Influence of Fiber Packing on the Deformation and Damage Development in Fiber Reinforced Laminates during Curing

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The influence of fiber packing on the deformation and damage evolution, during the process of manufacturing and subsequently, in fiber reinforced composites is investigated by considering a cluster of fibers within a matrix that is undergoing curing and subsequently subjected to mechanical loads. The residual stress development due to cure shrinkage and polymerization of the matrix, during the curing process, is modeled using the finite element (FE) framework. The model is based on the notion of polymer networks that are continuously formed in a body of changing shape due to changes in temperature, chemistry and external loads. Nonlinear material behavior due to post-damage response that can lead to cracking in the matrix is incorporated through a crack band model that preserves mesh objectivity in the FE calculations. The combined thermo-chemo-mechanical response of the composite due to curing and subsequent mechanical loading is investigated in detail in this paper.

I.   INTRODUCTION

Fiber reinforced polymer matrix composites (FRPC) are manufactured using a variety of manufacturing processes, predicated on the type of FRPC considered. During this process, the constituents undergo thermo-chemo-mechanical response and their states become altered. Therefore, for better designing of these laminates, a thorough understanding of the material state is required, and the influence of processing on the subsequent material response in service needs to be quantified. Along with design aspects like geometrical properties and layup, manufacturing induced effects have to be accounted for. In order to be able to predict the final properties of a "processed" structural material, a computational model that is used to study deformation response, must also include processing induced effects.

FRPCs are modeled in this paper. Previous studies by Song and Waas[1] on dry fiber FRPC manufactured using a VARTM process have shown that the use of original "virgin" matrix properties in numerical predictions can lead to inaccurate determination of the composite response to loading. Also, work by Rabearison et al.[2] showed that cracks are developed due to gradients in cure thick carbon epoxy tubes. The virgin matrix material showed different behavior as opposed to *in-situ* matrix material behavior obtained from experiments.[3] That is, the presence of fibers in a laminate and the curing cycle alter the behavior of the matrix during the curing process. Defect formation due to constrained curing of epoxy has also been demonstrated by Plepys and Ferris[4] and Chekanov et al.[5] Basically, the system undergoes shrinkage due to chemical processes, and thus, builds internal stresses. Depending on the constituent chemistry and thermal cycle that is used during cure, a fiber reinforced composite can and may undergo damage and cracking during the curing process, most likely due to gradients in stress and strain fields generated during cure, resulting in significant residual stresses. Therefore, it is important to account for curing induced effects on the response of the final laminate used in component design. In this paper, the *evolution* of temperature and the degree of cure for the matrix material system is determined through a coupled system that considers the heat equation and an empirical curing law.[6]

To investigate the effects of the curing process on the ultimate material behavior, the effects of geometry (packing details) and how these influence the pre-service stress distribution, and possible damage formation

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through cracking, is studied. The residual stresses that develop are a function of the cure progression in the system, and this progression which is related to the temperature field is also influenced by the packing details. Therefore, the prediction of temperature distribution, cure evolution and residual stress development in fiber reinforced laminates are all coupled, and this aspect is investigated in this paper.

Fiber-matrix microstructural models are constructed, with discrete fibers embedded in matrix, with 4 different fiber packing in 2D. The evolution of temperature, and the degree of cure is determined for these models, followed by solidification or curing of the epoxy. After curing, the models are subjected to tensile loading to determine the corresponding load-displacement responses, and the influence of curing on the responses is investigated.

II.   FIBER-MATRIX UNIT CELL

Four 2D representative unit cells (RUCs), with randomly packed fibers and a fixed fiber volume fraction are modeled as shown in Fig. 1. These models are subjected to temperature change that represents a typical cure cycle, and the curing process is simulated. Subsequently, the RUCs are subjected to tensile loading to determine the load-displacement responses of the cured models.
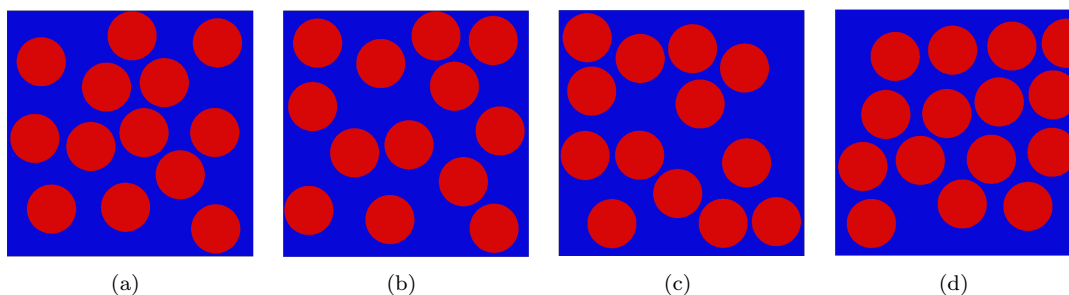

