

**Polymer Nanocomposite Dielectrics:
the Critical Roles of Interface and the Primary Interfacial Polarization**

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Polymer nanocomposites, which combines high dielectric permittivity (ϵ_r) of ceramics and high breakdown strength (E_b) of polymers, are promising dielectrics for high power electrostatic capacitors. Capable of delivering ultrahigh power, they are the major enabler for a number of modern electrical and electronic devices. Tremendous efforts have been made to break the adverse coupling between ϵ_r and E_b and raise their low energy density, make them viable for energy storage applications. The interface between ceramics and polymers plays critical roles in determining the dielectric behavior of polymer nanocomposite dielectrics. We will begin with the exploration of the intricate microstructure of the interface regions and then correlate the microstructures with distribution of charge and chemical species within the interface regions. In the light of these core information, we will discuss the primary interface polarization mechanism in polymer composite dielectrics. We show that the promise of polymer nanocomposite as viable dielectrics for both passive electronic and energy storage devices lies in the fine modulation of interface structure. Along this line, recent progresses are reviewed. By modulating the ceramic/polymer interface or employing novel design paradigm of nanocomposites, concomitant enhancement of ϵ_r and E_b is achieved, giving rise to ultrahigh energy density (enhanced by ~ 1800% over biaxially-oriented polypropylene, the bench-mark polymer dielectric). Besides energy density, thermal stability of polymer nanocomposite dielectrics, especially at high temperature and high voltage, is very recently considered as of critical significance. Further improvement of energy storage performance may lie in the synergistic modulation of electrical, thermal and mechanical properties of polymer nanocomposites.