

Series development of the stress tensor for the shear viscosity of gases in the transition regime

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Abstract

We discuss some relevant features of the variation of the effective viscosity of gases with the pressure in the transition regime. Both qualitative arguments and formal ones are given in order to explain the regular pattern of oscillation of the effective viscosity with pressure that has been experimentally observed in gases under the transition regime. Using the Boltzmann equation under the relaxation time approximation, an infinite series expression for the tangential stress P_{zx} in terms of the spatial derivatives of the gas velocity is deduced. For constant boundary conditions an appropriate cut off of this series up to fifth order in the mean time between collisions produces an ordinary differential equation that can be easily solved for P_{zx} assuming laminar flow and a small dependence of the fluid velocity with the spatial distance "z", from the walls of the container.

Key words: Gases; transition regime; viscosity.

Desarrollo en series del tensor de esfuerzos de la viscosidad de gases en régimen de transición

Resumen

Se discuten algunas características importantes de la variación de la viscosidad efectiva de los gases con la presión, en el régimen de transición. Se presentan argumentos tanto cualitativos como formales con el objeto de explicar el patrón regular de oscilación de la viscosidad efectiva con la presión, el cual ha sido experimentalmente observado en gases bajo Régimen de Transición. Usando la ecuación de Boltzmann en la aproximación del tiempo de relajación, se dedujo una expresión en la serie infinita para el esfuerzo tangencial P_{zx} en términos de las derivadas espaciales de la velocidad de un gas. Para condiciones de contorno constantes, un corte apropiado de esta serie hasta orden quinto en el tiempo promedio de colisiones, da lugar a una ecuación diferencial ordinaria que puede ser fácilmente resuelta para P_{zx} suponiendo flujo laminar y una pequeña dependencia de la velocidad del fluido con la distancia espacial "z" desde las paredes del recipiente.

Palabras clave: Gases; régimen de transición; viscosidad.

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Introduction

The first experimental evidence of the oscillating dependence of the shear viscosity of gases with pressure in the transition regime was reported in 1978 (1). Since then, significant advances both in the theoretical understanding of the phenomenon (2-7) and its experimental measurement (1,4,8-9) had been made. According to the theory it has been realized that the first order gradient expansions of the stress do not explain flows in the transition regime (10-13) and therefore higher order equations in the Chapman-Enskog expansion appear to be necessary (10-14). In order to illustrate this point let us consider the Couette flow of a gas between infinite parallel plates moving relative to each other and separated by a distance L . This problem, although simpler than the flow between rotating coaxial cylinders, already presents the referred effect. The mean force on a plane parallel to the plates and passing through the point z lying between $z=0$ and $z=L$ is due to the momentum transfer per unit area and unit time, and is given by the usual approximate expression (15):

$$P_{zx} = \frac{1}{6} nm\bar{v} v_z(z, \ell_c) v_x(z, \ell_c), \quad [1]$$

where z is an axis perpendicular to the infinite parallel plates, x the axis along which the relative motion of the plates occurs, ℓ_c the mean free path, and m and \bar{v} the mass and mean speed of a molecule respectively. Since we are considering the steady state of a laminar flow, we must have unaccelerated layers, which in turn requires that P_{zx} be independent of z . Therefore, the difference $v_x(z, \ell_c) - v_x(z, \ell_c)$ appearing as a factor in Eq. [1] must be independent of z . This occurs when $v_x(z)$ is linear in z , or, more generally, when $v_x(z)$ is a linear combination of a periodic function of z with period $2\ell_c$, and a linear term in z . The simplest function $v_x(z)$ fulfilling this condition is given by (3):

$$v_x(z) = A(\ell_c) \sin \frac{\pi z}{\ell_c} + B(\ell_c) \cos \frac{\pi z}{\ell_c} + C(\ell_c)z + D(\ell_c) \quad [2]$$

where the coefficients A , B , C and D may depend on ℓ_c (or equivalently on the density $n = \frac{1}{\sqrt{2}\sigma\ell_c}$).

When $v_x(z)$ is linear in z , i.e. $A(\ell_c) = B(\ell_c) = 0$, we have from Eqs. [1] and [2] that $P_{zx} = \frac{1}{3} nm\bar{v}\ell_c C(\ell_c)$. Then, we obtain in this case the usual result for the continuous regime: $P_{zx} = \eta \Delta v / L$ with $\eta = \frac{1}{3} nm\bar{v}\ell_c$ and $v_x(z) = v_0 + (\Delta v / L)z$. Note that v_0 and v_L may depend on ℓ_c through the slip at the boundary layers and $\Delta v = v_L - v_0$ (13).

The need to go beyond a linear approximation for $v_x(z)$, appears when one realizes that, depending on ℓ_c , and on the chemical and physical properties of the solid surfaces and the gas, the quantities $a_0 = (v_x / z)_{z=0}$ and $a_L = (v_x / z)_{z=L}$ may take values which are not necessarily equal to $\Delta v / L$. In other words, to describe cases where $a_0 \neq \Delta v / L$ and/or $a_L \neq \Delta v / L$, we must use for $v_x(z)$ a more general (flexible) function of z than the linear one, still fulfilling that the P_{zx} given in Eq. [1] is independent of z . This leads us to take into account all the four terms in Eq. [2], which together with the four boundary conditions $v_x(0) = v_0, v_x(L) = v_L, (v_x / z)_{z=0} = a_0$ and $(v_x / z)_{z=L} = a_L$, yields (3):

$$P_{zx} = \eta \frac{1 - \frac{\ell_c(a_L - a_0)}{\Delta v} \tan \frac{\pi L}{2\ell_c}}{1 + \frac{2\ell_c}{\pi L} \tan \frac{\pi L}{2\ell_c}} \frac{\Delta v}{L} \quad [3]$$

