# Method for determining the shear stress in cylindrical systems

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We develop a method for determining the elements of the pressure tensor at a radius $r$ in a cylindrically symmetric system, analogous to the so-called "method of planes" used in planar systems [B. D. Todd, Denis J. Evans, and Peter J. Daivis, Phys. Rev. E **52**, 1627 (1995)]. We demonstrate its application in determining the radial shear stress dependence during molecular dynamics simulations of the forced flow of methane in cylindrical silica mesopores. Such expressions are useful for the examination of constitutive relations in the context of transport in confined systems.

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## I. INTRODUCTION

The method of planes [1] has been widely used to evaluate elements of the pressure tensor at a given plane during molecular dynamics simulations. Elements of the pressure tensor are obtained from a frequency-space expression for the Navier-Stokes momentum conservation equation, and the derivation relies on the uniform behavior of the system in two of the three Cartesian directions describing the system. Consequently, it is well suited to determining the shear stress $P_{xy}(x)$, as a function of position $x$ across a slit pore, in nonequilibrium molecular dynamics (NEMD) simulations. In combination with a knowledge of the streaming velocity profile $u(x)$ across the pore, this method provides a means of determining the local shear viscosity $\eta(x)$ in accordance with the usual definition for Newtonian fluids, $P_{xy}=-\eta(\partial u/\partial x)$. Combined with other methods, it can provide a useful tool in examining the applicability of this relation to confined fluids.

In the NEMD simulation of forced flow in cylindrical pores, we anticipate an analogous uniformity in the system, but now in the angular and axial degrees of freedom. In such systems, it is the radial dependence of the shear stress that one wishes to determine. In Sec. II, we develop a method analogous to the method of planes, where the mean shear stress at a particular radius is determined. For convenience, we call this the method of cylinders. In Sec. III, we test this method, and compare it with an alternative method based on the direct integration of the momentum conservation equation.

## II. DERIVATION

The starting point for our derivation is the conservation equation for momentum,

$$\frac{\partial \mathbf{J}(\mathbf{r},t)}{\partial t}=-\boldsymbol{\nabla}\cdot[\mathbf{P}(\mathbf{r},t)+\rho(\mathbf{r},t)\mathbf{u}(\mathbf{r},t)\mathbf{u}(\mathbf{r},t)], \tag{1}$$

where $\mathbf{J}$ represents the momentum density, $\mathbf{P}$ represents the pressure tensor, $\rho$ represents the fluid mass density, and $\mathbf{u}$ represents the streaming velocity, each at a position $\mathbf{r}$ in our system at time $t$. The momentum density $\mathbf{J}$ can be written as

$$\mathbf{J}(\mathbf{r},t)=\sum_i m_i\mathbf{v}_i(t)\,\delta(\mathbf{r}-\mathbf{r}_i(t)),$$

where $m_i$ is the mass of the $i$th particle, $\mathbf{v}_i$ its velocity, and $\mathbf{r}_i$ its position.

In Ref. [1], the authors considered transport between planar interfaces, and solved Eq. (1) in frequency space, assuming uniformity in directions parallel to the interfaces over time. Let us instead consider the case where the system is confined by a cylindrical interface, that is, by the surface $r=R$ for the set of cylindrical coordinates $(r,\phi,z)$. In our case, the cylindrical symmetry implies uniformity in the $z$ and $\phi$ directions. In cases of such symmetry, the frequency space is studied using the Hankel transform, rather than the Fourier transform, defined for an arbitrary function $g(r)$ as [2]

$$G(q)=\int_0^\infty g(r)J_0(qr)r\,dr, \tag{2}$$

where $J_0$ is the zeroth-order Bessel function

$$J_0(z)=\frac{1}{2\pi}\int_0^{2\pi}e^{-iz\cos\phi}d\phi.$$

It is also convenient to introduce the first-order Bessel function $J_1$, which obeys the identity [3]

$$J_1(z)=-J_0'(z). \tag{3}$$

The condition of uniformity allows us to take averages over $\phi$, $z$, and $t$. Strictly, one should leave this averaging until the end of the derivation. However, one can greatly simplify the algebra without affecting the result by performing the spatial averages immediately, leaving the time averaging to the end. For each component $J_\alpha$ of the vector $\mathbf{J}$, $\alpha\in\{r,\phi,z\}$, we define

