

Application of finite element modeling and viscoelasticity theory in characterization and prediction of dielectric relaxation process in polymer nanodielectrics

Zhao, He, hezhao2012@u.northwestern.edu; Li, Yang; Li, Xiaolin; Brinson, L. Catherine, Northwestern University, United States; Huang, Yanhui; Krentz, Timothy M.; Wu, Ke; Breneman, Curt; Schadler, Linda, RPI, United States

ABSTRACT

Nanodielectrics, typically defined as polymer composites with nanosized ceramic fillers, have demonstrated significant improvements in electrical endurance, breakdown strength and dielectric constant relative to their constituent materials, which leads to enhanced energy storage capabilities. The key role played by the large interfacial area surrounding nanofillers proves to be essential to the enhancement, yet quantitative models to predict the altered dielectric properties in the interfacial area are rarely seen. In this presentation, we apply a finite element modeling approach, originally developed for viscoelasticity analysis, to predict the frequency and temperature dependence of dielectric permittivity spectra in polymer nanodielectrics containing functionalized silica fillers. The dispersion state of nanofillers in the finite element model is determined from descriptor-based analysis of scanning electron micrographs, and the interfacial area surrounding the fillers is explicitly configured into the geometry. The dielectric permittivity spectra of the polymer matrix are imported into the model using a series of Debye relaxation functions. The analogy between dielectric permittivity and viscoelastic modulus allows for a simple mathematical conversion between the two physically distinct quantities, which enables the usage of Prony Series when fitting the dielectric spectrum. With the assistance of a earlier developed algorithm to fit the viscoelastic modulus, the parameters of Debye relaxation series function are obtained. Using the above morphology and physical property inputs, dielectric spectroscopy experiments over a range of frequencies and temperatures can be simulated. Properties of the interfacial region are obtained through an iterative comparison between model output and experimental results. It is observed that the distribution of dielectric relaxation times of the interface could be expressed using those of the polymer matrix multiplied by frequency shift factors that vary with different functionalization of the silica filler surfaces. Our results indicate that surface energy parameters of the filler and the polymer matrix can vary the dielectric response of the composites, which is consistent with earlier observations of the viscoelastic properties of polymer nanocomposites. Further discussion on the results also provides insight into the underlying dielectric relaxation mechanism in the interfacial area.