The P_{zx} given in Eq. [3] oscillates with density n in all situations where $(a_L - a_0) \neq 2\Delta v / L$. On the other hand, if the boundary conditions are equal to those corresponding to the linear expression $v_x(z) = C(\ell_c)z + D(\ell_c)$, i.e. $a_0 = a_L = \Delta v / L$, then Eq. [3] reduces to the usual expression $P_{zx} = \eta \Delta v / L$, as expected. Furthermore, in

the continuous regime limit $\ell_c \rightarrow 0$ the expression given in Eq. [3] also reduces to $P_{zx} = \eta \Delta v / L$, even when $(a_2 - a_0) \neq 2\Delta v / L$, which is also the expected result.

A similar expression for P_{zx} , as is given in Eq. [3], can be obtained from a less crude procedure consisting of a series development of the stress tensor of the shear viscosity of gases which can be deduced from the Boltzmann equation using the relaxation time approximation (2). As shown by the experiments (1, 4, 8-9) this simple formalism is able to justify (2): 1) the oscillating variation of the shear viscosity of gases, with respect to pressure in the transition regime; 2) the position of the principal signals of that non monotonic variation of the viscosity as a function of pressure; 3) the movement of those signals with an external thermal gradient at constant pressure; 4) the position in pressure of the signals as a function of the distance between the sliding parallel plates; and 5) appears to reproduce some interesting features of the shear viscosity of thin liquids films sheared between parallel plates at molecular separations (16-17).

According to the referred formalism a third order expression for P_{zx} has the following form (2):

$$P_{zx} = \eta \frac{u_x}{z} - \gamma \frac{3u_x^3}{z^3},$$

[4]

where $\eta = (\pi/8)nm\bar{v}\ell_c$ and $\gamma = (3\pi^2/64)nm\bar{v}\ell_c^3$, and u_x is velocity of the gas in the x direction (parallel to the shearing plates). Assuming a steady state condition, ($P_{zx} = \text{constant}$), the solution of the third order differential equation (Eq. [4]), leads to a very similar expression to that previously deduced above from intuitive arguments (Eq. [3]):

$$P_{zx} = \eta \frac{\Delta u}{L} \frac{1}{1 - 2 \tan(\omega L / 2) / \omega L} \frac{(a_1 - a_0) \tan(\omega L / 2) / \Delta u \omega}{\omega L},$$

5]

where $\omega = (\eta/\gamma)^{1/2}$ and $\Delta u = \Delta v$.

If the approximations leading to Eq. [5] are carefully reviewed, it can be observed that: 1) the Boltzmann approach in the time-relaxation approximation should be a good approximation in the range of pressure and temperature used in the experiments reported; 2) the supposed re-establishment of a local maxwellian distribution after the collisions implied in the time relaxation approximation cannot be completely guaranteed, though the existence of a total maxwellian distribution at very low pressures has been experimentally confirmed (18); 3) the experimental apparatus employed in the viscosimetric measurements presents a cylindrical geometry, similar in some respects but not equivalent to the parallel plate case previously considered; 4) the constancy of the boundary conditions with respect to pressure is a rough approximation since the mechanism by which the gas molecules exchange energy and momentum with the walls of the container is complex and its effect upon the velocity distribution function difficult to estimate; 5) although Eq. [4] leads to the well known expression for P_{zx} in the continuous regime

$$P_{zx} = \eta \frac{u}{z} - \eta \frac{\Delta u}{L},$$

the zero pressure limit of this expression is not correct, since the coefficient of viscosity does not depend on pressure due to their linear dependence on the density and on the mean free path ($\ell_c = \frac{1}{\sqrt{2}\sigma n}$; where σ is the molecular cross section and n the density of the gas under consideration).

In what respect to point 5) above, that deficiency can be easily corrected with the employment of a suitable expression for the mean free path of the gas molecules which takes into account that there is a physical limit to the zero pressure mean free path (ℓ_c), given by the size of the container. Thus if one defines an "effective" or "total" mean free path (ℓ_{total}) as (15):

$$\ell_{total} = \bar{v} \tau_{total} = \frac{\ell_c L}{L} \quad [6]$$

where $\ell_c = \bar{v} \tau_c$ is the usual mean free path, being \bar{v} and τ_c , the mean molecular velocity and the mean time between collisions, the coefficient of viscosity will drop to zero as the density diminishes. Furthermore, if ℓ_c is substituted by ℓ_{total} in equation [4], and the resulting differential equation solved, the same expression previously obtained in (2) for the viscosimetric cross section of a gas is regained (see Appendix I), while the plot of P_{zx} vs. density tends to zero as $n \rightarrow 0$, and the curve only shifts to higher pressures preserving its original shape.

In what respect to the boundary conditions (point 4 above) a numerical evaluation of P_{zx} for constant, sinusoidal and random boundary conditions has been recently made for the specific case of two concentric cylinders with constant relative motion, which resemble the cylindrical geometry of the viscometer used in experimental measurements (point 3 above). As those calculations showed, the detailed variation of the viscosity with respect to pressure is naturally modified, but the periodicity of the oscillations does not change, preserving the general pattern of variation fairly unchanged (6). In any case arguments in favor of the near constancy of the boundary conditions (a_0 and a_1 in Eq. [5]) for gases in the transition regime had already been given (1).