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$$J_\alpha(r,t)=\frac{1}{2\pi Z}\int_Z\int_0^{2\pi}J_\alpha(\mathbf{r},t)d\phi dz,$$

where $Z$ is the range of the $z$ coordinate. We define analogous averages $u_\alpha(r,t)$, $\rho(r,t)$ and $\mathbf{P}_\alpha(r,t)=[P_{\alpha r}(r,t), P_{\alpha\phi}(r,t),P_{\alpha z}(r,t)]$—now all functions of $r$ rather than $\mathbf{r}$—and note that the vector equation, Eq. (1), becomes three scalar equations of the form

$$\frac{\partial J_\alpha(r,t)}{\partial t}=-\boldsymbol{\nabla}\cdot[\mathbf{P}_\alpha(r,t)+\rho(r,t)u_\alpha(r,t)\mathbf{u}(r,t)]. \quad (4)$$

In order to obtain an expression for the pressure tensor, our next step is to determine the Hankel transform of both sides of Eq. (4). In cylindrical coordinates, the momentum density component can be written as

$$J_\alpha(r,t)=\frac{1}{2\pi Z}\int_Z\int_0^{2\pi}\sum_i m_i v_{\alpha i}(t)\times\frac{\delta(r-r_i(t))\delta(\phi-\phi_i(t))\delta(z-z_i(t))}{r}d\phi dz$$
$$=\frac{1}{2\pi Z}\sum_i m_i v_{\alpha i}(t)\frac{\delta(r-r_i(t))}{r},$$

so that its Hankel transform is given by

$$J_\alpha(q,t)=\frac{1}{2\pi Z}\sum_i m_i v_{\alpha i}(t)J_0(qr_i(t)),$$

with time derivative

$$\frac{\partial J_\alpha(q,t)}{\partial t}=\frac{1}{2\pi Z}\sum_i [F_{\alpha i}(t)J_0(qr_i(t))-qm_i v_{\alpha i}(t)v_{ri}(t)J_1(qr_i(t))].$$

This gives us an expression for the Hankel transform of the left side of Eq. (4). In order to calculate the Hankel transform of the right side of Eq. (4), we consider a (pseudo) vector field, $\mathbf{X}(r)=[X_r(r),X_\phi(r),X_z(r)]$. Its divergence is given by

$$\nabla\cdot\mathbf{X}(r)=\frac{1}{r}\left[\frac{\partial}{\partial r}[rX_r(r)]+\frac{\partial}{\partial\phi}X_\phi(r)+\frac{\partial}{\partial z}[rX_z(r)]\right]$$
$$=\frac{1}{r}\frac{\partial}{\partial r}[rX_r(r)].$$

Through integration by parts, it is straightforward to show that the Hankel transformation of this divergence is

$$\int_0^\infty\frac{1}{r}\frac{\partial}{\partial r}[rX_r(r)]J_0(qr)rdr=q\int_0^\infty X_r(r)J_1(qr)rdr.$$

Substituting $\mathbf{X}(r)=\mathbf{P}_\alpha(r,t)+\rho(r,t)u_\alpha(r,t)\mathbf{u}(r,t)$ gives us the Hankel transform of the right side of Eq. (4). Having found expressions for the Hankel transform of both sides of Eq. (4), we equate them to obtain

$$\frac{1}{2\pi Z}\sum_i [F_{\alpha i}(t)J_0(qr_i(t))-qm_i v_{\alpha i}(t)v_{ri}(t)J_1(qr_i(t))]$$
$$=q\int_0^\infty[P_{\alpha r}(r,t)+\rho(r,t)u_\alpha(r,t)u_r(r,t)]J_1(qr)rdr. \quad (5)$$

We recognize on both sides of Eq. (5) contributions from two types of momentum transfer—a configurational contribution corresponding to transfer via forces, and a kinetic contribution corresponding to transfer via convection. We therefore separate and equate the two contributions as follows:

$$\frac{1}{2\pi Z}\sum_i F_{\alpha i}(t)J_0(qr_i(t))=-q\int_0^\infty P^U_{\alpha r}(r,t)J_1(qr)rdr, \quad (6)$$

$$\frac{q}{2\pi Z}\sum_i m_i v_{\alpha i}(t)v_{ri}(t)J_1(qr_i(t))$$
$$=q\int_0^\infty[P^K_{\alpha r}(r,t)+\rho(r,t)u_\alpha(r,t)u_r(r,t)]J_1(qr)rdr, \quad (7)$$

where $\mathbf{P}^U$ and $\mathbf{P}^K$ correspond to the configurational and kinetic parts of the pressure tensor, respectively, such that $\mathbf{P}=\mathbf{P}^U+\mathbf{P}^K$.

