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From self-assembly to mechanical behaviour: a computational data-driven framework for block copolymers

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Abstract

As inherently heterogeneous materials, block copolymers can demonstrate non-trivial mechanical behaviour such as hyperelasticity, viscoplasticity and hysteresis. Whilst there exist some continuum approaches to modeling the mechanical response of block copolymers [1], models that encompass the full physical detail have not yet been developed. Of interest is the tendency of block copolymer materials to self-assemble into well-defined microstructures/morphologies, and the effect that this self-assembly has on the mechanical response. A practical, physically-based computational approach for block copolymers should thus encompass both the self-assembly process as well as their microscopic features, to fully capture microstructure-mechanical property relations.

In this work, we propose a computational framework that combines self-consistent field theory simulations (SCFT) with molecular dynamics (MD), and a data-driven approach based on the Gaussian process regression. The SCFT approach enables to obtain an equilibrium structure, with additional parameters calculated via Monte Carlo (MC) simulations [2]. The results of the SCFT calculation are subsequently used to generate density biased random walks to build a particle-based molecular model [3-4], capturing a microstructure comprised of glassy and rubbery phases. A coarse-grained MD is then employed to predict the mechanical deformation of those triblock copolymer systems for selected microstructural parameters (block size) and different loading conditions (strain rate-dependent, monotonic, and cyclic). The Gaussian process (GP) regression is then applied to train data based on those selected conditions, to generalize our predictions to our microstructures and loading conditions.

The proposed data-driven computational framework can serve as a bridge between the microscopic physics of a block copolymer and the macroscopic description of the system's large deformation behavior, as well as a powerful method of studying the considerably complex deformation mechanisms that give rise to non-linear phenomena.

References

[1] C. P. Buckley, D. S. A. De Focatiis and C. Prisacariu; In *Constitutive Models for Rubber VII; 1st Ed.; S. Jerrams, N. Murphy,* CRC Press: London, pp. 3-10, 2011
[2] A. Chremos, A. Nikoubashman, A. Z. Panagiotopoulous, J Chem Phys 140, 054909, 2014
[3] A. F. Terzis, D. N. Theodorou, A. Stroeks, Macromolecules 33, 4, pp. 1385-1396 2000
[4] T. Aoyagi, T. Honda, M. Doi, J. Chem. Phys. 117, pp. 8153-8161 2002