Taking into account all discussed factors, it still remains that the series development of P_{zx} may be still short at third order. This leads us to a more thoughtful consideration of the series development of the stress tensor previously employed. According to the authors of this paper, the order up to which the series development of P_{zx} should be preserved might be set up through direct contrast with the experimental results. Thus, for the continuous case the first order term is enough, and it is experimentally found that the coefficient of

viscosity does not depend on density for pressures between 1 atm and 0.01 atm (15). Due to the inability of this expression to justify the oscillations of viscosity found in the transition regime, a third order term is maintained for this specific case, and eq. [5] results. If other experimental phenomena (such as the *detailed* variation of effective viscosity with pressure in the transition regime) cannot be explained by the third order expression, it would be useful to study higher order contributions.

At this point however, it is important to remark that a series development of P_{zx} in terms of the mean free path ℓ_c and the spatial derivatives of the gas velocity may not be convergent. On the other hand, if the effective mean free path given by Eq. [6] is used in the series development of P_{zx} , all the coefficients of the series, except the first, present a maximum and remain bound as the pressure drops to zero (see Appendix II).

The present paper presents an infinite series development of the stress tensor of a gas in the transition regime in terms of the spatial derivatives of the velocity of the gas. Additionally, the resolution of the fifth order differential equation found for P_{zx} under constant boundary conditions is also given.

Results and Discussion

Path integral formulation

Under the path integral formulation the distribution function is given by:

$$f(\mathbf{r}, \mathbf{v}, t) = \int f^0(\mathbf{r}_0, \mathbf{v}_0, t - \tau) d(e^{-\tau/\tau_c}), \quad [7]$$

which integrated by parts gives:

$$f(\mathbf{r}, \mathbf{v}, t) = f^0(\mathbf{r}_0, \mathbf{v}_0, t) - \int_0^t \frac{df^0}{dt}(\mathbf{r}_0, \mathbf{v}_0, t - t) e^{-t/\tau_c} dt. \quad [8]$$

It is supposed that f^0 could be taken as a maxwellian distribution:

$$f^0 = g(U_x, U_y, U_z) \frac{n(m\beta / 2\pi)^{3/2}}{\exp(\beta m U^2 / 2)}, \quad [9]$$

where $U_x = v_x - u_x(z)$, $U_y = v_y$, $U_z = v_z$.

$$\frac{dg}{dt} = \frac{g}{U_x} \frac{u_x}{z} v_z, \quad [10]$$

$$\text{and, } \frac{dz}{dt} = \frac{dz}{dt_0} t - t_0. \quad [11]$$

Then expression [7] gives:

$$f(\mathbf{r}, \mathbf{v}, t) = g(U_x, U_y, U_z) \int \frac{g}{U_x} \frac{u_x}{z} v_z e^{-t/\tau} dt. \quad [12]$$

The last integral in Eq. [12] can be integrated by parts to obtain:

$$f(\mathbf{r}, \mathbf{v}, t) = g(U_x, U_y, U_z) \left[\frac{g}{U_x} \frac{u_x}{z} v_z \tau - \frac{2g}{U_x^2} \frac{u_x}{z} v_z^2 - \frac{g}{U_x} \frac{2u_x}{z^2} v_z^2 e^{-t/\tau} dt \right]. \quad [13]$$

Now since the factor $e^{-t/\tau} dt$ still persists we can integrate by parts four more times to get:

$$f(\mathbf{r}, \mathbf{v}, t) = g(U_x, U_y, U_z) \left[\frac{g}{U_x} \frac{u_x}{z} v_z \tau - \frac{2g}{U_x^2} \frac{u_x}{z} v_z^2 \tau^2 - \frac{g}{U_x} \frac{2u_x}{z^2} v_z^2 \tau^3 - \frac{3g}{U_x^3} \frac{u_x}{z} v_z^3 \tau^3 - \frac{g}{U_x} \frac{3u_x}{z^3} v_z^3 \tau^3 - 3 \frac{2g}{U_x^2} \frac{2u_x}{z^2} \frac{u_x}{z} v_z^3 \tau^3 - 6 \frac{3g}{U_x^3} \frac{2u_x}{z^2} \frac{2u_x}{z^2} v_z^4 \tau^4 - 3 \frac{2g}{U_x^2} \frac{2u_x}{z^2} v_z^4 \tau^4 - 4 \frac{2g}{U_x^3} \frac{3u_x}{z^3} \frac{u_x}{z} v_z^4 \tau^4 - \frac{4g}{U_x^4} \frac{u_x}{z} v_z^4 \tau^4 - \frac{g}{U_x} \frac{4u_x}{z^4} v_z^4 \tau^4 - 12 \frac{3g}{U_x^3} \frac{u_x}{z} \frac{2u_x}{z^2} v_z^5 \tau^5 - 3 \frac{3g}{U_x^3} \frac{u_x}{z} \frac{2u_x}{z^2} v_z^5 \tau^5 \right]$$

$$10 \frac{3g}{U_x^3} \frac{3u_x}{z^3} \frac{u_x}{z} v_z^5 \tau^5 - 10 \frac{4g}{U_x^4} \frac{2u_x}{z^2} \frac{u_x}{z} v_z^5 \tau^5 - 10 \frac{2g}{U_x^2} \frac{3u_x}{z^3} \frac{2u_x}{z^2} v_z^5 \tau^5 - 5 \frac{2g}{U_x^2} \frac{4u_x}{z^4} \frac{u_x}{z} v_z^5 \tau^5 - \frac{5g}{U_x^5} \frac{u_x}{z} v_z^5 \tau^5 - \frac{g}{U_x} \frac{5u_x}{z^5} v_z^5 \tau^5. \quad [14]$$