We now solve for the elements of the pressure tensor, beginning with $\mathbf{P}^U_r$. Multiplying both sides of Eq. (6) by $J_1(qr')$ and integrating with respect to $q$ leads to

$$\frac{1}{2\pi Z}\sum_i F_{\alpha i}(t)\int_0^\infty J_0(qr_i(t))J_1(qr')dq$$
$$=-\int_0^\infty P^U_{\alpha r}(r,t)\left[\int_0^\infty qrJ_1(qr)J_1(qr')dq\right]dr.$$

Using the identities [3,4]

$$\int_0^\infty qr'J_1(qr')J_1(qr)dq=\int_0^\infty qrJ_0(qr)J_0(qr')=\delta(r'-r),$$

$$\int_0^\infty J_1(qr')J_0(qr)dq=\frac{1}{2r'}[\mathrm{sgn}(r'-r)+1],$$

where $\mathrm{sgn}(x)$ is the usual signum function, and noting that since the $F_{\alpha i}(t)$ represent internal forces only, they sum to zero, we obtain

$$P^U_{\alpha r}(r,t)=\frac{-1}{4\pi rZ}\sum_i F_{\alpha i}(t)\mathrm{sgn}(r-r_i(t))$$
$$=\frac{1}{2A(r)}\sum_i F_{\alpha i}(t)\mathrm{sgn}(r_i(t)-r),$$

where $A(r)=2\pi rZ$ represents the surface area of the cylinder of radius $r$. Finally, we average over time, assuming a steady-state flow, to obtain

$$P^U_{\alpha r}(r)=\frac{1}{2A(r)}\overline{\sum_i F_{\alpha i}(t)\mathrm{sgn}(r-r_i(t))}, \tag{8}$$

where $\overline{a(t)}$ represents the time average of $a(t)$. In analogy with the result in Ref. [1], only those forces acting through the surface of the cylinder of radius $r$ contribute to $P^U_{\alpha r}(r)$.

Let us now consider the kinetic contribution to the pressure tensor, $\mathbf{P}^K_r$. As in the configurational case, we multiply both sides of Eq. (7) by $J_1(qr')$ and integrate with respect to $q$ to obtain

$$P^K_{\alpha r}(r,t)+\rho(r,t)u_\alpha(r,t)u_r(r,t)$$
$$=\frac{1}{2\pi rZ}\sum_i m_i v_{\alpha i}(t)v_{ri}(t)\,\delta(r_i(t)-r).$$

Finally, we average over time, and rearrange to obtain the expression

$$P^K_{\alpha r}(r)=\frac{1}{A(r)}\overline{\sum_i m_i v'_{\alpha i}(t)v'_{ri}(t)\,\delta(r_i(t)-r)}, \tag{9}$$

where $\mathbf{v}'_i$ is the peculiar velocity of particle $i$, $\mathbf{v}'_i=\mathbf{v}_i-\delta(r-r_i(t))\mathbf{u}(r,t)$. Note that the expression being averaged on the right side of Eq. (9) is not instantaneously equal to $P^K_{\alpha r}(r,t)$, but yields the same average over microscopically large time scales. In analogy with the result in Ref. [1], only particles passing through the surface of the cylinder of radius $r$ contribute to $P^K_{\alpha r}(r)$.

## III. COMPUTER SIMULATION

In order to test these expressions for the pressure tensor, we consider the case of forced flow along a cylindrical mesopore. The simulations model the flow of Lennard-Jones (LJ) methane in a silica pore of radius 1.919 nm (approximately 5 $\sigma_{CH_4}$). The wall is also modeled using LJ sites with fitted parameters, and the Lorentz-Berthelot rules are used to determine solid-fluid LJ interaction parameters. In addition, particles closer to the wall than the solid-fluid interaction potential minimum are diffusely scattered in the plane tangential to the pore wall, at the point where the radial velocity of the methane molecule is zero. This process randomly redistributes the particles' momentum in this plane, thus affecting the axial momentum, but not the radial momentum. We refer the reader to Ref [5] for further simulation details.

