

LHMM for Rheological Analysis of Polymer Melts with DPD-SPH Coupling

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This study analyzes the rheological behavior of polymer melts under complex shear flow conditions using a Lagrangian Heterogeneous Multiscale Method (LHMM). The method bridges scales by coupling Dissipative Particle Dynamics (DPD) and Smoothed Particle Hydrodynamics (SPH). At the microscopic level, DPD simulates polymer chains (8 to 32 beads connected by FENE bonds) to extract shear-thinning viscosity, relaxation times, and zero-shear-rate viscosity, which are fitted to the Carreau-Yasuda model. At the macroscopic level, SPH computes continuum flow dynamics, integrating stresses derived from the Irving-Kirkwood (IK) method in DPD.

Validation using benchmark tests (Double Poiseuille Flow and Flow Around a Cylinder) demonstrates the LHMM's ability to accurately predict shear-thinning, stress relaxation, and viscoelastic behaviors under steady and transient conditions. By integrating micro- and macro-scales, the LHMM provides a powerful framework for understanding polymer rheology in complex flows.

I. INTRODUCTION

Polymer melts exhibit complex rheological behaviors due to their inherent viscoelasticity and memory effects, especially under shear and elongational flow conditions. Accurate simulation of these dynamics requires capturing the coupling between microscopic and macroscopic scales, which is highly nonlinear. Traditional Eulerian-based methods struggle to address the history-dependent nature of polymer melts, limiting their applicability to these systems [1], [2]. This study introduces an LHMM that combines DPD and SPH to overcome these limitations [2]. The LHMM integrates microscopic stress contributions with macroscopic flow dynamics, enabling a robust framework for simulating non-Newtonian fluids [3].

The microscopic component uses DPD as a virtual rheometer to characterize key rheological properties of polymer melts, such as shear-thinning viscosity, mean relaxation times, and zero-shear-rate viscosity. Polymer chains are modeled with bead-spring systems connected by FENE bonds. These properties, derived from prior DPD/FENE simulations on viscosity-shear rate dependence [4], [5], are fitted to the Carreau-Yasuda model

to describe the rheological behavior under various conditions. The fitted parameters will be essential for defining the microscopic scaling in the proposed LHMM framework.

At the macroscopic scale, SPH captures continuum flow behaviors, such as velocity gradients and shear rates, while incorporating stress contributions derived from the IK method in DPD [2]. This bidirectional coupling ensures the LHMM can account for complex no-Newtonian responses and memory effects in polymer melts.

A. Lagrangian Heterogeneous Multiscale Methodology

The LHMM bridges microscopic and macroscopic scales to model the rheological behavior of polymer melts under complex flow conditions. It combines DPD simulations for microscale stress computation with SPH to resolve macroscopic flow dynamics. Using the Arbitrary Boundary Condition setup, this approach ensures seamless coupling between scales by accurately capturing stress contributions at the microscopic level while resolving velocity and stress fields and particles and bead chain distribution at the linking of macroscopic and microscopic scales, as illustrated in Fig. 1.

B. Microscale: DPD Rheometer Calibration and Carreau-Yasuda Model

Polymer melts are modeled using bead-spring systems with FENE bonds connecting monomers. The Arbitrary Boundary Condition methodology [6], shown in Fig. 1(a), ensures localized behavior within defined domains, enabling accurate velocity and stress calculations for multiscale coupling.

DPD simulations serve as a virtual rheometer to extract shear-dependent viscosity ($\eta(\dot{\gamma})$), zero-shear viscosity (η_0), infinite-shear viscosity (η_∞), and relaxation times (λ). Figure 1(b) shows η as a function of shear rate ($\dot{\gamma}$) for chains of length $N = 16$, revealing three regimes: random, stretched, and oriented. These results are fitted to the Carreau-Yasuda model (Eq.1), which characterizes the non-Newtonian behavior of polymer melts. The model is defined as:

$$\eta(\dot{\gamma}) = \eta_\infty + (\eta_0 - \eta_\infty) [1 + (\lambda\dot{\gamma})^a]^{(n-1)/a}, \quad (1)$$

