used in the explicit FE solver to introduce negligible damping to the system, which can control high frequency oscillations in the simulation and hence facilitate the convergence.
boundary conditions, prohibits the unrealistic accumulation of damage along lateral faces of RUCs. Also, the damage pattern leading to failure is within the RUC volume and not along the adherend surfaces, indicating a cohesive-type (versus an adhesive-type) failure mode under both loading conditions. The complex damage surfaces illustrated in Figs. 10 and 11 initiate at sites of stress concentrations along particle–matrix interfaces, then propagate and coalesce within the epoxy matrix. More information regarding damage mechanisms within the adhesive microstructure under different types of loading is provided in Section 6.

To ensure that \( l = 600 \mu m \) is indeed an appropriate size for the SRVE, in addition to comparing traction–separation curves associated with RUCs with various sizes (cf. Fig. 9), we study the failure responses of two other RUCs with the same size but with distinct microstructures. These RUCs, which have statistically equivalent periodic microstructures similar to that of the original RUC with \( l = 600 \mu m \) depicted in Fig. 4b are virtually reconstructed using the RSA-NURBS-GA algorithm described in Section 3.3. After construction of conforming meshes using CISAMR, the resulting damage patterns after failure within the microstructure of these RUCs subject to an out-of-plane compressive load is depicted in Fig. 12. A comparison between corresponding traction–separation responses is also provided in
Fig. 11. Damage patterns after failure in two RUCs with (a) \( l = 300 \, \mu m \) and (b) \( l = 900 \, \mu m \) subject to a macroscopic compressive load. Corresponding points on the macroscopic traction–separation responses are labeled similarly in Fig. 9b.

Fig. 12. (a–c) Damage patterns after failure subject to a macroscopic compressive load in statistically equivalent RUCs 1 (original), 2, and 3 with \( l = 600 \, \mu m \); (d) comparison between macroscopic traction–separation responses associated with each RUC.

Fig. 12d, where negligible differences between them (\(< 0.5\% \) in strength and fracture energy) verifies that RUC with \( l = 600 \, \mu m \) can properly serve as SRVE for this adhesive system.

Before we proceed to analyze the impact of various microstructural features on the failure response of SRVE, it is worth examining the impact of mesh structure on the convergence of simulation results. To ensure that conforming
Fig. 13. (a) Damage pattern after failure in the smallest RUC ($l = 300 \mu m$) subject to a compressive load, which is simulated using an FE model generated using CISAMR on a fine background mesh with $h = 2 \mu m$; (b) small portion of the corresponding conforming mesh; (c) comparison between the resulting macroscopic traction–separation response and that of the original FE model ($h = 3.33 \mu m$).

meshes generated using CISAMR are sufficiently refined to avoid mesh-dependency, we have simulated the failure response of the smallest RUC ($l = 300 \mu m$) under compression using an FE model generated using CISAMR on a finer background mesh with $h = 2 \mu m$. The simulated damage pattern after failure in this RUC and a small portion of the new conforming mesh used for discretizing that are depicted in Fig. 13a and b, respectively. The resulting macroscopic traction–separation response is compared to that obtained using the original mesh ($h = 3.33 \mu m$), whose nearly identical responses indicate the appropriateness of the latter without imposing an exceedingly high computational cost.

6. Results and discussions

In this section, we investigate the effects of the adherend surface roughness and microstructural defects on the failure response of SRVEs of the adhesive layer subject to various loading conditions. As a benchmark for comparison purpose, failure responses of SRVEs under the (unrealistic) assumption of perfectly strong interfacial bonding between embedded particles and the matrix are also studied. Note that in order to carry out such simulations, we simply remove the cohesive elements and surface contact along material interfaces in the FE model.

6.1. Effect of tensile load

Three snapshots of the initiation and evolution of damage within the original SRVE with no defects (no pre-existing voids, no surface roughness, but weak particle–matrix interfacial bonding) subject to a macroscopic tensile load are illustrated in Fig. 14. The solid view of the damage propagation corresponding to the inbox depicted in Fig. 14a is also shown at each stage of damage. According to this figure, the damage initially nucleates along particle–matrix interfaces, specially when the axis connecting high curvature regions of silica particles is aligned with the $x_{m_1}x_{m_2}$-plane, i.e., perpendicular to the loading direction. This is due to higher stress concentrations in these regions, which are magnified after particles are debonded from the surrounding matrix (cohesive damage). As shown in Fig. 14b, the damage then radially propagates around each particle, approximately parallel to the $x_{m_1}x_{m_2}$-plane. At the failure point, the damage zones formed at different heights in through-the-thickness direction are merged together, which form a jagged failure surface, as shown in Fig. 14c.