Simulations were performed over average fluid number densities ranging from $n=1.9$ nm$^{-3}$ to $n=11.1$ nm$^{-3}$. For the lower densities, an external force field of $F_z=2.66\times10^{-14}$ N per molecule was applied in the $z$ direction, whereas at higher densities a field of $F_z=5.32\times10^{-15}$ N per molecule was applied. All simulations were performed at 150 K. In each simulation the shear stress $P_{zr}(r)=P^U_{zr}(r)+P^K_{zr}(r)$ was calculated from the method of cylinders expressions Eqs. (8) and (9), using 500 cylinders with equally spaced radii across the pore. The calculation of $P^U_{zr}(r)$ is relatively straightforward, as a pair interaction only contributes where $r$ lies between the radial coordinates of the interacting particles. The calculation of $P^K_{zr}(r)$ is less straightforward. Suppose a particle (labeled $j$) crosses the cylinder at $r$ at time $t_X$ in the time interval $[t,t+\Delta t]$. The contribution to $P^K_{zr}(r)$ from this event would be

$$\frac{1}{A(r)\Delta t}\int_t^{t+\Delta t} m_j v'_{zj}(t)v'_{rj}(t)\,\delta(r_j(t)-r)\,dt$$
$$=\frac{1}{A(r)\Delta t}\int_t^{t+\Delta t} m_j v'_{zj}(t)\,\delta(t_X-t)\,dt=\frac{m_j v'_{zj}(t_X)}{A(r)\Delta t}.$$

Thus contributions to $P^K_{zr}(r)$ consist of the axial peculiar momentum of particles as they cross the cylinder at $r$.

In order to verify the results of this method, a second means of determining the shear stress was employed. The momentum conservation equation, Eq. (1), for forced flow leads to the following expression:

$$\frac{1}{r}\frac{\partial(rP_{zr}(r))}{\partial r}=n(r)F_z,$$

for the steady state, where $n(r)$ represents the number density at radius $r$. Thus we have an alternative expression for the shear stress,

$$P_{zr}(r)=\frac{F_z}{r}\int_0^r r'n(r')dr'. \tag{10}$$

In Fig. 1, we compare the values for the shear stress obtained using the method of cylinders (labeled MOC) with the values obtained by integrating the momentum conservation equation (labeled IMC), for the system with average number density $n=1.9$ nm$^{-3}$. We note that the two results obtained using these two methods agree to within the predicted error, which is smaller than the size of the symbols used in the figure, until a critical radius $R_0$ is reached. $R_0$ corresponds to the minimum in the solid-fluid interaction potential. This discrepancy is due to the diffuse boundary conditions that only occur for particles at $r>R_0$. The values of $P_{zr}(r)$ obtained from the IMC method report the shear stress required to maintain the particles contained within this volume at a

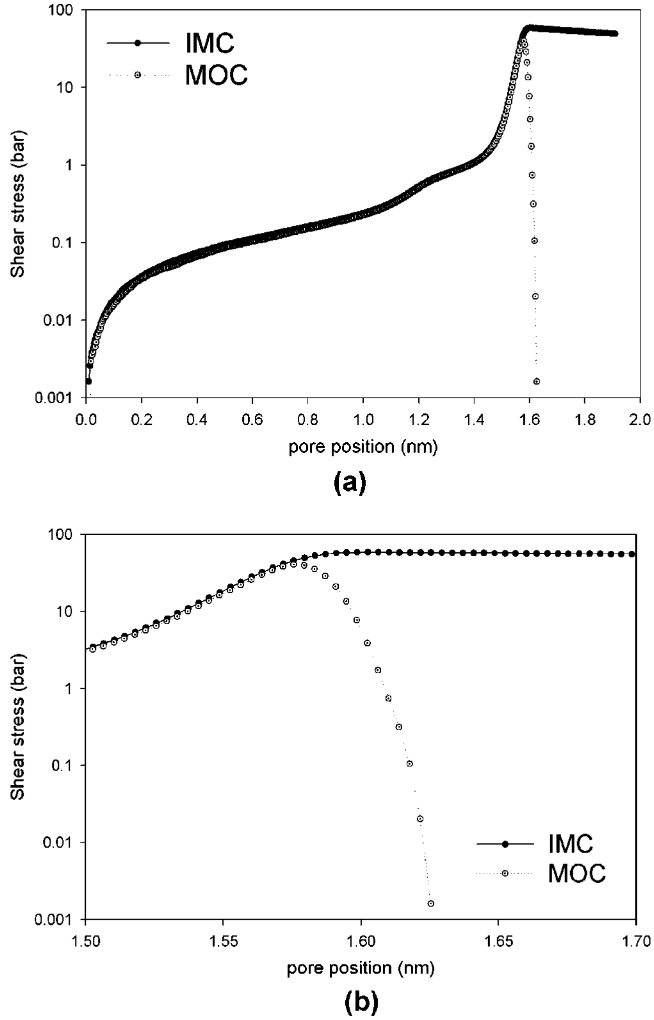


FIG. 1. Shear stress calculated using the method of cylinders (MOC)—Eqs. (8) and (9)—and integration of the momentum conservation equation (IMC)—Eq. (10)—for the forced flow of methane in a silica mesopore, $n=1.9\ \mathrm{nm}^{-3}$.

