Slip-link Simulations of Well-Entangled Linear Polymer Melts under Fast Flows: Role of the Stretch/Orientation-Induced Reduction of Friction

Takeshi Sato and Takashi Taniguchi
Department of Chemical Engineering, Kyoto University, Japan
sato.takeshi@cheme.kyoto-u.ac.jp

Macroscopic flows of entangled polymer melts are tightly related to the microscopic polymer chain dynamics. From a simulation perspective, to tackle this problem, a MultiScale Simulation (MSS) method is highly desirable. In the MSS method for flows of polymer melts, a microscopic coarse-grained model is combined with a macroscopic continuum model[1]. To develop the MSS method that can address polymer processing conditions, we need to construct a microscopic model that can properly predict rheological properties under fast flows.

In this study, we employ the slip-link (SL) model developed by Doi and Takimoto (DT model)[2] to predict rheological properties. To the best of our knowledge, the DT model is one of the most coarse-grained SL models. In the DT model[2], a polymer chain is expressed by a primitive path and slip-links on the primitive path. A slip-link represents an entanglement point between two different polymer chains. The DT model can successfully predict non-linear rheological properties for a moderate strain rate region where polymer chains are oriented without significant stretching. However, the rheological properties obtained by the DT model for fast flows with chain stretch ($\dot{\epsilon} > \tau_{\mathrm{R}}^{-1}$) deviate from experimental data especially of steady-state elongational viscosities.

To improve rheological properties obtained from the DT model for fast flows ($\dot{\epsilon} > \tau_{\mathrm{R}}^{-1}$), we focus on the stretch/orientation-induced reduction of molecular friction (SORF) originally proposed by Ianniruberto *et al*[3]. In this study, we have investigated the effect of SORF on the DT model. As shown in Fig. 1, we have found that SORF can improve the rheological properties obtained from the DT model especially under uniaxial elongational flows.

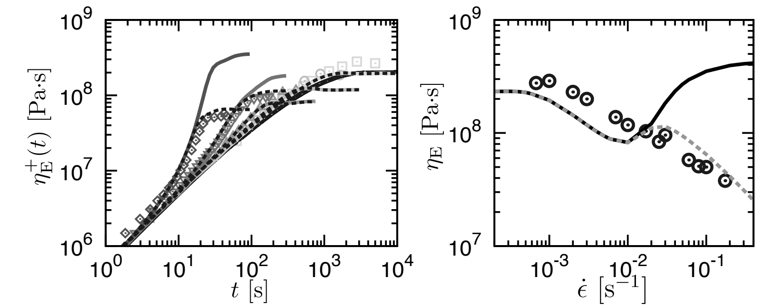

Fig. 1: Results obtained from the DT model for (left) transient uniaxial viscosities and (right) steady-state uniaxial elongational viscosities for a PS200K melt at 130°C. Dashed and solid lines indicate the SL simulation results with and without the stretch/orientation-induced reduction of friction, respectively. Experimental data from Refs. [4] and [5] are shown with symbols.

[1] Sato, T.; Harada, K.; Taniguchi, T. *Macromolecules* **2019**, 52, 547-564.
[2] Doi, M.; Takimoto, J. *Phil. Trans. R. Soc. Lond. A* **2003**, 361, 641-652.
[3] Ianniruberto, G.; Brasiello, A.; Marrucci, G. *Proc. 7th Annu. Eur. Rheol. Conf.* **2011**, 61.
[4] Bach, A.; Almdal, K.; Rasmussen, H.; Hassager, O. *Macromolecules* **2003**, 36, 5174-5179.
[5] Nielsen, J. K.; van Meerveld, J.; Öttinger, H. C. *J. Rheol.* **2006**, 50, 453-476.