(a)   (b)   (c)   (d)

Figure 1. 2D Fiber-Matrix Unit Cells with Randomly Packed Fibers (Blue: Matrix, Red: Fiber) with Fiber Volume Fraction of 50 %

The fibers are modeled as transversely isotropic linear elastic material. The matrix material is initially a liquid Epon862 resin which is cured to form a solid material. The material properties of the fiber-matrix system is obtained from the paper by Heinrich et al.[3]

III.   EVOLUTION OF TEMPERATURE AND DEGREE OF CURE

Curing of Epon862 matrix material with curing agent Epikure9553 is modeled here. The degree of cure in the matrix is given by,

$$\phi(t) = \frac{H(t)}{H_r} \tag{1}$$

where, H(t) is the heat generated and $\phi(t)$ is the degree of cure at time t. The process of curing of an epoxy is modeled by a curing kinetics equation of a highly exothermic chemical reaction. Equation for a one way coupled non-linear partial differential equation for spatial and temporal variation of temperature $T(x,t)$ and the corresponding degree of cure $\phi(x,t)$ is,

$$\rho c_p \frac{\partial T}{\partial t} = \frac{\partial}{\partial x}(K\frac{\partial T}{\partial x}) + \rho \frac{d(\phi(t)H_r)}{dt} \tag{2}$$

The degree of cure as a function of time for Epon862 and Epikure 9553 (given in Heinrich et al.[3]) was obtained experimentally using Raman light scattering (RLS). A non-linear least squares approach was used to fit the experimental data. The above partial differential equation is solved for the models to determine the temperature distribution over the entire cure cycle.

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IV. EVOLUTION OF STRESSES DURING CURING

A model for evolution of stress given in Heinrich et al.,[3] incorporating the concept of network formation and its contribution to stress evolution is used here. The epoxy resin, which is the matrix used, is initially mixed with a hardener in liquid state. The mixture is poured over fibers in a mold. The pure epoxy starts to harden upon mixing through the formation of cross links. The entire configuration is stress free initially, and at time t_1, the first network forms, which is accompanied by cure shrinkage, along with additional strains due to external loads or other thermal effects. A second network forms at a later time t_2, but with a different reference configuration, since some networks have already formed at time t_1. The process of network formation continues, and the reference configuration is continually changing as time evolves. But, the current configuration is the same for all the networks, since they occupy the same volume. This results in different stresses in each network. The externally applied loads are balanced out by the sum of stresses in all of the networks formed at any time.

A 1D formulation of the stress evolution is derived similar to the derivations given in Mei[7] and Hossain et al.[8] The stress $\sigma(t,s)$ at time t in a network formed at time t_1, which is proportional to the mechanical strain $\epsilon_{mech}(t,s)$ relative to its stress free configuration with a proportionality constant $E(s)$, which is the elastic modulus of the network, is obtained as,

$$\sigma(t) = \int_0^t \frac{d\phi}{ds} E(s)(\epsilon(t) - \epsilon(s) - \alpha(T(t) - T(s)) + \epsilon_c(s))ds \tag{3}$$

where, $\alpha(s)$ is the coefficient of thermal expansion and $\epsilon_c(s)$ is the cure shrinkage of the network formed at time s. The above equation can be generalized to 3D as given in Heinrich et al.:[3]

$$\begin{aligned}
\underline{\sigma} = \int_0^t \frac{d\phi}{ds} \underline{1}[&K(s)tr(\underline{\epsilon}(t) - \underline{\epsilon}(s) + \underline{\epsilon}_c(s) - \underline{1}\alpha(s)\Delta T(t,s)) \\
&+ 2\mu(s)(\underline{\epsilon}(t) - \underline{\epsilon}(s) + \underline{\epsilon}_c(s) - \underline{1}\frac{1}{3}tr(\underline{\epsilon}(t) - \underline{\epsilon}(s) + \underline{\epsilon}_c(s))) \\
&+ (1 - \phi(t))K_{liquid}\underline{1}(\underline{\epsilon}(t) - \underline{1}\alpha_{liquid}\Delta T(t))]
\end{aligned} \tag{4}$$

