

## Supplementary Information

### ***In situ* Investigation of the Rheological and Dielectric Properties of a Cross-linking Carbon Nanotube-Thermosetting Epoxy**

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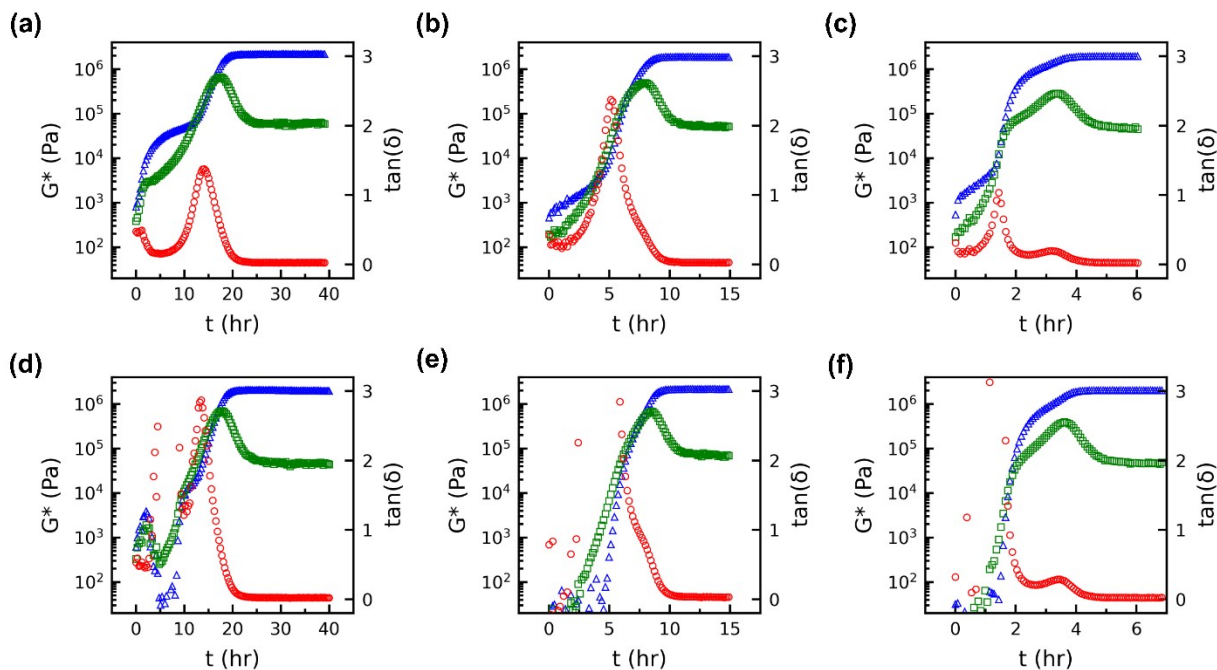
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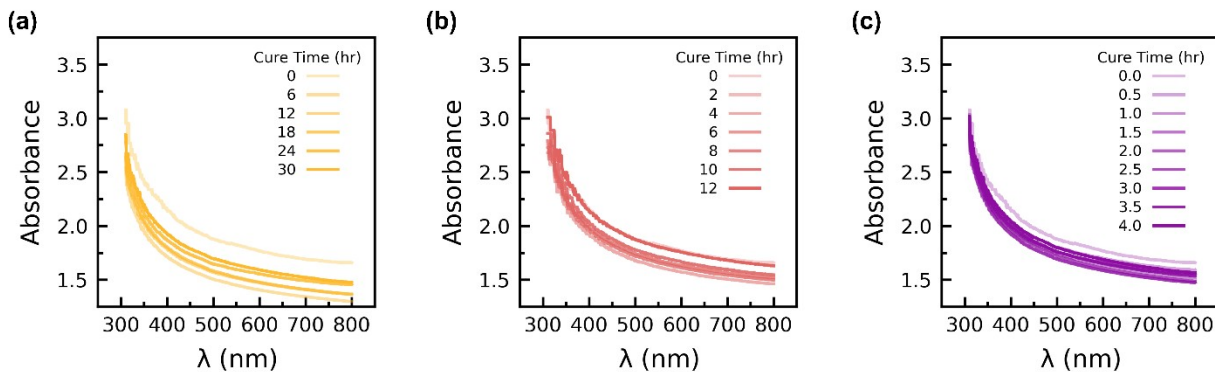
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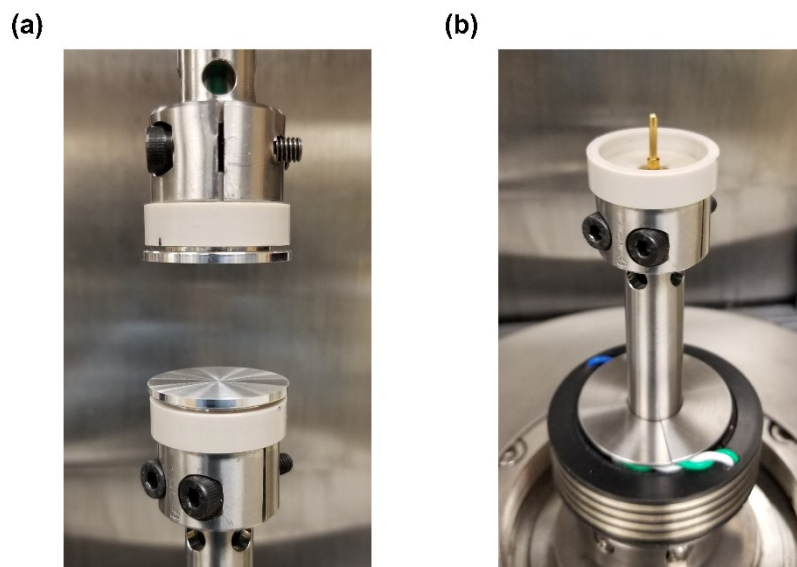
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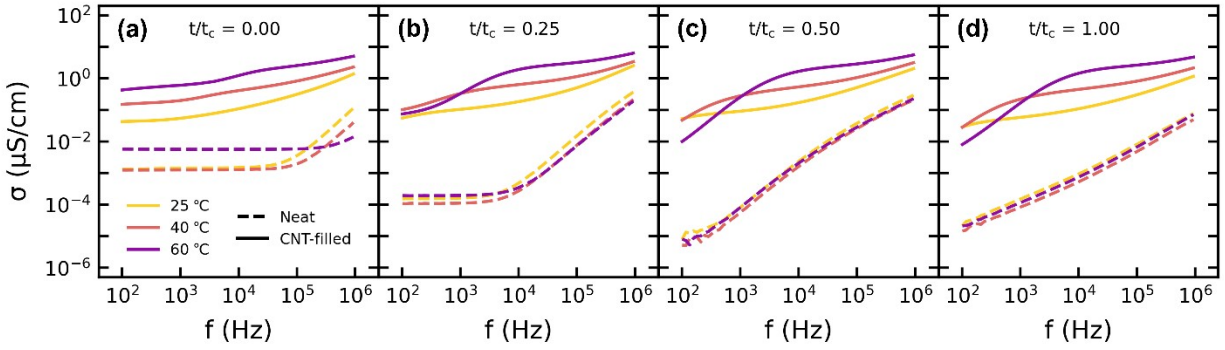
**Figure S1:** Time evolution ( $t$ ) of the storage modulus ( $G'$ ,  $\Delta$ ), loss modulus ( $G''$ ,  $\square$ ), and  $\tan(\delta)$  ( $\circ$ ) of the CNT-filled [(a) 25, (b) 40, and (c) 60 °C] and neat [(d) 25, (e) 40, and (f) 60 °C] thermosetting epoxies. The noise in the data at early times for the neat epoxies is due to insufficient torque contribution to the rheometer as the samples are primarily a liquid. In contrast, CNT-filled epoxies show an enhancement of  $G'$  over  $G''$  from  $t = 0$ . This elastic behavior indicates the formation of a CNT mechanical network in the system.



