Shear Thickening and Turbulence in Simple Fluids

Denis J. Evans and Gary P. Morriss
Research School of Chemistry, Australian National University, Canberra, Australian Capital Territory 2601, Australia
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In 1984 Erpenbeck observed a shear-induced alignment of particles into strings in nonequilibrium molecular-dynamics simulations of shear flow in the hard-sphere fluid. In this paper we show that this effect arises from the use of a thermostat which assumes a stable linear velocity profile. The use of a thermostat which does not bias the streaming velocity profile causes the string phase to vanish.

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To date all thermostatted nonequilibrium molecular-dynamics (NEMD) simulations of shear flow assume that the temperature, T_B, for the shearing system can be defined from the equation

$$dNk_B T_B = \langle \sum m(\mathbf{v}_i - \mathbf{n}_x \gamma y_i)^2 \rangle . \tag{1}$$

In this equation d is the number of dimensions while N is the number of particles. The term $\mathbf{n}_x \gamma y_i$ is the *expected* streaming velocity for particle i. Once the form of the streaming-velocity profile is established it is a simple matter to use peculiar velocity scaling,[1] the Gaussian isokinetic method,[2] or the Nose[3] method to thermostat the shearing system. We should point out that for the Gaussian method, the peculiar kinetic energy is a constant of the motion and therefore the ensemble average employed in (1) is unnecessary. We shall call all thermostats which operate under some *expected* streaming-velocity profile profile-biased thermostats.

At small shear rates and low Reynolds number, the Lees-Edwards shearing periodic boundary conditions[4] do indeed lead to a planar velocity profile of the form assumed in (1). Recently, however, simulations by Erpenbeck,[5] Woodcock,[6] and Heyes, Morriss, and Evans[7] have been performed at very high shear rates where the Reynolds numbers ($R = \rho m \gamma L^2/\eta$) have been very large ($10^3$–$10^5$). The assumption of a linear streaming-velocity profile under these conditions is extremely dubious. Suppose that at high Reynolds number the linear velocity profile assumed in (1) is not stable. In a freely shearing system with Lees-Edwards geometry, this might manifest itself as an S-shaped kink developing in the velocity profile. If (1) is used to define the temperature, the thermostat will interpret the development of this secondary flow as a heating up of the system. The thermostat would extract sufficient heat from the system to prevent this.

For simulations at high Reynolds numbers one needs a thermostat which makes no assumptions whatever about the form of the streaming-velocity profile. The thermostat should not even assume that a stable profile exists. These ideas led us to develop what we call a profile-unbiased thermostat (PUT). We use the Irving-Kirkwood definition of the local peculiar kinetic energy density, $E_K(\mathbf{r},t)$,[8] as a basis for defining a local profile-unbiased measure of the temperature. In a d-dimensional fluid where local thermodynamic equilibrium holds the local temperature is related to the peculiar kinetic energy density and to the local streaming velocity by the equation

$$E_K(\mathbf{r},t) = d[n(\mathbf{r},t) - 1]k_B T(\mathbf{r},t)/2 = \sum \tfrac{1}{2} m[\mathbf{v}_i(t) - \mathbf{u}(\mathbf{r},t)]^2 \delta(\mathbf{r}_i(t) - r). \tag{2}$$

$T(\mathbf{r},t)$ is a measure of the instantaneous temperature at position $\mathbf{r}$ at time t. $n(\mathbf{r},t)$ and $\mathbf{u}(\mathbf{r},t)$ are the usual instantaneous measures of the number density and streaming velocity at $\mathbf{r},t$,

$$n(\mathbf{r},t) = \sum \delta(r_i(t) - \mathbf{r}) \tag{3}$$

and

$$n(\mathbf{r},t)\mathbf{u}(\mathbf{r},t) = \sum \mathbf{v}_i(t)\delta(\mathbf{r}_i(t) - \mathbf{r}). \tag{4}$$

In Eqs. (2)–(4) δ is the Dirac delta function. In an actual simulation it must of course be realized by a finite representation. In our numerical work we use finite square area elements with an area chosen so that on average, they contain two particles. If local thermodynamic equilibrium does not hold then there is little that we can say about a microscopic definition of the thermodynamic temperature. Indeed the status of thermodynamics applied to such systems is still uncertain. It is true, however, that the peculiar kinetic energy per degree of freedom is used universally in computer simulations as a "kinetic" definition of the temperature. Furthermore, its maintenance at a constant average value is a *necessary* condition for the attainment of a steady state.