The expression for the stress tensor is (15):

$$P_{zx} = m \int d^3 U f(\mathbf{r}, \mathbf{v}, t) U_z U_x \quad [15]$$

In order to calculate P_{zx} from Eqs. [14] and [15] the derivatives of the function g should be evaluated:

$$\begin{aligned} \frac{g}{U_x} &= \beta m U_x g - \frac{2g}{U_x^2} = \beta m g - (\beta m u_x)^2 g \\ \frac{3g}{U_x^3} &= 3(\beta m)^2 U_x g - (\beta m U_x)^3 g \\ \frac{4g}{U_x^4} &= 3(\beta m)^2 g - 6(\beta m)^3 U_x^2 g - (\beta m U_x)^4 g \\ \frac{5g}{U_x^5} &= 15(\beta m)^3 U_x g - 10(\beta m)^4 U_x^3 g - (\beta m U_x)^5 g. \end{aligned} \quad [16]$$

Since $U_z = v_z$ all the terms of the series [14] which are even in v_z give rise to odd integrands of U_z when substituted in equation [15] and vanish when integrated in the interval $(-\infty, +\infty)$. All the terms which contain even spatial derivatives of g also disappear, since these derivatives are even in U_z (as a matter of fact do not contain U_z) and also give rise to odd integrands. Finally, the rest of the terms that are product of spatial derivatives of different order in u_x cancel out when the appropriate derivative of g is substituted and the integrals evaluated (See Appendix III). If one calls f_{zx} the sum of all the terms of f which produce non vanishing contributions to P_{zx} , then:

$$f_{zx} = v_z \tau \left[\frac{g}{U_x} \frac{u_x}{z} v_z^3 \tau^3 - \frac{g}{U_x} \frac{3u_x}{z^3} v_z^3 \tau^3 - \frac{g}{U_x} \frac{5u_x}{z^5} v_z^5 \tau^5 \right]. \quad [17]$$

By induction it is seen that for n^{th} order:

$$f_{zx} = \sum_{i=1,3,5,7}^n (v_z \tau)^i (\beta m U_x g) \frac{u_x}{z^i} \quad [18]$$

Inserting this expression in equation [15] we get:

$$P_{zx} = \sum_i^{ODD} m^2 n \beta \tau^i \frac{m \beta}{2\pi} \frac{u_x}{z^i} \quad [19]$$

$$d^3 U U_x^2 U_z^{i-1} \exp(\beta m U^2 / 2)$$

where

$$d^3 U U_x^2 U_z^{i-1} \exp(\beta m U^2 / 2)$$

$$dU_y \exp(\beta m U_y^2 / 2) dU_x U_x^2 \exp(\beta m U_x^2 / 2)$$

$$dU_z U_z^{i-1} \exp(\beta m U_z^2 / 2)$$

$$\pi^{1/2} \frac{\beta m}{2}^{1/2} \frac{\pi^{1/2}}{2} \frac{\beta m}{2}^{3/2} \frac{\beta m}{2} \frac{1}{z} \quad [20]$$

$$\int_0^\infty dx x^{i-1} \exp(-x^2).$$

Let us call $M(i)$ the remaining integral of equation [20]:

$$M(i) = \int_0^\infty dx x^{i-1} \exp(-x^2) \quad [21]$$

$$2 \int_0^\infty dx x^{i-1} \exp(-x^2) = \Gamma(i/2),$$

where Γ is the well known gamma function which has the following properties:

1. $\Gamma(j) = (j-1)\Gamma(j-1)$
2. $\Gamma(1/2) = \sqrt{\pi}$
3. $\Gamma(1) = 1$ [22]

Using [22]:

$$M(i) = \sqrt{\pi} 2^{(i-1)/2} \sum_{p=0}^{(i-1)/2} (2p-1) \quad [23]$$

Inserting Eq. [21] in [20] and this result in [19], one gets:

$$P_{zx} = \sum_{p=0}^{(i-1)/2} m^{(i-1)/2} n \beta^{(i-1)/2} \tau^i \frac{u_x}{z^i} \quad [24]$$

Changing the index so that the summatory runs on all positive integers ($i=2j+1$):

$$P_{zx} = \sum_j^{(d-1)/2} m^j n \beta^{(j-1)} \tau^{2j+1} \frac{u_x}{z^{2j+1}} \quad [25]$$

where d is the degree up to which the series would be preserved, and j runs over all positive integers starting with zero.

Finally taking into account that $\beta = 1/kT$ and $\tau_{total} v = l_{total}$ a series expression for P_{zx} is reached:

$$P_{zx} = nmv \sum_j^{(d-1)/2} \frac{l_c L}{L} \frac{\tau^{2j+1}}{8} \frac{u_x}{z^{2j+1}} \quad [26]$$

Resolution of the differential equation of order d

Once d is selected, Eq. [26] becomes a differential equation in u_x , which can be solved when the stress tensor is independent of the distance z . This laminar flow condition is justified if the period of the viscometer is chosen large enough to avoid turbulent flow. Whenever this holds true, the resulting differential equation is a non homogeneous ordinary differential equation of order d with constant coefficients. The solution of the correspondent homogeneous equation is (19):

$$u_x = \sum_{i=1}^d c_i \exp(\lambda_i z) \quad [27]$$

where the lambdas (λ) are solutions of the characteristic equation of the correspondent homogeneous differential equation.