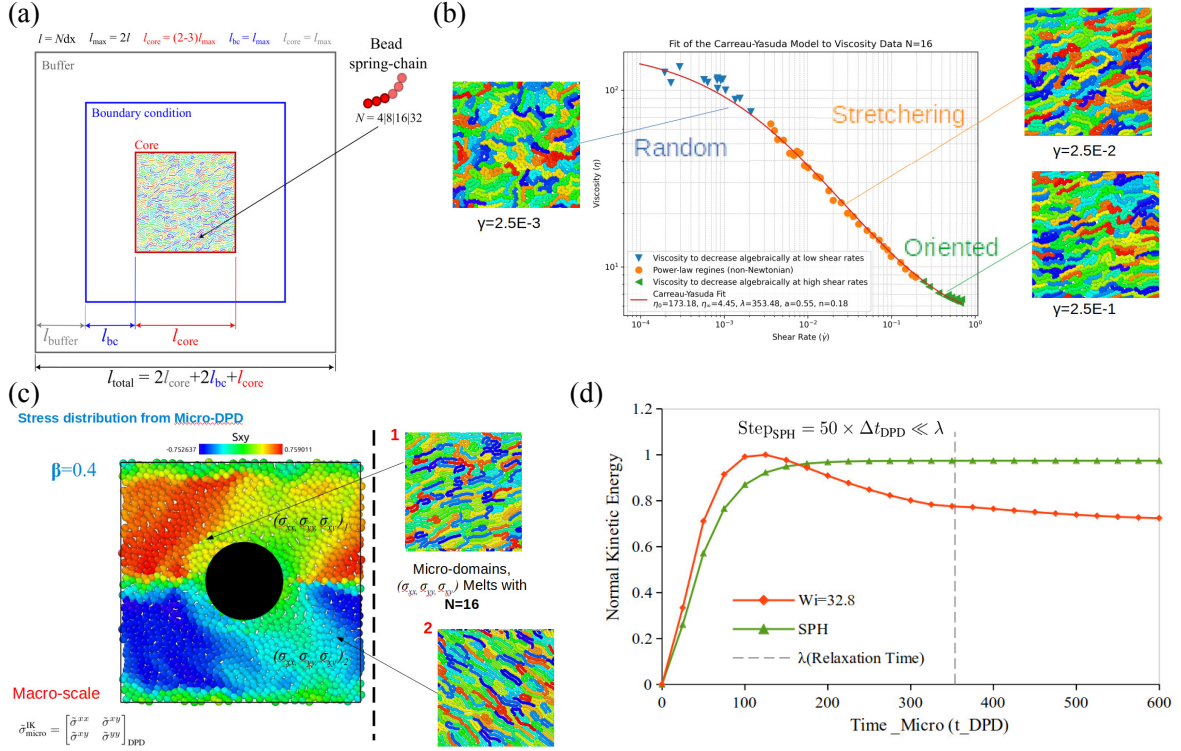


Fig. 1. (a) Schematic representation of the Arbitrary Boundary Condition setup [6], showing the coupling framework between DPD and SPH. (b) Shear viscosity (η) as a function of shear rate ($\dot{\gamma}$) for polymer melts of chain length $N = 16$. Data points correspond to DPD simulations, while solid lines represent the fits of the Carreau-Yasuda model. Different regions indicate random, stretched, and oriented chain distributions. (c) Shear stress distributions from DPD simulations, mapped to SPH particle distributions in the Flow Around a Cylinder test. Velocity distributions in the core domain highlight the effect of micro-macro coupling. (d) Comparison of kinetic energy evolution for SPH simulations with and without microscale coupling, demonstrating the influence of LHMM on transition times and steady-state stabilization.

where a , and n are transition sharpness parameter and power-law index, respectively. Table I summarizes the fitted parameters for different chain lengths (N).

TABLE I
FITTED PARAMETERS FOR THE CARREAU-YASUDA MODEL FOR DIFFERENT POLYMER CHAIN LENGTHS (N).

N	η_0	η_∞	λ	a	n
8	34.29	3.15	89.75	0.95	0.37
16	173.18	4.45	353.48	0.55	0.18
32	242.83	4.81	1307.88	1.23	0.29

The Weissenberg number ($Wi = \lambda\dot{\gamma}$) measures the balance of elastic and viscous forces. It effectively models transient and steady-state dynamics for Wi values from 0.1 to 50, encompassing both low- and high-elasticity regimes [2], [6].

C. Macroscale: SPH and Stress Coupling from DPD

At the macroscopic scale, SPH is used to compute continuum flow dynamics, incorporating stress contributions derived from DPD simulations. The total stress tensor is expressed as:

$$\tau = -p\mathbf{I} + \eta_s \nabla^2 \mathbf{v} + \frac{\beta}{1-\beta} \left(\frac{\eta_s}{\eta_0} \right) \tilde{\sigma}^{\text{IK}}, \quad (2)$$

where p is the pressure, η_s is the solvent viscosity, and $\tilde{\sigma}^{\text{IK}}$ is the microscopic stress tensor calculated using the IK method. The parameter β scales the microscopic stress contribution and is defined as:

$$\beta = \frac{\eta_0^*}{\eta_0^* + \eta_s}, \quad \text{where } \eta_0^* = \epsilon \eta_0. \quad (3)$$

In this context, ϵ serves as a scaling factor that connects macroscopic and microscopic viscosities, specifically linking the zero-shear viscosity of the polymer melt (η_0) to η_s . This coupling ensures that non-Newtonian effects, captured at the microscale (see Fig. 1(c)). The velocity gradient $\nabla \mathbf{v}$ at macroscopic particles is computed as:

$$\nabla \mathbf{v}_i = \sum_j \mathbf{v}_{ij} \nabla W(r_{ij}, h), \quad (4)$$

where \mathbf{v}_{ij} is the relative velocity, W is the SPH kernel function, r_{ij} is the distance between particles, and h is the smoothing length. This gradient reconstructs the velocity \mathbf{v}_I of microscale particles of the different domains of the arbitrary boundaries as $\mathbf{v}_I = \nabla \mathbf{v}_i \cdot \mathbf{r}'_I$, where \mathbf{r}'_I is the relative position of the microscale particle. The method integrates the macroscopic velocity fields of each particle into the microscale domain by

utilizing the arbitrary boundary conditions outlined in [6]. This approach ensures consistency across the boundary conditions, as shown in Fig.1(a). Additionally, as illustrated in Fig.1(c), the method combines microscale stress tensors with SPH velocity gradients to accurately model non-Newtonian flows. At the same time, the DPD stress distributions dynamically adjust to the macroscopic particles, as depicted in Fig.1(c).

