Synchronized molecular-dynamics simulation for the thermal lubrication of a polymeric liquid between parallel plates

Shugo Yasuda [a], *, Ryoichi Yamamoto [b]

[a] *Graduate School of Simulation Studies, University of Hyogo, Japan*
[b] *Department of Chemical Engineering, Kyoto University, Japan*

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ABSTRACT

The Synchronized Molecular-Dynamics simulation, which was recently proposed by authors (Yasuda and Yamamoto, 2014), is applied to the analysis of polymer lubrication between parallel plates. The changes in rheological properties, conformational change of polymer chains, and temperature rise due to the viscous heating are investigated with varying values of thermal conductivity of the polymeric liquid. It is found that with a small applied shear stress on the plate, the temperature of the polymeric liquid only slightly increases in inverse proportion to the thermal conductivity; the apparent viscosity of the polymeric liquid is little affected by changing the thermal conductivity. However, at a large shear stress the transitional behaviors of the polymeric liquid are observed due to the interplay of the shear deformation and viscous heating by changing the thermal conductivity. This transition is characterized by the Nahme–Griffith number Na, which is defined as the ratio of the viscous heating to the thermal conduction at a characteristic temperature. When the Nahme–Griffith number exceeds unity, the temperature of the polymeric liquid increases rapidly and the apparent viscosity also exponentially decreases as the thermal conductivity decreases. The conformation of polymer chains is stretched and aligned by the shear flow when $Na < 1$, but the coherent structure becomes disturbed by the thermal motion of molecules when $Na > 1$.

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1. Introduction

Molecular dynamics (MD) simulations are used to investigate the material properties and internal structures of complex liquids and soft matter. In MD simulations, the dynamics of all of the model molecules that comprise the materials are calculated and the material properties and internal structures of the materials are analyzed from these results [1,2]. The MD simulation is applicable for a wide variety of complex liquids and soft matter consisting of complicated molecules including polymer molecules and biological molecules. However, due to the enormous computational time to calculate the dynamics of all of the molecules, MD simulation is not yet applicable to problems that concern mesoscopic and macroscopic scale motions far beyond the molecular size, where the dynamics of molecules are strongly coupled to the macroscopic transports of complex liquids. At microscopic scales, the macroscopic transports are usually ignored and the MD simulation is performed under a certain ideal environment. To develop a simulation technology to analyze the molecular

dynamics coupled with macroscopic transports for complex liquids is a significant challenge from both a scientific and an engineering point of view. Multiscale modeling is a promising candidate by which to address this challenge.

The multiscale simulation for the flow behaviors of complex fluids was first advanced in the CONNFFESSIT approach for polymeric liquids by Laso and Öttinger [4–6], where the local stress in the fluid solver is calculated using a microscopic simulation instead of using any constitutive relations. The GENERIC approach is also presented for non-isothermal polymeric flows by making important corrections and clarifications to the CONNFFESSIT scheme [7]. The strategy exploited in the CONNFFESSIT approach is also introduced into heterogeneous multiscale modeling (HMM), which was proposed by E and Enquist [8], as a general methodology for the efficient numerical computation of problems with multiscale characteristics. HMM has been applied to various problems, such as the simple polymeric flow [9], the coarsening of a binary polymer blend [10] and the channel flow of a simple Lennard-Jones liquid [11]. The equation-free multiscale computation proposed by Kevrekids et al. is based on a similar idea and has been applied to various problems [12,13]. De et al. proposed the scale-bridging method, which can correctly reproduce the memory effect of a polymeric liquid and demonstrate the non-linear viscoelastic behavior of a polymeric liquid in slab and cylindrical geometries

[14,15]. The multiscale simulation for polymeric flows with the advection of memory in two and three dimensions was developed by Murashima and Taniguchi [16–18]. We have developed a multiscale simulation coupling of MD and transport equations. The multiscale method was first developed for simple fluids [19] and subsequently extended to polymeric liquids with the memory effect [20–23]. Recently, we proposed the synchronized molecular dynamics (SMD) method in which the multiscale method was extended to treat the coupled heat and momentum transfer of complex liquids [24].

The earlier proposed multiscale methods were based on the concept of utilizing the microscopic simulation as a closure of macroscopic transport equations, instead of using any complicated constitutive relations. However, some important characteristics of microscopic dynamics of complex liquids, e.g., conformational changes of polymer chains and heat generation under shear flows, are not correctly treated by the microscopic simulation. The concept of the SMD method is different from those of the earlier proposed methods. In the SMD simulation, MD simulations are utilized to investigate the microscopic dynamics of local fluid elements in macroscopic flows. The MD simulations associated with each fluid element are independently performed to calculate the microscopic dynamics of molecules as well as the various local quantities, but the MD simulations are synchronized to satisfy the macroscopic heat and momentum transfers at a certain time interval. Thus, the SMD method allows for the extension of the conventional MD simulation to the analysis of the microscopic dynamics in macroscopic dynamical systems.

In a previous study [24], the SMD method was applied to polymer lubrication generating the viscous heating between parallel plates. The rheological properties, conformation of polymer chains, and temperature rise due to the viscous heating were investigated for various applied shear stresses on the plate. We found that an interesting transitional behavior of the conformation of polymer chains occurs with a rapid temperature rise for a large applied shear stress due to the coupling of the shear deformation and heat generation under the shear flow.

