Abstract Submitted
for the MAR15 Meeting of
The American Physical Society

Stochastic model prediction of the Kovacs' "expansion gap" effect for volume relaxation in glassy polymers GRIGORI MEDVEDEV, JAMES CARUTHERS, Purdue University — The classic series of experiments by A. Kovacs on volume relaxation following temperature jumps for poly(vinyl acetate), PVAc, in the Tg region revealed the richness and complexity of the viscoelastic behavior of glassy materials. Over the years no theoretical model has been able to predict all the features of the Kovacs data, where the so-called "expansion gap" effect proved to be particularly challenging. Specifically, for a series of up-jump experiments with different initial temperatures, Ti, but with the same final temperature, as the relaxation approaches equilibrium it would be expected that the effective relaxation time would be the same regardless of Ti; however, Kovacs observed that the dependence on Ti persisted seemingly all the way to equilibrium. In this communication we will show that a recently developed Stochastic Constitutive Model (SCM) that explicitly acknowledges the nano-scale dynamic heterogeneity of glasses can capture the "expansion gap" as well as the rest of the Kovacs data set for PVAc. It will be shown that the success of the SCM is due to its inherent thermo-rheological complexity.

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Date submitted: 10 Nov 2014 Electronic form version 1.4