where, K, μ, α and $\underline{\epsilon}_c$ are the per network bulk modulus, shear modulus, coefficient of thermal expansion and cure shrinkage, respectively, and are inputs to the model. These per network input parameters are extracted from the plane wave modulus, shear modulus and degree of cure determined experimentally for the entire epoxy system. The variation of the elastic modulus E with respect to degree of cure ϕ is shown in Fig. 2.
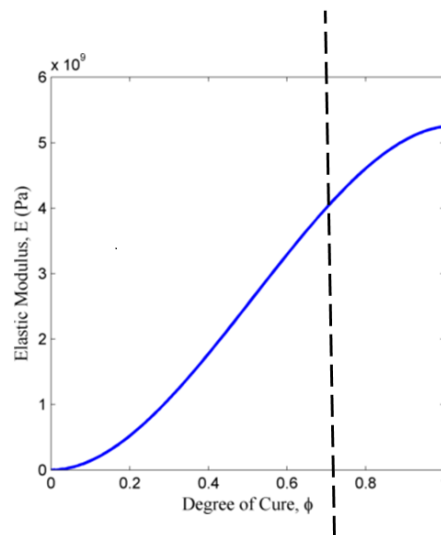

Figure 2. Variation of Elastic Modulus with respect to the Degree of Cure

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V.   MODELING DAMAGE DUE TO CURE SHRINKAGE

Due to excessive cure shrinkage and residual stress build up during the curing process, the epoxy material can degrade in the presence of fibers. Therefore, stress evolution should account for the possibility of progressive damage during the curing process. A new methodology is formulated to incorporate possible damage evolution in the matrix during curing. An extra dimension is added to the variation of the material properties. That is, the instantaneous material properties are assumed to be function of the degree of cure (ϕ) as well as the amount of strain accumulated at a material point. The properties are degraded through a physics based approach. The degradation of the material is governed by the amount of energy dissipated during the damage process governed by a cohesive law. That is, the material properties are degraded over a process zone. Thus, the elastic modulus which was originally considered as a function of ϕ only as shown in Fig. 2 is now generalized as shown in Fig. 3.
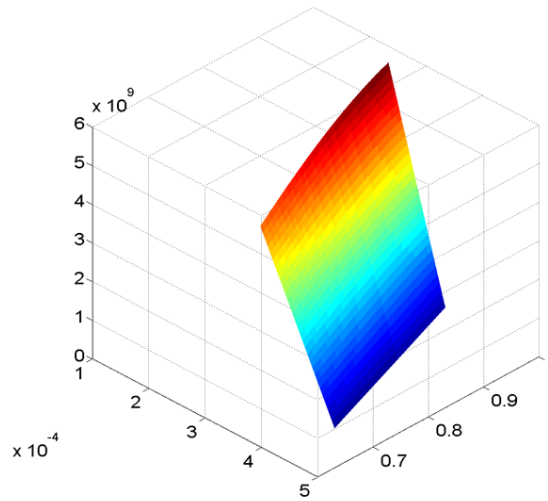

Figure 3. Variation of Elastic Modulus with respect to the Degree of Cure and Strain at a Material Point

The crack band approach given in Bazant and Oh[9] is incorporated within the curing framework to account for damage induced due to cure shrinkage. A typical stress-strain material law is shown in Fig. 4(a). It is assumed that the material starts to degrade (or stiffness reduces) beyond a critical stress (σ_0) value corresponding to varying degree of cure. This critical stress is a function of the degree of cure. Thus, here, it is assumed that the strength of the material is a function of the degree of cure. Further, the fracture energy released upon complete failure of the material is also dependant on the degree of cure. It is assumed that the fracture energy associated with a material that has a smaller degree of cure (more ductile) is higher than that of a material with a higher degree of cure. Clearly, these assumptions need to be verified by carrying out fracture tests on the matrix material cured to different degrees. The assumption adopted in this paper is that fracture energy (G_C) is not constant, but reduces linearly with the degree of cure, (ϕ). By adopting the crack band model, the damage is introduced in a mesh objective manner. That is, the energy dissipated during the post-peak response (failing part, that corresponds to a negative tangent stiffness) is preserved when the mesh size is changed.

