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Advanced Modeling of Polymer Non-Linear Stress Relaxation – Poly (methylmethacrylate) and Polycarbonate

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Abstract. Sound models of temperature- and strain-dependent non-linear stress relaxation are still lacking. Very recent work has shown that focusing on polymers' local, non-affine, strains and stresses provides an adequate basis for developing such models and accurately predicting experimental stress relaxation moduli, the values of meaningful physical parameters and long time behavior, from experiments spanning only a few hours. A new modeling strategy that explicitly considers such non-affine local stresses and strains was applied to two amorphous polymers – a poly(methylmethacrylate), PMMA, and a bisphenol-A polycarbonate, PC. The results support a view of the stress relaxation process where a temperature-dependent, truncated, approximately log-normal distribution of local cooperative (or clustering) transitions are involved, at and above a minimum (or primitive) relaxor size. Within this view, cooperativity (via the average and maximum cluster sizes) increases with decreasing temperatures. Beyond the reasonable agreement with the experiments, the model succeeds in predicting (1) the effect of increases in the fully relaxed modulus, E_∞, as in semi-crystalline or strongly cross-linked polymers, (2) the strict inapplicability of time-temperature and strain-time super-positions, (3) an extended, Kohlrausch-Williams-Watts, type of relaxation response, spanning 12 or more time decades, and (4) specific, meaningful, physical parameters: a minimum activation energy (close to those of corresponding β-type transitions), the (occupied + free) volume of the primitive relaxor, and the approximate crossover temperature, T_c, and frequency, ν_c, both of critical importance in condensed matter dynamics. The model also has the potential of incorporating the effect of changes in free volume and allows very fast computations, irrespective of the experimental time scale.

Keywords: Stress relaxation - mechanical properties of solids.
PACS: 62.40.+i

SCOPE

Focusing on the material's *non-affine local strains and stresses* provides a sound basis for modeling *stress relaxation* in a physically realistic way and accurately predicting decreasing (from E_0 to E_∞) experimental *stress relaxation moduli* (to within 1% or lower relative errors), the values of meaningful physical parameters and very long time behavior, from experiments spanning less than 3 hours.

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