A particular solution of Eq. [26] cut off at order d , could be easily chosen as $-P_{zx}z/\eta$. The general solution of equation [26] is then:

$$u_x = \sum_{i=1}^d c_i \exp(\lambda_i z) - P_{zx} \frac{z}{\eta} \quad [28]$$

In order to get P_{zx} which in above equation is one of the coefficients, boundary conditions on u_x and its derivatives should be selected. If the value of the velocity u_x and its derivatives are established at $z=0$ and $z=L$, then differentiating Eq. [28] $(d-1)/2$ times, gives $d+1$ algebraic equations with $d+1$ unknowns. Thus an expression for the stress tensor can be deduced.

For first order, the resulting homogeneous differential equation is:

$$0 = \eta \frac{u_x}{z} \quad [29]$$

the characteristic equation is $\lambda = 0$, and the general solution is:

$$u_x = c_1 z \frac{P_{zx}}{\eta} \quad [30]$$

If $u_x(z=L) = u_L$, and $u_x(z=0) = u_0$, the following system of equations results:

$$\begin{aligned} u_L &= c_1 P_{zx} L / \eta, \\ u_0 &= c_1. \end{aligned} \quad [31]$$

Hence:

$$P_{zx} = \eta(u_L - u_0) / L. \quad [32]$$

being [32] the usual expression for the tangential stress in the continuous regime, η being the usual coefficient of shear viscosity.

For third order the homogeneous differential equation is:

$$0 = \eta \frac{u_x}{z} - \gamma \frac{u_x^3}{z^3} \quad [33]$$

and the characteristic equation is:

$$\eta \lambda - \gamma \lambda^3 = 0. \quad [34]$$

Solving Eq. [34] one gets: $\lambda_1 = i(\eta/\gamma)^{1/2}$, $\lambda_2 = -i(\eta/\gamma)^{1/2}$, and $\lambda_3 = 0$.

The general solution of Eq. [33] is therefore:

$$u_x = c_1 \exp(\lambda_1 z) + c_2 \exp(\lambda_2 z) - P_{zx} \frac{z}{\eta} + c_4. \quad [35]$$

Differentiating Eq. [35], and specifying the boundary conditions for u_x and u_x' at $z=L$ and $z=0$, a system of four equations with 4 unknowns is obtained, from which the expression of P_{zx} given in Eq. [5] is obtained ($\omega = i\lambda_2$).

As a final example it might be noticed that for fifth order, the procedure outlined in the previous section leads to:

$$u_x(z) = c_1 e^{\lambda_1 z} + c_2 e^{\lambda_2 z} + c_3 e^{\lambda_3 z} + c_4 e^{\lambda_4 z} + c_5 e^{\lambda_5 z} - z P_{zx} / \eta \quad [36]$$

where the lambdas are the solutions of the characteristic equation:

$$\alpha \lambda - \lambda^4 - (\gamma/\alpha) \lambda^2 - (\eta/\alpha) = 0 \quad [37]$$

where:

$$S^2 = \frac{\gamma}{\alpha} S - \eta/\alpha = 0, \quad S^2 = \lambda^4 \quad [38]$$

$$\lambda = \frac{\gamma \sqrt{\gamma^2 - 4\eta\alpha}}{2\alpha}^{1/2}, \quad [39]$$

and α is the coefficient of the fifth order derivative in Eq. [26], and $\lambda_5 = 0$.

The general expression of the stress tensor generated from the correspondent system of six equations (u_x, u_x, u_x for $z=L$ and $z=0$) is:

$$P_{zx} = N / D \quad [40]$$

where:

$$\begin{aligned} N = & \eta \frac{\Delta u}{L} [\exp(2AL/l) - \exp(-2AL/l)] \\ & [A^4 B^2 - 2A^2 B^4 + B^6][L] \\ & [2A^6 - 6A^4 B^2 + 6A^2 B^4 - 2B^6][L] \\ & [\cos(2BL/l)][2A^6 - 4A^4 B^2 + 2A^2 B^4][L] \\ & \eta^2 (a_0 - a_L) l [\exp(AL/l) - \exp(-AL/l)] \\ & [\cos(BL/l)] [2A^3 B^2 - 2AB^4] \\ & [\exp(2AL/l) - \exp(-2AL/l)] [A^3 B^2 - AB^4] \\ & [2A^4 B - 2A^2 B^3][\sin(2BL/l)] \\ & \eta (b_L - b_0) l^2 [\exp(AL/l) - \exp(-AL/l)] \\ & [\sin(BL/l)] [4A^3 B - 4AB^3] \\ & [\exp(2AL/l) - \exp(-2AL/l)] \\ & [A^2 B^2 - B^4] [2A^4 - 2B^4] \\ & 2[A^4 - A^2 B^2][\cos(2BL/l)] \end{aligned} \quad [41]$$

and

$$\begin{aligned} D = & [\exp(AL/l) - \exp(-AL/l)] \\ & [\cos(BL/l)][8A^3 B^2 l - 8AB^4 l] \\ & [\exp(AL/l) - \exp(-AL/l)] \\ & [\sin(BL/l)][8A^4 B l - 8A^2 B^3 l] \\ & [\exp(2AL/l) - \exp(-2AL/l)] \\ & [A^4 B^2 L - 2A^2 B^4 L + B^6 L] \\ & [\exp(2AL/l) - \exp(-2AL/l)] \\ & [4A^3 B^2 l - 4AB^4 l] \\ & [2A^6 L - 6A^4 B^2 L + 6A^2 B^4 L - 2B^6 L] \\ & [\cos(2BL/l)][2A^6 L - 4A^4 B^2 L + 2A^2 B^4 L] \\ & [\sin(2BL/l)][8A^4 B l - 8A^2 B^3 l] \end{aligned} \quad [42]$$

where $A = 0.44880405$, $B = 0.67533176$, and $b_L = u_x(z=L)$ and $b_0 = u_x(z=0)$.

