Investigating Filler Reinforcement and Nonlinear Viscoelastic Behavior in Polymer Composites

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Solid fillers have been known to enhance the linear viscoelastic responses of polymer melts and elastomers. Nonlinear viscoelastic behavior of such systems is closely related to the reinforcement of the linear viscoelascity. Understanding such phenomena as the Payne effect (where the storage modulus is measured to decrease in oscillatory shear with the amplitude of the oscillation and with time for a fixed amplitude) requires a better understanding of the filler reinforcement mechanism. Recent publications[1,2] from two different groups prompted our present study. Using monodisperse 1,4-polybutadiene melts as the matrix and nano-silicon oxide particles of 15 nm diameter as the fillers, we carried out a variety of viscoelastic and NMR-spin-echo diffusion measurements to elucidate the important role of the filler-filler networking in controlling the observed linear and nonlinear behavior at temperatures over 100 degrees above the glass transition temperature of PBD.

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[1] S.S. Sternstein and A. Zhu, *Macromolecules* **35**, 7262 (2002); *Composites Sci. and Techn.* **63**, 1113 (2003). This work claims that the reinforcement arises primarily from the entrapped chain entanglement due to chain adsorption on filler surfaces instead of the filler-filler networking.

[2] H. Montes, F. Lequeux and J. Berriot, *Macromolecules*, **36**, 8107 (2003). This work advocates that a glassy layer formed around each filler is responsible for the enhanced linear viscoelascity and for the observed nonlinear viscoelastic behavior such as the Payne effect.