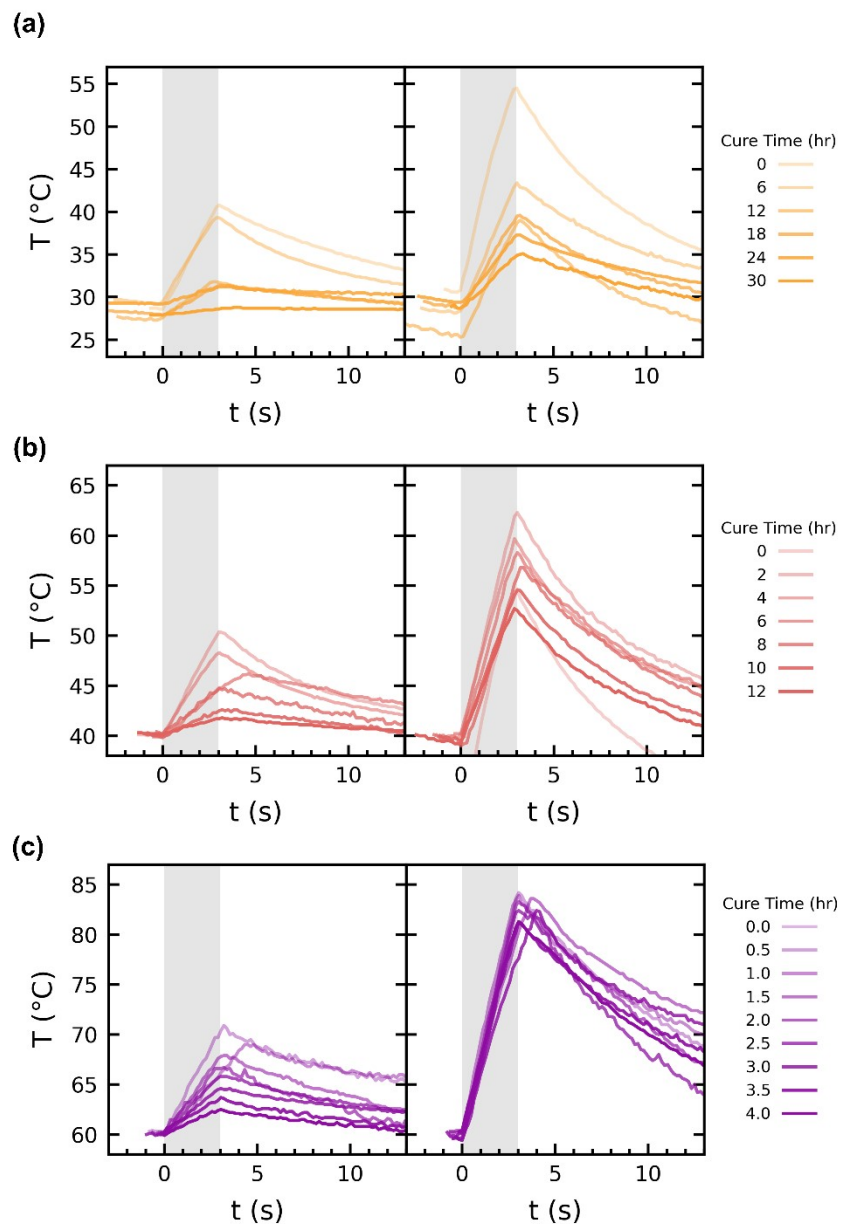
**Figure S2:** UV-vis absorbance as a function of wavelength ( $\lambda$ ) of the CNT-filled epoxies cured at (a) 25, (b) 40, and (c) 60 °C taken with increasing time in hours. Within the first few hours, the absorbance across all wavelengths and samples substantially decreases. This is likely a result of CNT sedimentation when the epoxy system is still a liquid, and the viscosity remains low. The drop in absorbance, however, is not as significant with increasing cure temperature possibly due to the better dispersity of the CNTs. As the samples begin to solidify, the epoxy system slowly becomes less transparent, and the absorbance subsequently rises.



