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CURE KINETICS AND INTERFACIAL PHENOMENA IN POLYMER MATRIX COMPOSITES

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Polymer matrix composites with textile reinforcement are used in a wide range of aerospace and industrial applications. Continuum mechanical predictions of the composite behaviors have been inaccurate, possibly because of the lack of information with regard to polymer materials properties, especially near the interfaces with the reinforcing fibers. Concurrent micro-Brillouin and Raman light scattering provides sufficiently high spatial resolution to probe the mechanical properties and chemical composition of the interphase regions of the matrix, without interfering with the thermo-mechanical equilibrium of the material. Using this technique, we mapped the elastic properties of epoxy resin in between and within the fiber tows of a composite, revealing that the modulus exhibits a marked spatial inhomogeneity in proximity of fibers, with a decrease of up to 5% compared to that of bulk epoxy resin in the regions of highest fiber density (see Fig. 1).¹ We estimate that it would take a deformation of four times the failure strain to cause such a change in modulus based on residual stresses. Hence, the origin must lie elsewhere.

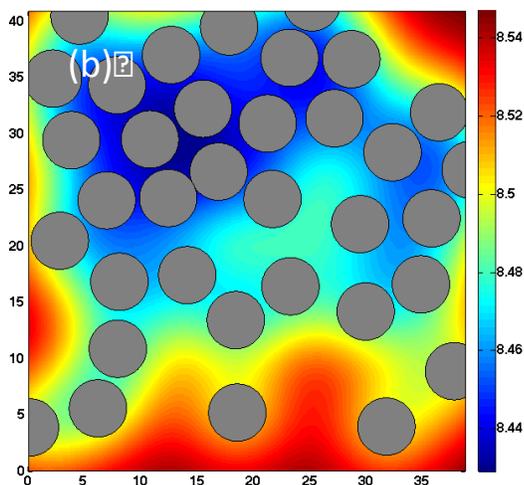


Figure 1 – Longitudinal elastic modulus as a function of the location within a fiber tow

Using the same methodology, we then monitored the elastic properties *in situ*, during epoxy cure under different thermal and chemical conditions. We find that depending on the reaction rate, the elastic modulus evolves differently as a function of the degree of cure: the faster the rate, the more the modulus lags behind of what would be expected from the amount of cross-links that have formed according to the degree of cure. This is because the overall modulus is based on the stiffness resulting from bonded and non-bonded network connections, the latter arising from the optimization of network packing that ensues after a slow structural relaxation.² Provided enough time, the same final modulus is reached, unless network formation is impeded by the under-supply of hardener.

To interpret and enhance these results, experiments are complemented with molecular dynamics simulations of the interface. Accordingly, the one-sided confinement of polymer adjacent to a fiber surface results in clearly detectable structural features, e.g., layering and densification, as well as changes in the elastic properties within a spatial extent that reaches significantly beyond the region of distinguishable structural features.

In conclusion, we attribute the inhomogeneity in mechanical properties to a combination of hardener depletion and an impediment of structural relaxation due to unilateral confinement that lowers the extent and effectiveness of non-bonded interactions.

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