In principle any of the three schemes mentioned above could be used to thermostat the system. Because of the discontinuous nature of this definition of the temperature [(2)–(4)] we rescaled the peculiar velocities of each particle at every time step in such a way as to keep the average of temperature of the simulation, $T_U(t) = \int d^d r\, T(\mathbf{r},t)/V$, constant.

The system studied was 896 soft disks with

$$\phi(r) = \epsilon(\sigma/r)^{12} \tag{5}$$

at a temperature $k_B T_U/\epsilon = 1$ and a density $\rho\sigma^2 = 0.9238$. The potential (5) was truncated at a distance 1.5σ. This system was chosen because it has been studied extensively before.[6,7,9]

Figure 1 shows the shear viscosity as a function of the logarithm of the shear rate. All units are reduced in units of m, σ, ϵ. The crosses show the earlier results obtained with the linear-profile thermostat. The circles show the new results obtained with the profile-unbiased thermostat. The low-shear-rate "turnover" regime has been described before.[9] We have argued that secondary flows are responsible for the turnover, but the kinetic energies involved are so small compared to thermal kinetic energies ($\sim 5\%$) that we believe that they are little affected by the linear-profile thermostat.

At high shear rates the viscosity falls very sharply with the formation of the string phase. The string-phase behavior is highly nonergodic with a viscosity which depends upon how the system was prepared. Different preparative histories lead to different numbers of strings being formed parallel to the streamlines. Once a certain number of strings have formed that number is essentially preserved by the high-speed grazing collisions between particles in neighboring strings. As has been reported by Woodcock,[6] the initial transients associated with formation of the strings can lead to *transient* shear-thickening behavior.

A typical instantaneous snapshot of an atomic configuration in the string phase is shown in Fig. 2. A reasonably complete picture of this phase is given in Ref. 7. One indication of the lack of realism of the results obtained by use of profile-biased thermostats is given in Fig. 1 of Ref. 7. This figure shows an atomic snapshot at a shear rate which is close to the transition from the homogeneous to the string phase. Figure 1 of Ref. 7 shows the implausible coexistence of string and amorphous phases. This coexistence is implausible because these two phases must be described by different thermophysical properties. In particular, they must have different shear viscosities. It is easily seen that unless the thermostat exerts stabilizing stresses on the system, the coexistence of two phases across a region with a linear velocity profile is impossible. In such a circumstance the condition for stability of two phases is that the stress rather than the strain rate should be constant across the cell. The streaming-velocity profile would, in this circumstance, not be constant across the interface of the two coexisting phases. It is the assumption of a stable linear profile in the formulation of the thermostat used in that work which in fact stabilizes the coexistence observed in Ref. 7.

It is easy to see at a microscopic level how the stand-

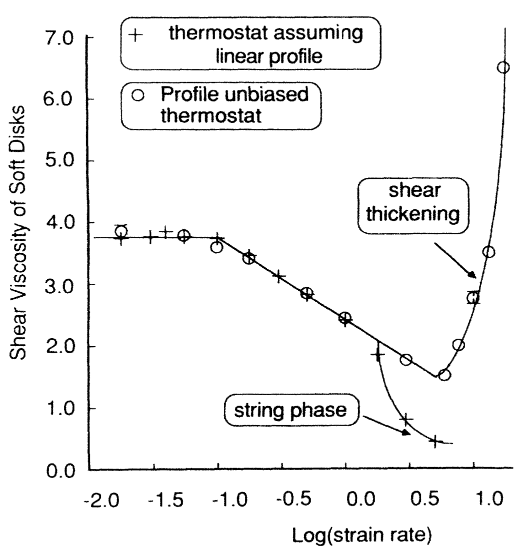


FIG. 1. Viscosity as a function of the base-10 logarithm of the shear rate for a two-dimensional soft-disk fluid. The standard NEMD algorithm leads to the formation of a string phase at high shear rates. This phase's existence is brought about by the assumption of a linear velocity profile in the formulation of the thermostat. This assumption is incorrect at high Reynolds numbers.