**Figure S3:** Optical images of the dielectric disposable plate geometries mounted on an ARES-G2 rheometer. (a) 25 mm aluminum plates assembled on the top and bottom geometries held at loading gap. (b) Bottom geometry with the plate removed to reveal a pogo pin that connects externally to a Keysight Impedance Analyzer.



**Figure S4:** Frequency sweep ( $f$ ) of the conductivity ( $\sigma$ ) of the neat and CNT-filled epoxies at  $t/t_c$  values of (a) 0.0, (b) 0.25, (c) 0.5, and (d) 1.0. Legend shown in (a) applies to all subplots. Two distinct regions can be observed in the neat epoxies for  $t/t_c < 0.5$ : a direct current (DC) plateau in the conductivity at low frequencies that turns to a sharp increase for higher frequencies. This plateau originates from mobile charge carriers in the system,<sup>1,2</sup> possibly originating from the deprotonation of the alcohol or amine groups from the epoxy and curing agent, respectively. For  $t/t_c \geq 0.5$ , this DC plateau is lost as the cross-linked network is formed, signifying the arrest of the mobile charge carriers in the cured epoxy network. All CNT-filled epoxies also exhibit a DC plateau that is enhanced with increasing cure temperature from electronic contributions. At higher frequencies, the conductivity steadily rises, consistent with dielectric measurements made of CNT-filled liquids and polymers.<sup>3,4</sup> As a function of time, the DC plateau in all CNT epoxies is lost, but the high-frequency conductivity is maintained throughout the cure at all temperatures.



**Figure S5:** Radio-frequency (RF) heating of the neat (left) and CNT-filled (right) thermosetting epoxies cured at (a)  $\sim 25$ , (b) 40, and (c) 60 °C for given times in hours during the curing period. The sample temperature ( $T$ ) was monitored as a function of time ( $t$ ) as the epoxies were exposed to an RF field for 3 sec at  $t = 0$ . The heating rate was calculated by measuring the slope of the temporal plot highlighted by the shaded region. The temperature data at the start of cure for 40 and 60 °C epoxies was omitted as the starting temperature was not at the cure temperature.

## References

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