Notice that the boundary conditions at each wall of the viscometer appear as sums in Eq. [41], so that only the values of Δu , ($a_0 + a_L$) and $(b_0 + b_L)$ are needed instead of the six boundary conditions that should be in principle required for a fifth order development of P_{zx} . The behavior of this high order expression of P_{zx} as a function of density along with its dependence under distinct boundary conditions will be given in following articles.

Conclusions

As shown in this article, the use of the Boltzmann equation under the time relaxation approximation allows to deduce a general series expression for the stress tensor for a gas. Appropriate cut off of the series to a given order open the possibility of a systematic study of the influence of high order terms in the behavior of the stress tensor, as well as the search of qualitative criteria in order to bound this series in an appropriate way for different regimes.

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Appendix I

Separation in density of the regions of non monotonic variation of the gas viscosity, employing a suitable expression for the mean free path in the transition regime

As was previously found [2] the regions of rapid variation in a P_{zx} vs pressure plot satisfy the following condition:

$$\Delta\omega L = 2\pi \quad [I.1]$$

Now if the Eq. [6] is used to calculate $\omega = (\eta/\gamma)^{1/2}$ and this expression is substituted in the left handside of Eq. [I.1], one gets:

$$\Delta\omega L = \frac{8}{3\pi} \left(\frac{L}{l_c} \right)^{1/2} \frac{L}{l_c} = \frac{8}{3\pi} \frac{L}{l_c} \left(\frac{L}{l_c} \right)^{1/2}$$

Introducing $l_c = (2^{1/2} \sigma n)^{-1}$ in the above equation, the known expression for the viscosimetric cross reported in references (3) and (7) is reproduced:

$$\Delta n \sigma = \frac{1}{(4/3\pi^3)^{1/2} L} \quad [I.2]$$

where $\Delta n = n - n_0$.

Thus, the approximate expression of the mean free path for low pressures given by Eq. [3] does not change the separation in density of the regions of non-monotonic variation of the shear viscosity (Δn).

Appendix II

Mathematical form of the maxima of the coefficients of Eq. [23]

If usual expression of the mean free path (l_c) is used instead of Eq. [6], the coefficients of the series development of the stress tensor go to infinity very rapidly as the pressure diminishes. Though the value at which

they get out of bounds depends on the cross section of the gas, it is usual that at $T = 300$ K this pressure stays around $10 \mu\text{mHg}$ (i.e. for He it is located around $17 \mu\text{mHg}$ and for N_2 it is around $8 \mu\text{mHg}$). If the wall correction on the mean free path is taken into account, the coefficients increase toward a maximum but then drop towards zero as pressure is further decreased.

As was previously noted (Eq. [26]), the referred coefficients have the following form:

$$mv \frac{1}{\sigma l_c \sqrt{2}} \frac{l_c L}{l_c L} \frac{\pi}{8} \frac{d-1}{2} \frac{(d-1)^{d-1}}{p^0} (2p-1) \quad \text{[II.1]}$$

where d is again the order of the coefficient ($d=1,3,5\dots$). In order to get the position of the extrema, we have the condition:

$$\frac{1}{l_c^2} \frac{l_c L}{l_c L} \frac{d}{l_c} \frac{l_c L}{l_c L} \frac{d-1}{L} \frac{L}{l_c L} = 0 \quad \text{[II.2]}$$

After some arithmetic, the expression:

$$l_c = L(d-1) \quad \text{[II.3]}$$

is obtained; which in terms of the pressure is equal to:

$$P = \frac{KT}{\sigma \sqrt{2}} \frac{1}{L(d-1)} \quad \text{[II.4]}$$

As can be deduced from [II.3] and [II.4], the coefficient of first order do not present a maximum value but instead, decreases monotonically with the gas density. The position of the maxima of the rest of the coefficients moves towards zero as the order of the coefficient increases. For most gases, these maxima lay between $1.5 \mu\text{mHg}$ and $0 \mu\text{mHg}$.

Introducing Eq. [II.3] in Eq. [II.1], one gets for the coefficient of order d :

$$mv \frac{1}{\sigma L (d-1) \sqrt{2}} \frac{(d-1)L^2}{L (d-1) L} \frac{\pi}{8} \frac{d-1}{2} \frac{(d-1)^{d-1}}{p^0} (2p-1) \quad \text{[II.5]}$$

From which the final expression for the maximum value of the coefficient can be deduced:

$$\frac{mv}{\sigma \sqrt{2}} \frac{L (d-1)^{d-1}}{d^d} \frac{\pi}{8} \frac{d-1}{2} \frac{(d-1)^{d-1}}{p^0} (2p-1) \quad \text{[II.6]}$$

Figure 1 illustrates the dependence on pressure of the coefficients (given in Eq. [II.1]) of the terms contributing to P_{zx} (Eq. [26]) up to fifth order. This figure corresponds to a helium gas at $T = 298$ K, confined between solid surfaces separated by a distance $L = 2$ cm. According to this figure the first order coefficient largely predominates for pressures beyond $50 \mu\text{mHg}$, and this tendency increases for still larger pressures. In the continuous regime it may then be expected that only the first order term contributes to P_{zx} . In the transition regime it can be seen in this figure that the inclusion of the third order term may be expected to be adequate to describe P_{zx} for pressures larger than $\sim 20 \mu\text{mHg}$. Still for pressures below this value the fifth order term should be included. Moreover, according to our numerical results (which are not illustrated in Figure 1) for pressures below $10 \mu\text{mHg}$ still higher order terms also contribute to P_{zx} in a sizable way. However, the important feature of the oscillations of viscosity with pressure may be expected to occur according to our model beyond $20 \mu\text{mHg}$ for the system corresponding to Figure 1. Thus the cut off of the series for P_{zx} up to third, or even fifth order, seems to be appropriate for the description of the expected oscillations. Experimental results tend to confirm this view.