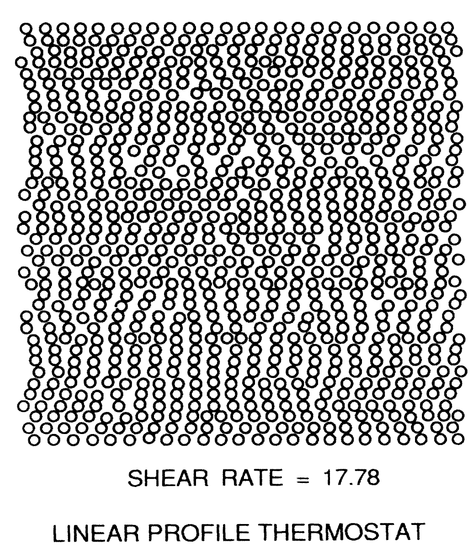


FIG. 2. Instantaneous atomic configuration within the string phase. This phase is stabilized by the linear-profile thermostat and disappears when a profile-unbiased thermostat is used instead. The streaming velocity is parallel to the x axis while the velocity gradient is parallel to the y axis.

ard thermostats can contribute to momentum transport in fluids. The equations of motion for planar Couette flow with either a Gaussian or Nose thermostat can be written in the form[10]

$$d\mathbf{r}_i/dt=\mathbf{p}_i/m, \quad d\mathbf{p}_i/dt=\mathbf{F}_i-\alpha(\mathbf{p}_i/m-\mathbf{n}_x\gamma y_i). \tag{6}$$

The thermostatting multiplier α takes on different forms depending on whether the thermostat is Gaussian isokinetic/isoenergetic or Nose in form. If $\mathbf{J}(\mathbf{r},t)=mn(\mathbf{r},t)\mathbf{u}(\mathbf{r},t)$ is the local momentum density, Eqs. (6) imply[8]

$$d\mathbf{J}(\mathbf{r},t)/dt=-\nabla\cdot(\mathbf{P}+\rho\mathbf{u}\mathbf{u})-\alpha\sum(\mathbf{p}_i/m-\mathbf{n}_x\gamma y)\delta(\mathbf{r}_i-\mathbf{r}). \tag{7}$$

The last term on the right-hand side of (7) influences the momentum transport. For steady-state planar Couette flow at *any* Reynolds number, the profile-biased thermostat forces the velocity profile to be linear with $\sum(\mathbf{p}_i/m-\mathbf{n}_x\gamma y)\delta(\mathbf{r}_i-\mathbf{r})=0$, for all $\mathbf{r}$. This does not happen for our PUT thermostat. Instead of Eqs. (6) we have

$$d\mathbf{r}_i/dt=\mathbf{p}_i/m, \quad d\mathbf{p}_i/dt=\mathbf{F}_i-\alpha[\mathbf{p}_i/m-\mathbf{u}(\mathbf{r},t)]\delta(\mathbf{r}_i-\mathbf{r}). \tag{6$'$}$$

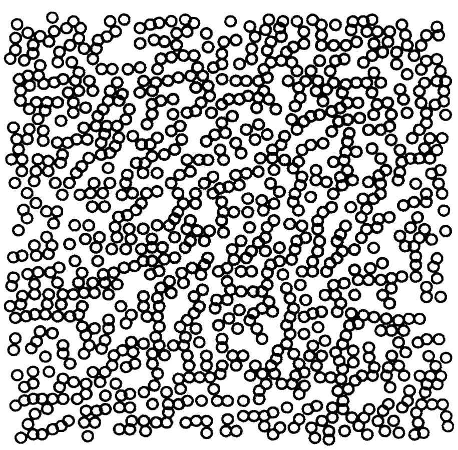
Differentiating the momentum density using (6′) we find that instead of (7) we have

$$d\mathbf{J}(\mathbf{r},t)/dt=-\nabla\cdot(\mathbf{P}+\rho\mathbf{u}\mathbf{u})-\alpha\sum[\mathbf{p}_i/m-\mathbf{u}(\mathbf{r},t)]\delta(\mathbf{r}_i-\mathbf{r})=-\nabla\cdot(\mathbf{P}+\rho\mathbf{u}\mathbf{u}). \tag{7$'$}$$

Provided that the "delta functions," $\delta(\mathbf{r}_i-\mathbf{r})$, are small compared to the characteristic lengths of inhomogeneities in the fluid properties, the profile-unbiased thermostat does not contribute to momentum transport. It may be thought that these results contradict the proof by Evans and Morriss[11] of the identity of Green-Kubo response functions for Newtonian and Gaussian isothermal dynamics. There is, however, no contradiction because the Green-Kubo response pertains only to the limiting small-field regime with a Reynolds number of zero.