steady state, that is, the shear stress required to balance the external force field $F_z$. Thus, IMC values give the *total* shear stress experienced by particles at a given radius. However, the values of $P_{zr}(r)$ obtained from the MOC reflect fluid-fluid momentum exchange only, and hence give the *fluid* contribution to the shear stress experienced by particles at a given radius. Consequently, the difference between the two values represents the shear stress exerted by the pore wall at a given radius. This momentum exchange between solid and fluid, arising from the diffuse boundary conditions, represents velocity slip at the wall. Such slip can be represented by the introduction of a frictional boundary condition to a hydrodynamic model of the form $k\rho(r_0)u(r_0) = P_{zr}(r_0)$ where $r=r_0$ represents the boundary of the model [5–7]. In Ref. [5], $r_0$ is the location of the solid-fluid potential minimum.

We observe similar behavior at all tested densities. Figure 2 shows the results obtained for a system with mean number density of $n=11.0\ \mathrm{nm}^{-3}$—the densest system examined in this work.

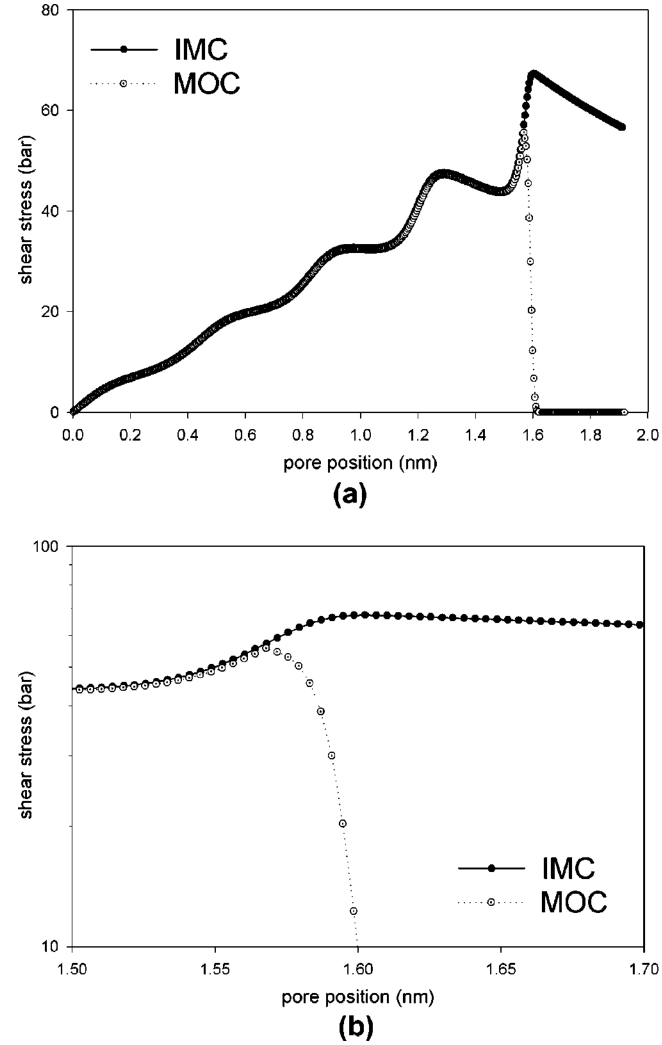


FIG. 2. Shear stress calculated using the method of cylinders (MOC)—Eqs. (8) and (9)—and integration of the momentum conservation equation (IMC)—Eq. (10)—for the forced flow of methane in a silica mesopore, $n=11.0\ \mathrm{nm}^{-3}$.

## IV. CONCLUSION

We have derived an expression for determining the radial dependence of elements of the pressure tensor in systems with cylindrical symmetry. This method of cylinders is analogous to a previously derived approach used in systems with planar symmetry. We have successfully tested this expression in examining the radial dependence of the shear stress, through a comparison with an alternative expression based on integrating the momentum conservation equation (IMC method). However, where the method of cylinders measures only that part of the shear stress exerted by other particles, the IMC method measures contributions from external sources as well. We anticipate that the method of cylinders will be useful in the study and development of localized expressions for fluid shear viscosity, for confined fluids.

The physical interpretation of the resultant expressions for the method of planes and the method of cylinders are similar, and suggestive of a general principle for other geometries in which the frequency-space analysis is more difficult to develop.

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