The curing cycle shown in Fig. 4(b) is utilized in the present study. The post cure state of the 4 models are studied for a fixed critical stress (σ_0) and fracture toughness (G_C).

Results obtained for the post-cured state corresponding to the 4 different microstructures are shown in Fig. 5. The regions failed due to excessive cure shrinkage are shown in red.

From Fig. 5, it is evident that the material has degraded during the curing process, much prior to being subjected to service loads (for example, tensile loading). It is also observed that the position and area of failed regions change with varying microstructure within the models. Clearly, the amount of degradation and subsequent failure, which are governed by the stress (and strain) evolution in the matrix will change due to the relative positions of the fibers. Thus, the details of fiber packing, keeping all other aspects fixed, will clearly influence the cured state of the composite prior to it being subjected to mechanical loading.
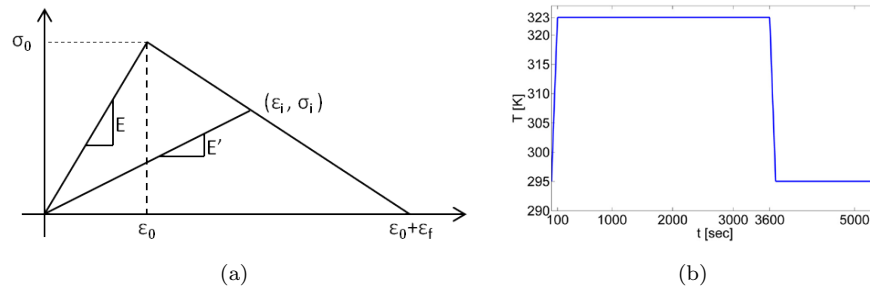
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Figure 4. (a)Modified Stress-strain Material Curve incorporating Failure (b) Curing Cycle
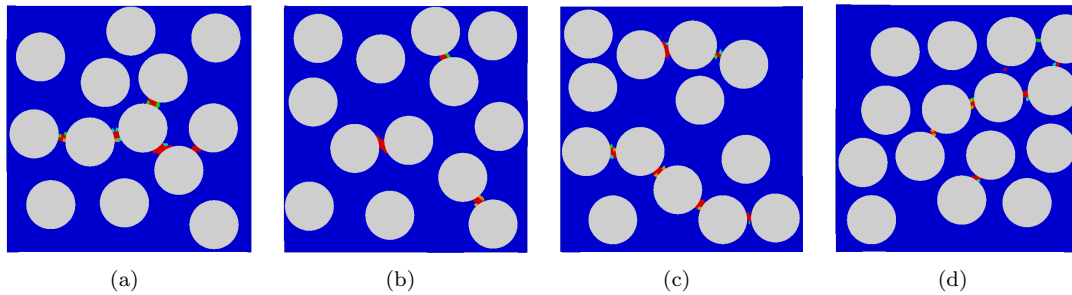

Figure 5. Cured Fiber-Matrix Unit Cells with Red Regions indicating Failed Matrix due to Excessive Cure Shrinkage

VI.   TENSILE FAILURE PREDICTION

Subsequent to curing, the effect of mechanical loading is investigated by the studying the tensile response of the cured microstructures. Subjecting each model to the same maximum displacement, the tensile response of the 4 models are shown in Fig. 6 .
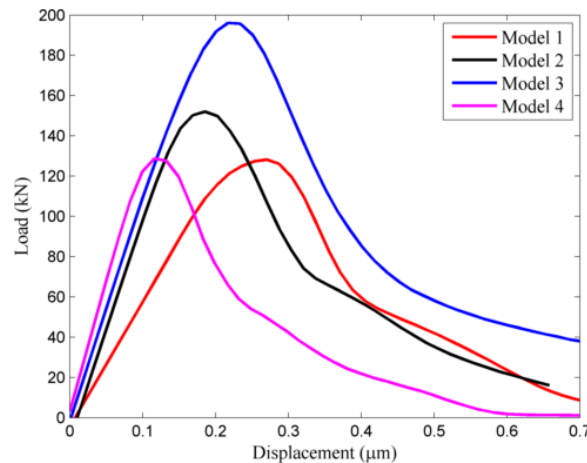

Figure 6. Load-Displacement Responses of the RUCs subjected to Tensile Loading

The tensile response of the 4 models vary significantly for the same applied tensile displacement. Both the initial stifness and the maximum load differ significantly from one microstructure to the other. The change in initial stiffness of the models is directly indicative of the influence of the damage that occurred during the curing process, as different regions of the matrix has reduced stiffness due to degradation.