The same coefficients presented in Figure 1 are shown in Figure 2, but this time

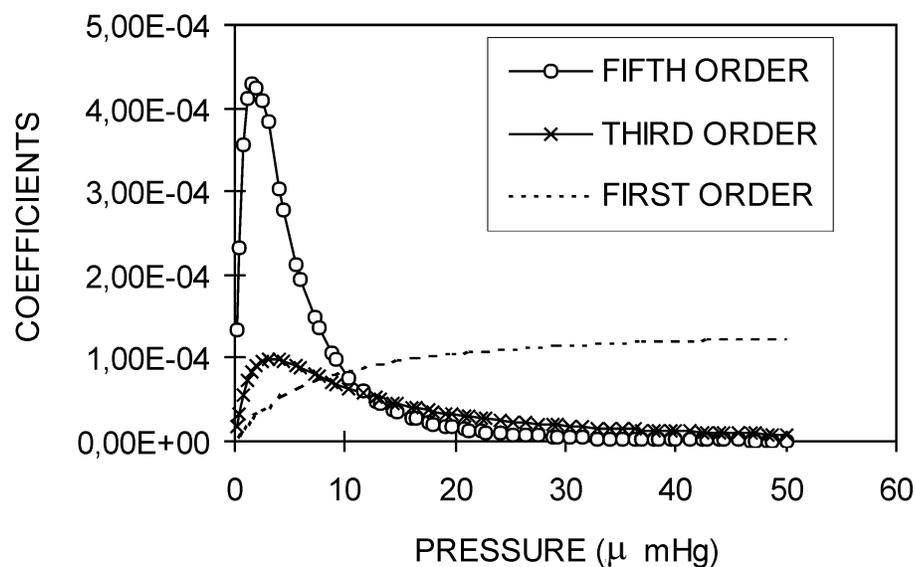


Figure 1. Coefficients up to fifth order in the spatial derivative of the fluid velocity in the expression for P_{zx} as a function of pressure, Eq. [II.1]. The numerical values corresponds to a Helium gas at $T = 298$ K between solid surfaces separated by a distance $L = 2$ cm.

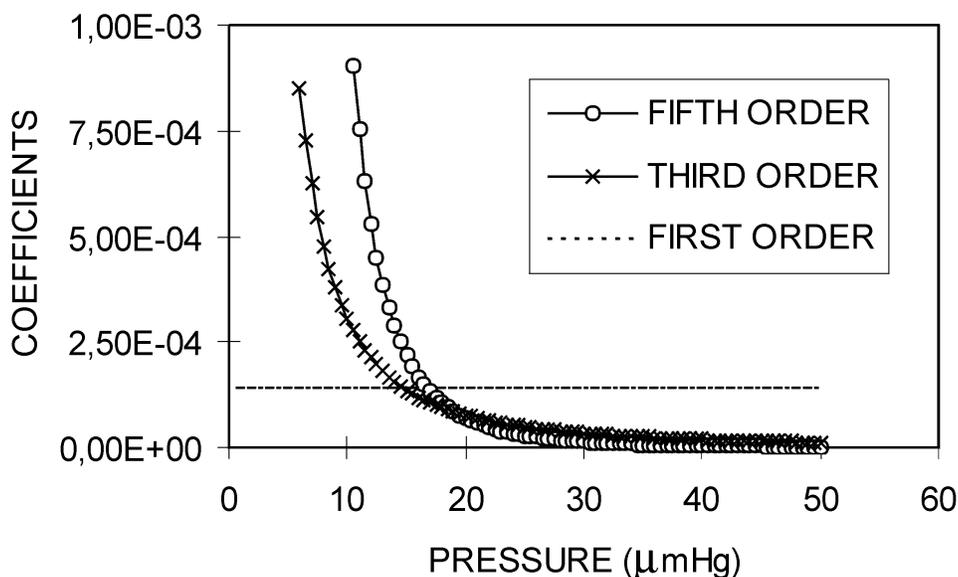


Figure 2. The same coefficients vs. pressure illustrated in Fig. 1 but this time excluding the wall correction for the mean free path (see Eq. [6]).

calculated excluding the wall correction for the mean free path, i.e. calculated with ℓ_c instead of ℓ_{total} , Eq. [6]. Two important features should be remarked: (i) The inclusion of the

wall correction avoids the divergence of the coefficients as the pressure goes to zero; (ii) The region where the high order coefficients are relevant is more extended when the wall

correction is not included. This clearly shows the two-fold importance of the wall correction for the mean free path.

