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New Mechanism Responsible for the Tg-broadening and Nonlinear Response of Nanoparticle-Reinforced Elastomers

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Particle-filled elastomers are employed extensively in modern industry, because they are high-elasticity and relatively low-cost materials that can potentially have their physical characteristics matched to a given design specification. Despite their technological importance, particle-filled elastomers are extremely complicated with pronounced mechanical nonlinearity and temperature dependence and Tg-broadening. In this presentation, we report the fundamentals accounting for this nonlinear effect and Tg-broadening using hairy polymeric nanoparticles of well-defined core-shell structure that are dispersed in polymer matrices of identical chains. By adjusting the filler interaction potential, we are able to show that much of this rheological phenomenon comes from the phase behavior of particles in the polymer matrices and there is a strong connection between the rheological dynamics of particle-filled system and the thermodynamics of phase behavior of particles. The phenomenon of Tg-broadening in one-phase regime appears to differ significantly from that in two-phase regime despite the fact that the polymer around the particles is the same. The strain-induced nonlinearity in particle-filled system is found to display features of singularity near the phase transition point. Although there were attempts in the past to interpret this phenomenon as arising from formation of polymer glassy shells around particles, the present study clearly demonstrates that a glassy shell model is not capable to describe the behavior of particle-filled systems in a unified way. Our finding also suggests new mechanism responsible for the Tg-broadening and nonlinear response of nanoparticle-reinforced elastomers.