In this study, we investigate the effects of changing the thermal conductivity of the polymeric liquid on the behaviors of the rheological properties, conformation of polymer chains, and heat generation in the polymer lubrication between parallel plates. In the following, we first describe the problem considered in the present paper, followed by the proposed simulation method. The SMD simulation of polymer lubrication is performed, and the results are discussed; these results are mainly the rheological properties and the coupling of the conformations of the polymer chains with heat generation under shear flows. Finally, a short summary is given.

2. Problem and method

We consider a polymeric liquid contained in a gap of width H between parallel plates with a constant temperature T_0 (see Fig. 1(a)). The polymeric liquid is composed of short Kremer–Grest chains [25] of ten beads, in which all of the bead particles interact with a truncated Lennard-Jones potential defined by

$$U_{LJ}(r) = \begin{cases} 4\epsilon\left[\left(\dfrac{\sigma}{r}\right)^{12} - \left(\dfrac{\sigma}{r}\right)^6\right] + \epsilon, & (r \leqslant 2^{1/6}\sigma), \\ 0, & (r > 2^{1/6}\sigma), \end{cases} \tag{1}$$

and consecutive beads on each chain are connected by an anharmonic spring potential,

$$U_F(r) = -\frac{1}{2}k_c R_0^2 \ln\left[1 - \left(\frac{r}{R_0}\right)^2\right], \tag{2}$$

with $k_c = 30\,\epsilon/\sigma^2$ and $R_0 = 1.5\sigma$. The polymeric liquid is in a quiescent state with a uniform density ρ_0 and a uniform temperature

T_0 before a time $t = 0$. Hereafter, the y-axis is perpendicular to the parallel plates, and the boundaries between the upper and lower plates and the polymeric liquid are located at $y = H$ and 0, respectively. The upper plate starts to move in the x direction with a constant shear stress σ_0 at a time $t = 0$, whereas the lower plate is at rest.

The macroscopic behavior of the polymeric liquid is described by the following transport equations:

$$\rho_0 \frac{\partial v_x}{\partial t} = \frac{\partial \sigma_{xy}}{\partial y}, \tag{3a}$$

$$\rho_0 \frac{\partial e}{\partial t} = \sigma_{xy}\dot{\gamma} - \lambda \frac{\partial^2 T}{\partial y^2}, \tag{3b}$$

where v_α is the velocity, $\sigma_{\alpha\beta}$ is the stress tensor, e is the internal energy per unit mass, and $\dot{\gamma}$ is the shear rate, i.e., $\dot{\gamma} = \partial v_x/\partial y$. Hereafter, the subscripts α,β, and γ represent the index in Cartesian coordinates, i.e., $\{\alpha,\beta,\gamma\} \in \{x,y,z\}$. Here, we assume that the macroscopic quantities are uniform in the x and z directions, $\partial/\partial x = \partial/\partial z = 0$, and the density of the polymeric liquid is constant. Fourier's law for a heat flux with a constant and uniform thermal conductivity λ is also considered in Eq. (3b). Note that the thermal conductivity of polymeric liquids is anisotropic under shear flows in general [26–29], and some experimental studies have reported that the linear stress-thermal relation between the stress tensor and thermal conductivity holds [30–33]. However, in the present study, we consider only the isotropic thermal conductivity as the first step because the effect of shear thinning of the viscosity is thought to be more crucial to viscous heating under strong shear flows than that of the anisotropy of the thermal conductivity. We also assume that the velocity and temperature of the polymeric liquid are the same as those of the plates at the boundaries, i.e., the non-slip and non-temperature-jump boundary conditions.

The effect of viscous heating is estimated using the ratio of the first and second terms in Eq. (3b) to be $\sigma_0\dot{\Gamma}H^2/\lambda\Delta T_0$. Here, $\dot{\Gamma}$ is the gross shear rate of the system, which is defined by the ratio of the velocity of the upper plate v_w to the width of the gap H, $\dot{\Gamma} = v_w/H$, and ΔT_0 is a characteristic temperature rise for the polymeric liquid. In the present problem, we consider a characteristic temperature necessary to substantially change the viscosity of the polymeric liquid, i.e., $\Delta T_0 = |\eta_0/(\partial\eta_0/\partial T_0)|$, where η_0 is the characteristic viscosity of the polymeric liquid at a temperature of T_0. Thus, the Nahme–Griffith number Na, defined as

$$Na = \frac{\sigma_0\dot{\Gamma}H^2}{\lambda|\partial\log(\eta_0)/\partial T_0|^{-1}} \tag{4}$$

represents the effect of viscous heating on the changes in the rheological properties [3,34]. Usually, in lubrication systems and in high-speed processing operations with polymeric liquids, the Nahme–Griffith number is not negligibly small because of the large viscosity and the small thermal conductivity of the polymeric liquid [34]. For example, when a lubrication oil in a gap with a width of 10 μm is subjected to shear deformation with a strain rate of 1×10^4 s^{-1}, the Nahme–Griffith number is estimated to be $Na \gtrsim 0.1$. Thus, the rheological properties of the lubricant in such micro devices must be significantly affected, not only by the large velocity gradient but also by the temperature increase caused by local viscous heating. To predict the rheological properties of the polymeric liquid in these systems, one must consider the temperature variation in Eq. (3b) coupled with Eq. (3a).

Viscous heating is one of the most fundamental and important problems in lubrication systems. However, it is difficult to analyze the behaviors of polymer chains under the coupling of strong shear flows and viscous heating using conventional simulation