Before analyzing the effects of voids and surface roughness on the mechanical behavior of the adhesive layer, it is worthwhile to investigate the impact of particle–matrix interfacial bonding on the SRVE’s failure response. For this purpose, although physically unrealistic, we consider an SRVE with strong bonding between these phases. Fig. 15 shows a comparison between different stages of damage in SRVEs with strong and weak interfaces, as well as corresponding macroscopic traction–separation responses, indicating the significant impact of interfacial bonding strength on the failure response. For example, although at $\|u_{M_1}\| = 5.3 \mu m$ the damage has already initiated in the SRVE with weak particle–matrix interfacial bonding, the SRVE with strong (perfect) bonding is undamaged. Also,
Fig. 14. FE simulation of the initiation and evolution of damage in intact SRVE subject to a macroscopic tensile load. The right column shows a small portion of the damaged SRVE corresponding to the inbox shown in figure a.

According to Fig. 15(b,e), while the former case has reached mechanical failure at $\|u_M\|=8.4\ \mu m$, the latter is in early stages of damage initiation. As illustrated in Fig. 15c, the SRVE under the assumption of strong silica–epoxy bonding experiences failure at a larger displacement jump ($\|u_M\|=9.6\ \mu m$) and has a thicker, more tortuous damage pattern in the matrix compared to the case with weak bonding (Fig. 15e). This corresponds to a higher absorbed strain energy before the failure, as well as an approximately 35% increase in the strength, as depicted in Fig. 15f. Although not feasible to achieve a perfect bonding between silica and epoxy, this study suggests that appropriate surface treatment of particles to increase this interfacial bonding strength could considerably improve the failure response of the adhesive subject to a tensile load.

The simulated damage pattern in an SRVE with non-smooth adherend surfaces (weak silica–epoxy bonding) is illustrated in Fig. 16. The FE model for this simulation is built by incorporating a thin layer of steel (AISI 302) adherend, with an elastic modulus of $E_s = 180$ GPa and Poisson’s ratio of $\nu_s = 0.30$. Note that cohesive elements are not incorporated along adherend–adhesive interfaces, as debonding along this surface (adhesive failure) is often
Fig. 15. Initiation and evolution of damage in intact SRVEs with (a–c) strong and (d,e) weak bondings along particle–matrix interfaces subject to a macroscopic tensile load; (f) corresponding traction–separation response of each SRVE.

not the dominating failure mode and thereby not considered in this study. Fig. 16 shows a very similar damage pattern throughout the thickness of the adhesive layer as that of the original SRVE (smooth surface) depicted in Fig. 14c, indicating that the morphology of adherend surfaces has a negligible impact on the failure response. In particular, no damage nucleation is observed along the non-smooth adherends–adhesive interfaces in this case. Note that small differences between damage patterns corresponding to SRVEs with smooth and rough adherend surfaces are mainly due to the threshold used for hiding the undamaged matrix phase during the visualization. This observation is confirmed by the negligible difference between macroscopic traction–separation responses of these SRVEs, as shown in Fig. 17a. The fact that the adherends’ surface morphology does not affect the failure response suggests that, upon identical surface preparation (e.g., plasma treatment) to achieve the same chemical bonding strength, a mild surface roughness could improve the overall mechanical integrity of the joint by increasing the bonding area of adhesive–adherend interfaces.

The simulated damage pattern after the failure in an SRVE with a $V_f^{\text{void}} = 2\%$ volume fraction of randomly distributed ellipsoid-shaped voids subject to a tensile load is depicted in Fig. 18. Although this damage pattern is
Fig. 16. Damage pattern after failure in SRVE with surface roughness subject to a macroscopic tensile load.

