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Rate- and Temperature-Dependent Softening in Polymer Glasses

LEON GOVAERT, Eindhoven University of Technology

It is well established that physical aging in polymer glasses leads to an increase in density, elastic modulus, yield stress and also strain softening. The latter, sometimes referred to as "mechanical rejuvenation," is the phenomenon where the post-yield stress level initially decreases with further deformation until strain hardening sets in. In all constitutive models for glasses proposed until now, the rate and temperature-dependence of the yield stress is regarded to remain unchanged during strain softening. In the present study, it is demonstrated that a large number of polymer glasses (PMMA, PLLA, PS, PVC) display a pronounced change in kinetics (strain-rate dependence) after yield. The phenomenon finds its origin in the fact that, in specific ranges of temperature and strain rate, two different molecular mechanisms may contribute to the yield stress. Due to strain softening the post-yield response is only controlled by one of the two, resulting in a strain-rate and temperature dependence of the yield drop. The universality of the phenomenon is discussed in connection to the alleged influence of secondary transitions on the impact response of polymer glasses. A modification to the traditional models is proposed that enables an accurate description of the mechanical response of solid polymers in the transition range.