Also, the damaged regions catalyze failure when the models are further subjected to tensile loading. This is very clear from Fig. 7 and Fig. 8, which display failed regions at the global peak and failed regions of the load-displacement responses shown in Fig. 6. The direction and the regions of failure have originated around the initial damaged regions after curing, and these are shown in Fig. 5. Therefore, it is evident that fiber packing has significant influence on both curing and subsequent tensile response of these composites.

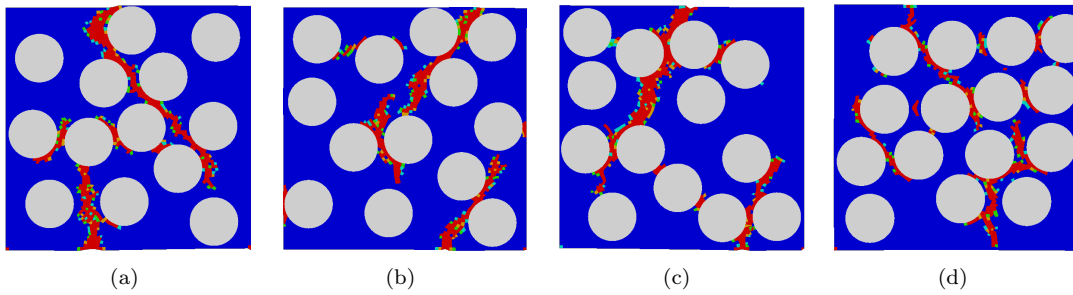American Institute of Aeronautics and Astronautics

Figure 7. RUCs indicating Failed Regions corresponding to the Peak Load of the Tensile Load-Displacement Responses
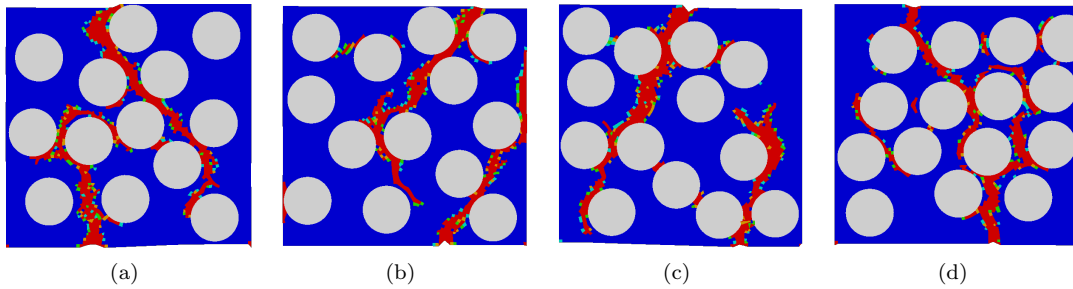

Figure 8. RUCs indicating Failed Regions corresponding to the Complete Failure of the Tensile Load-Displacement Responses

VII.   CONCLUSION AND FUTURE WORK

Preliminary results that show the influence of packing on the deformation, damage development during curing, and subsequent mechanical loading of FRPCs have been presented. The model and the methodology formulated above have demonstrated that fiber packing has a significant influence on the extent and pattern of damage and failure in FRPCs. Significant damage accumulation occurs during the curing process due to cure shrinkage, and pockets of the matrix fail before the culmination of the cure process. Further, the tensile response of the FRPC, represented here by a representative volume element, was seen to be influenced by the damaged regions due to cure, and the initial stiffness as well as the peak loads varied between different microstructures. Therefore, the results from multiple renditions of packing are used to inform the significance (or lack of) of considering cure induced effects on subsequent mechanical response of the composites. This paper clearly suggests that a stochastic modeling approach, considering spatial variability of material properties, need to be used while moving up from the microstructural models to macroscale models, for predicting mechanical loading responses in cured CFRPs. This latter aspect is the subject of ongoing work.

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