II. VALIDATION CASES

The LHMM framework was initially validated using benchmark problems to evaluate its capacity to capture transient and steady-state behaviors under complex flow conditions with $N=16$. Two representative cases were analyzed, as illustrated in Fig. 2.

A. Double Poiseuille Flow

This scenario explores the evolution of velocity profiles (v_x) from initialization through transient stages to achieve steady-state conditions. The results demonstrate the emergence of shear-thinning behavior, which is driven by the microscale dynamics captured through DPD simulations (see Fig. 2(a)). The stabilized velocity profiles are consistent with the predictions made by the Yasuda model, aligning well with the rheology of polymer melts. Additionally, Fig. 1(d) compares kinetic energy evolution between the SPH-only and LHMM approaches. The LHMM effectively captures the transient coupling effects between micro- and macroscales, ensuring stable behavior over the long term.

B. Flow Around a Cylinder

This case analyzes the LHMM framework under complex boundary conditions. During the transient phase, the velocity profiles show deformation, which is later followed by stabilization. This stabilization results in a flattened profile that is characteristic of shear-thinning effects (see Fig. 2(b)). These outcomes reflect the micro-level stress distributions obtained from DPD simulations and align with the predictions of the Yasuda model for the flow of polymer chains.

CONCLUSION

The proposed Lagrangian Heterogeneous Multiscale Method offers a promising framework for modeling polymer melts under complex flow conditions by integrating DPD and SPH. This approach has shown potential in capturing key rheological properties and non-Newtonian behaviors, with applications in understanding polymer melt dynamics.

Using DPD simulations as a virtual rheometer, the study identified rheological parameters such as shear-dependent viscosity, zero-shear-rate viscosity, and relaxation times, which were fitted to the Carreau-Yasuda model. These results form a foundation for incorporating microscopic stress contributions into a multiscale framework.

While benchmark cases like Double Poiseuille Flow and Flow Around a Cylinder have demonstrated the effectiveness of the LHMM, further validation and refinement are necessary to fully assess its robustness and versatility. Future research should explore additional flow configurations and parameter ranges to expand its potential applications.

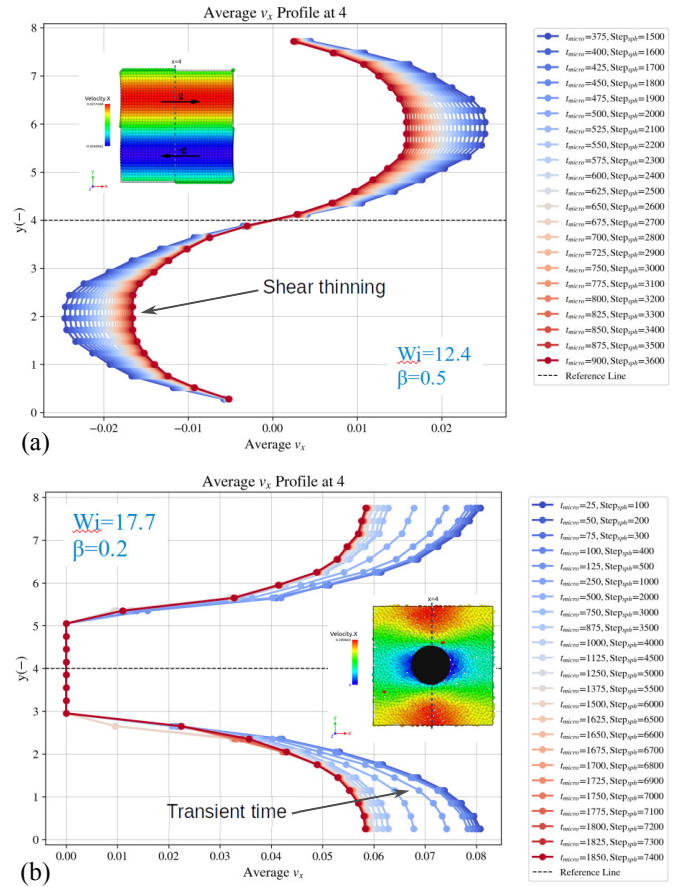


Fig. 2. Validation cases for the LHMM framework. (a) Double Poiseuille Flow: Temporal evolution of velocity profiles (v_x) illustrating shear thinning effects and stabilization under non-Newtonian behavior, consistent with predictions of the Yasuda model. (b) Flow Around a Cylinder: Transient response and steady-state velocity profiles, demonstrating velocity profile deformation and flattening due to shear-thinning behavior, in agreement with micro-level DPD predictions and the Yasuda model.

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