Fig. 17. (a) Macroscopic traction–separation responses of SRVEs with no defects, with pre-existing voids, and with surface roughness subject to a macroscopic tensile load. The points labeled (a,b,c) correspond to three stages of damage in the intact SRVE shown in Fig. 14; (b) comparison of traction–separation responses of SRVEs with weak and strong bondings along particle–matrix interfaces.

slightly different than that of the intact SRVE (Fig. 14c). Fig. 18b clearly shows that even in the presence of voids, high curvature regions of particle–matrix interfaces still serve as primary sites of damage nucleation. Thereby, the macroscopic traction–separation responses illustrated in Fig. 17a show less than 0.5% decrease in the strength and the absorbed energy. In other words, a small volume fraction of voids in the microstructure, which is practically inevitable during accelerated manufacturing processes of real-world structures, has a negligible impact on the failure response of this heterogeneous adhesive.

The analysis presented in the paragraph above might seem to be counter-intuitive at first glance, as pre-existing voids in the adhesive layer are manufacturing defects that are expected to undermine its mechanical integrity. This expectation is in accordance with some experimental and numerical studies available in the literature for other types of composite materials, which show a significant reduction in the strength and fracture toughness due to such flaws [110,111]. However, the traction–separation curves presented in Fig. 17a can be justified taking into account the
Fig. 18. (a) Damage pattern after failure in SRVE with pre-existing voids subject to a macroscopic tensile load; (b) small portion of the damaged SRVE corresponding to the inbox shown in figure a.

relatively weak interfacial bonding between silica particles and epoxy matrix of the heterogeneous adhesive studied in this work. The debonding between these phases leads to magnified stress concentrations and plastic deformations in the matrix along sharp edges of particles, which are even more pronounced than those caused by ellipsoid-shaped voids. Thereby, even in the presence of voids, particle–matrix interfaces still serve as primary sites of damage nucleation in the microstructure subject to a tensile load.

In order to verify this claim, Fig. 17b compares the failure responses of SRVEs containing pre-existing voids with weak and strong particle–matrix interfacial bondings. Note that the presence of voids ($V_{\text{void}} = 2\%$) considerably reduces both the strength and toughness of the SRVE with strong interfacial bonding compared to a similar SRVE with no defects, although its strength and ductility are still higher than those of the original SRVE (actual adhesive) with weak silica–epoxy interfaces. A one-to-one comparison of evolving damage patterns in a small portion of defective SRVEs with strong and weak interfacial bondings is illustrated in Fig. 19, which better elucidates distinct damage mechanisms in these SRVEs: while in the former the damage initiates in the vicinity of voids, in the actual SRVE, the damage nucleates at an earlier stage along delaminated particle–matrix interfaces. It is worth mentioning that this discussion on the impact of particle–matrix adhesion is in accordance with the results of a similar experimental study presented in [112] for a particulate polymer composite.

6.2. Effect of compressive load

Two snapshots of the evolving damage pattern in the original SRVE subject to a compressive load are shown in Fig. 20, which correspond to points (a) and (b) on its macroscopic traction–separation response depicted in Fig. 21a. Unlike the case of tensile loading, where the damage initiates along the surfaces of a few particles and then immediately propagates in the matrix (semi-brittle failure), here the matrix surrounding more than half of the particles is significantly damaged with a limited propagation away from those particles at $\|u_{\text{M}_3}\| = -8.25 \mu\text{m}$. This localized accumulation of damage accounts for the nonlinear hardening in the traction–separation response of the SRVE before reaching the peak traction (Fig. 21a), which is not observed for the case of tensile loading. Note that in addition to the punching effect of upper and lower regions of particles into the matrix under compression, the damage localization along particle peripheries is highly enhanced by magnified stress concentrations caused by their debonding from the matrix. Also, Fig. 20a shows that most damage sites in the SRVE correspond to regions that two or more particles are in close proximity due to higher stress concentrations, while the matrix surrounding isolated particles experiences less damage. This re-emphasizes the importance of incorporating the actual spatial arrangement of particles in synthesized adhesive SRVEs (using the RSA-NURBS-GA algorithm) for the reliable prediction of its failure response. Further, the final damage pattern in this SRVE is depicted in Fig. 20b, showing a jagged failure surface formed by the coalescence of particle-induced damage sites.