The viscosities obtained by use of the profile-unbiased thermostat agree at low shear rates (or Reynolds numbers) with the results obtained by use of the linear-profile thermostat. At high Reynolds numbers the PUT system shows extreme shear thickening. This is not a transient phenomenon. Looking at snapshots of atomic configurations (Fig. 3) reveals that in the shear-thickening regime there is comparatively little long-range positional order. The system is homogeneous and, relative to the biased profile configuration, isotropic.

In Fig. 4 we can see that there is a high degree of lo-



SHEAR RATE = 17.78

PROFILE UNBIASED THERMOSTAT

FIG. 3. Atomic configuration observed at the same nominal shear rate, temperature, and density as Fig. 2. The configuration was generated by use of a thermostat which makes no assumption whatever of the form, or even the stability, of the streaming velocity field. The streaming velocity is parallel to the x axis while the velocity gradient is parallel to the y axis.

SHEAR RATE =17.78

PROFILE UNBIASED THERMOSTAT

FIG. 4. Instantaneous velocity vectors for the configuration shown in Fig. 3. For display purposes the velocities shown are taken relative to the linear profile $\mathbf{n}_x\gamma y$. The velocity vectors show a high degree of local correlation. Convective momentum transport dominates the system.

cal velocity correlation which has no effect on the temperature T_U. Temperature is a measure of the fluctuations of particle velocities about the local streaming-velocity field. The degree of turbulent convection found in PUT systems in the shear-thickening regime can be seen by comparison of the temperature T_U with that based on an assumed linear velocity profile, T_B. Below the transition to the thickening regime these two temperatures are equal. At a strain rate of 17.78, T_B is approximately 24. The unbiased temperature T_U is of course equal to the set value, 1.0. The ratio of these two values provides a measure of the enstrophy of turbulence in the system.

Within the shear-thickening regime, the viscosity shown in Fig. 1 is computed by inclusion of contributions to momentum transport from convection. This value is the same as that computed from the energy balance equation:

$$\langle dE/dt \rangle = \eta_E \gamma^2 V. \tag{8}$$

This equation defines the so-called eddy viscosity η_E. The eddy viscosity is not a true thermophysical property. Its value is dependent upon the Reynolds number as well as upon the thermodynamic variables of temperature, density, and shear rate. The laminar viscosity is defined[8] as the ratio of the appropriate element of the pressure tensor to the strain rate as observed in a free-streaming coordinate system.

The PUT system is ergodic with properties which are independent of the preparative history. Indeed, to check this point we started one run using the profile-unbiased thermostat from a string-phase configuration. After equilibration to the steady state it was indistinguishable from systems which had been prepared solely with use of PUT thermostat.

We have pointed out that for high–Reynolds-number flows, thermostats which assume a specific streaming velocity profile can lead to spurious results. Within the laminar-flow regime, profile-unbiased thermostatting leads to results identical to those obtained by the assumption of a linear profile for planar Couette flow. This indicates that for low–Reynolds-number flows, thermophysical properties are remarkably insensitive to the details of the thermostatting mechanism. Thus low–Reynolds-number simulations that are thermostatted by profile-biased or profile-unbiased methods would seem to provide a well-defined realistic statistical mechanical model of the corresponding experimental systems. On the other hand, future NEMD simulations of high–Reynolds-number flows will have to employ profile-unbiased thermostatting together with local definitions of thermophysical properties in order to yield results which are not dictated by the form of the assumed velocity profile used in the thermostat. Clearly the simulation should predict the correct streaming-velocity profile. It should not be predetermined by assumptions made in the formulation of the thermostat.

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