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Shear-Induced Carbon Nanotube Migration and Morphological Development in Polylactide/Poly(vinylidene fluoride) Blend Nanocomposites and Their Impact on Dielectric Constants and Rheological Properties

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## Abstract:

Polymer composites featuring high dielectric constants are known to be useful materials for the development of modern transducers, piezo- and thermal-sensors, and energy storage devices. In this study, blend nanocomposites of carbon nanotube (CNT)-containing (0.25, 0.75, and 1.0 wt %) polylactide (PLA)/poly(vinylidene fluoride) (PVDF) (70/30 w/w) are developed by melt-mixing PVDF-CNT composite with PLA in a twin-screw extruder. The novelty of this process lies in the fact that the CNTs can migrate from the PVDF to the interface between PVDF and PLA, which could potentially improve the dielectric constant (ɛr) and elastic moduli G'of the composites. For purposes of comparison, the PLA/PVDF/CNT composite is processed using melt extrusion of all three components together. Furthermore, the effects of CNT network formation are understood using high (L) and low (S) CNT aspect ratios. The dielectric and rheological properties of the composite are measured under different shearing and annealing conditions using a dielectro-rheological device. The results show that the dielectric properties are temperature sensitive, exhibiting higher values at higher temperatures, and that L-CNTs form stronger networks than S-CNTs in nanocomposites. Moreover, the CNTs can migrate toward the interface and coarsen the morphologies of the composite when annealing at a temperature of 200 °C. Such phenomena enhance the εr of the PLA/(PVDF + 0.75 wt % L-CNT), from εr ≈ 22 × 103 at t = 0 to  $\varepsilon r \approx 3 \times 104$  after 2 h of annealing at 1 kHz. In contrast, under constant shear rates the  $\epsilon$ r value of the same composite reduces from  $\epsilon$ r  $\approx$  22 x 103 to  $\epsilon$ r  $\approx$  8.2 x 103 after 1 h. However, under the specific measurement conditions of a 0.001 s-1 shear rate applied for 300 s, there is a resulting overshoot in both the viscosity and dielectric constant. Such an observation indicates the suitability of these composites for potential application in highly sensitive strain sensors.