Fig. 21a clearly shows that the presence of adherend surface roughness or voids has practically no impact on the failure response of the adhesive layer subject to a compressive load. The damage pattern after the failure in
Fig. 19. Initiation and evolution of damage in SRVEs with pre-existing voids subject to a macroscopic tensile load: (a,b) strong and (c,d) weak bonding along particle–matrix interfaces.

the former is depicted in Fig. 21b, indicating that no damage is nucleated along upper and lower surfaces of the adhesive layer bonded to steel adherends. The damaged SRVE with pre-existing voids after the failure is depicted in Fig. 22, showing the negligible effect of voids on the damage pattern in the matrix, which is highly governed by particle-induced damages. Both these observations are similar to those reported for SRVEs subject to a tensile load in Section 6.1, although even the small differences between traction–separation responses of SRVEs with and without defects in that case are vanished here. Fig. 21a also compares the failure responses of the original SRVE (weak particle–matrix interfaces) with that of a similar SRVE with strong interfacial bonding. According to this figure, the strength and toughness of this imaginary composite are approximately 33% and 59% higher than those of the actual adhesive (weak interfacial bonding), respectively. Similar to the case of tensile loading, this study shows the significant importance of the interfacial adhesion strength of embedded heterogeneities with the surrounding matrix on the failure response of the adhesive layer.

6.3. Effect of shear load

A similar study as those presented in Sections 6.1 and 6.2 on the impacts of pre-existing voids and adherend surface roughness on the failure response is performed here considering a shearing displacement jump $\|\mathbf{u}_{M_2}\|$. The simulated damage pattern in the SRVE with pre-existing voids after the failure is depicted in Fig. 23, showing that no damage has nucleated and propagated from these defects. This damage pattern is nearly identical to that of an SRVE with no defects. Similar to the cases of tensile and compressive loads, here the weak interfacial bonding along particle–matrix interfaces and the high curvature of particles govern the principal damage surface. However, unlike those case scenarios, this damage surface is not tortuous and is confined within a thin layer that is parallel to the $x_{m1}x_{m2}$-plane. Due to the negligible contribution of voids on the damage evolution in the matrix, as shown in Fig. 24a, the resulting macroscopic traction–separation response is only slightly different than that of an intact SRVE with no pre-existing voids. Also, note the considerably higher ductility ($\|\mathbf{u}_{M_2}\| = 36 \mu m$) but lower strength ($t_2 = 56$ MPa)
Fig. 20. FE simulation of the initiation and evolution of damage in the intact SRVE subject to a macroscopic compressive load.

Fig. 21. (a) Macroscopic traction–separation responses of SRVEs with no defects (weak and strong particle–matrix interfacial bondings), pre-existing voids, and surface roughness subject to a macroscopic compressive load. The points labeled (a,b) correspond to two stages of damage in the intact SRVE shown in Fig. 20; (b) damage pattern after the failure in SRVE with surface roughness.

of the adhesive layer subject to a shear load compared to SRVEs under tensile and compressive loads, which sustain $\|u_{M3}\| = 6.2 \mu m$, $t_3 = 126$ MPa and $\|u_{M3}\| = -9.7 \mu m$, $t_3 = -186$ MPa, respectively. The significant nonlinear
response of the adhesive (strain hardening) before reaching the peak shearing traction is another distinct behavior of this SRVE compared to previous types of loadings.

