Atomistic-to-continuum coupling for structural glasses using the energy-landscape picture

Georgios G. Vogiatzis†‡, M. Semkiv†‡, Lambèrt C.A. van Breemen† and Markus Hütter†*

† Polymer Technology, Department of Mechanical Engineering
Eindhoven University of Technology (TU/e)
P.O. Box 513, 5600 MB Eindhoven, The Netherlands
e-mail: m.huetter@tue.nl, web page: http://www.tue.nl/pt

‡ Dutch Polymer Institute (DPI)
P.O. Box 902, 5600 AX Eindhoven, The Netherlands
Web page: http://www.polymers.nl

ABSTRACT

Physical aging, which originates from slow ongoing changes in the microstructure, is a hallmark of structural glasses, and affects many physical properties, e.g. the yield stress. This presentation addresses the modelling of such materials in terms of the energy landscape.

In the first part, physical aging and mechanical rejuvenation are modeled phenomenologically using two temperatures, characterizing phenomenologically the inter- and intrabasin thermodynamics [1-3], respectively. Doing so in combination with finite-deformation non-isothermal mechanics [4] also leads to the conclusion [5, 6] that the stress of an aging material is in general hypoelastic, rather than hyperelastic.

In the second part, we proceed to unraveling the atomistic origins of physical aging and related features of the glass. The results of a Molecular Dynamics (MD) simulation of atactic polystyrene show that, in the course of time, the system visits several basins in the energy landscape, which are envisioned as discrete states [7, 8] connected to a network by infrequent transitions. It is shown that this network has scale-free and small-world characteristics [9]. In order to overcome time-scale limitations in MD simulations, we have implemented a computational technique that focusses on generating the network of discrete states connected by infrequent transitions over saddle points, while rapid intra-basin properties are captured by the basin free energy. Using this technique, good agreement is obtained with experimental data on infrared-spectroscopy, NMR, dielectric spectroscopy, and mechanical properties. The major benefit of the simulation technique is that it allows to make a direct link between the dynamics on atomistic scale and macroscopic observations.

Acknowledgments: GGV and MS acknowledge financial support by the Dutch Polymer Institute (DPI), project no. 745ft14 and project EU-FP-001 COMPNANO, respectively.

REFERENCES