The effect of the adherend surface roughness on the macroscopic traction–separation response of the adhesive layer subject to a shear load is illustrated in Fig. 24a. This figure shows a negligible decrease in the strength compared to an intact SRVE but a more notable impact on the fracture toughness than the cases of tensile and compressive loadings. The corresponding damage pattern in this SRVE is depicted in Fig. 24b, which is different from that of SRVEs with smooth adherend surfaces (with or without pre-existing voids). Unlike the latter cases, where the failure surface develops in the middle of the SRVE (Fig. 23), the non-smooth morphology of adherend surface induces a horizontal failure surface in the proximity of the lower face of SRVE. The matrix near the upper adherend’s surface is significantly damaged as well. Note that the principal damage surface is not exactly along the non-smooth lower surface, meaning the adhesive still experiences a cohesive failure mode subject to a shear load. However, amplified stress concentrations along interfaces of nearby particles lead to the nucleation and eventually growth of damage in these regions, which are different than the damage pattern observed in SRVEs with smooth surfaces. Despite distinct damage patterns, according to Fig. 24a, the presence of surface roughness does not significantly undermine the mechanical integrity of the joint subject to a shear load. Thereby, the increased adhesive-adherend bonding area due to the presence of this mild adherend surface roughness could still be beneficial by avoiding the adhesive-type failure.
Fig. 24a also illustrates the effect of strong particle–matrix interfacial bonding on the traction–separation response of the adhesive subject to a shear load. Unlike the tensile and compressive loading case scenarios, where the bonding strength has a significant impact on the adhesive’s failure response, here it only increases the strength and fracture toughness by 2% and 10%, respectively. This can be attributed to lower stress concentrations developed in the matrix surrounding particles compared to previous two loading cases, allowing a considerably higher plastic deformation before failure. This in turn mitigates the impact of particle–matrix interfacial strength on the failure response, as less debonding is observed along material interfaces during the initiation and evolution of damage in the matrix under shear load.

Note that the final damage surfaces shown in Figs. 23 and 24 are parallel to the $x_{m1}$-$x_{m2}$ plane, which correspond to the direction of the maximum shear stress under the mode II loading condition. On the other hand, the damage surfaces in the previous two case scenarios (tensile and compressive loads) are in the direction of the maximum tensile stress. This is mainly due to the significant plastic deformation occurred subject to a shear load (cf. Fig. 24a), which leads to a shear stress induced ductile failure in the matrix. This behavior is in contrary to that observed under tensile loading, where the brittle damage in the matrix caused by the maximum normal stress dominates the failure process. Although a more ductile failure is observed subject to a compressive load due to the initial punching of particles into the epoxy matrix, subsequent propagation of damage in the matrix is also a relatively brittle phenomenon.

7. Conclusion

A comprehensive numerical study was presented to illuminate the effects of pre-existing microstructural defects, adherend surface roughness, and particle–matrix interfacial bonding strength on the failure response of a heterogeneous structural adhesive. In order to carry out this study, a set of micro-CT images of the adhesive was processed to extract morphological features and statistical microstructural descriptors needed to reconstruct its microstructure. A NURBS-based reconstruction algorithm relying on RSA and GA was then utilized to synthesize realistic RUCs of the adhesive layer. These RUCs were transformed into appropriate periodic FE models using a parallel non-iterative mesh generation algorithm named CISAMR. The failure response of each RUC was approximated using an explicit dynamic FE solver relying on a phenomenological continuum ductile damage model for the epoxy matrix and a cohesive–contact model to simulate interfacial damage between the particles and the matrix. After calibrating damage models with experimental data, we identified the appropriate size of statistical SRVE for this adhesive system. It must be noted that this study, as well as the subsequent analysis on impacts of various microstructural features on the macroscopic traction–separation response involved simulating failure responses of tens of RUCs with realistic 3D microstructures. Such high-fidelity studies, which were not carried out in previous modeling efforts on this topic, were not feasible without implementing the automated computational framework presented in this work. Main outcomes of the study
aimed at linking the microstructure to the mechanical behavior of the adhesive subject to tensile, compressive, and shear loads are summarized below:

- Weak interfacial bonding between embedded particles and matrix could significantly undermine the strength and fracture toughness of the adhesive under tensile and compressive loads.
- While pre-existing voids considerably deteriorate the mechanical integrity of an adhesive with perfectly strong particle–matrix interfaces, they have a negligible impact on the failure response of the actual adhesive material with weak interfacial bonding.
- Similarly to pre-existing voids, a mild roughness of adherend surfaces does not cause a significant deterioration of the strength and ductility of the adhesive layer. Thus, a non-smooth adherend surface could improve the overall mechanical integrity of the joint by avoiding adhesive-type failure through increasing the adhesive-adherend bonded surface area.

Acknowledgments

This work has been supported by the Air Force Office of Scientific Research (AFOSR) under grant number FA9550-17-1-0350 and the Ohio State University Simulation Innovation and Modeling Center (SIMCenter) through support from Honda R&D Americas, Inc. The authors also acknowledge the allocation of computing time from the Ohio Supercomputer Center.

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