Appendix III

Vanishing integrals of equation [14]

After all the considerations concerning the parity of the variables of integration in Eq. [14] are made, the following high order terms still survive (those integrals of 3rd or lower order are already calculated in reference (2)):

$$1. \quad 15 \cdot 3 (\beta m)^2 U_x g \quad (\beta m U_x)^3 g \quad \frac{u_x}{z} \quad \frac{u_x}{z} \quad v_z^5 \tau^5 \quad \text{[III.1]}$$

$$2. \quad 10 \cdot 3 (\beta m)^2 U_x g \quad (\beta m U_x)^3 g \quad \frac{u_x}{z^3} \quad \frac{u_x}{z} \quad v_z^5 \tau^5 \quad \text{[III.2]}$$

$$3. \quad \frac{15 (\beta m)^3 U_x g \quad 10 (\beta m)^4 U_x^3 g \quad (\beta m U_x)^5 g}{\frac{u_x}{z} \quad v_z^5 \tau^5} \quad \text{[III.3]}$$

$$4. \quad \beta m U_x g \quad \frac{u_x}{z^5} \quad v_z^5 \tau^5 \quad \text{[III.4]}$$

Introducing the appropriate expression for the function $g(U_x, U_y, U_z)$, the first term (III.1) is equal to:

$$\begin{aligned} & 45 m^3 \beta^2 n \frac{m \beta}{2 \pi} \tau^{3/2} \tau^5 \\ & dU_y \exp(\beta m U_y^2 / 2) \quad dU_x U_x^2 \exp(\beta m U_x^2 / 2) \\ & dU_z U_z^6 \exp(\beta m U_z^2 / 2) \quad 15 m^4 \beta^3 n \frac{m \beta}{2 \pi} \tau^{3/2} \tau^5 \\ & dU_y \exp(\beta m U_y^2 / 2) \quad dU_x U_x^4 \exp(\beta m U_x^2 / 2) \\ & dU_z U_z^6 \exp(\beta m U_z^2 / 2) \\ & 45 m^3 \beta^2 n \frac{m \beta}{2 \pi} \tau^{3/2} \tau^5 \pi^{1/2} \frac{\beta m}{2} \tau^{1/2} \\ & \frac{\pi^{1/2}}{2} \frac{\beta m}{2} \tau^{3/2} \quad 15 \frac{\pi^{1/2}}{8} \frac{\beta m}{2} \tau^{7/2} \end{aligned}$$

$$\begin{aligned} & 15 m^4 \beta^3 n \frac{m \beta}{2 \pi} \tau^{3/2} \tau^5 \quad \pi^{1/2} \frac{\beta m}{2} \tau^{1/2} \\ & \frac{3 \pi^{1/2}}{4} \frac{\beta m}{2} \tau^{5/2} \quad 15 \frac{\pi^{1/2}}{8} \frac{\beta m}{2} \tau^{7/2} \quad 0 \end{aligned}$$

Expression [III.2] is equal to:

$$\begin{aligned} & 30 m^3 \beta^2 n \frac{m \beta}{2 \pi} \tau^{3/2} \tau^5 \\ & dU_y \exp(\beta m U_y^2 / 2) \quad dU_x U_x^2 \exp(\beta m U_x^2 / 2) \\ & dU_z U_z^6 \exp(\beta m U_z^2 / 2) \quad 10 m^4 \beta^3 n \frac{m \beta}{2 \pi} \tau^{3/2} \tau^5 \\ & dU_y \exp(\beta m U_y^2 / 2) \quad dU_x U_x^4 \exp(\beta m U_x^2 / 2) \\ & dU_z U_z^6 \exp(\beta m U_z^2 / 2) \\ & 30 m^3 \beta^2 n \frac{m \beta}{2 \pi} \tau^{3/2} \tau^5 \pi^{1/2} \frac{\beta m}{2} \tau^{1/2} \\ & \frac{\pi^{1/2}}{2} \frac{\beta m}{2} \tau^{3/2} \quad 15 \frac{\pi^{1/2}}{8} \frac{\beta m}{2} \tau^{7/2} \\ & 10 m^4 \beta^3 n \frac{m \beta}{2 \pi} \tau^{3/2} \tau^5 \quad \pi^{1/2} \frac{\beta m}{2} \tau^{1/2} \\ & \frac{3 \pi^{1/2}}{4} \frac{\beta m}{2} \tau^{5/2} \quad 15 \frac{\pi^{1/2}}{8} \frac{\beta m}{2} \tau^{7/2} \quad 0 \end{aligned}$$

Eq. [III.3] is equal to:

$$\begin{aligned} & 15 m^4 \beta^3 n \frac{m \beta}{2 \pi} \tau^{3/2} \tau^5 \\ & dU_y \exp(\beta m U_y^2 / 2) \quad dU_x U_x^2 \exp(\beta m U_x^2 / 2) \\ & dU_z U_z^6 \exp(\beta m U_z^2 / 2) \quad 10 m^5 \beta^4 n \frac{m \beta}{2 \pi} \tau^{3/2} \tau^5 \\ & dU_y \exp(\beta m U_y^2 / 2) \quad dU_x U_x^4 \exp(\beta m U_x^2 / 2) \\ & dU_z U_z^6 \exp(\beta m U_z^2 / 2) \\ & m^6 \beta^5 n \frac{m \beta}{2 \pi} \tau^{3/2} \tau^5 \end{aligned}$$

$$dU_y \exp(\beta m U_y^2 / 2) \quad dU_x U_x^6 \exp(\beta m U_x^2 / 2)$$

$$dU_z U_z^6 \exp(\beta m U_z^2 / 2)$$

$$15 m^4 \beta^3 n \frac{m \beta}{2 \pi} \tau^5 \pi^{1/2} \frac{\beta m}{2}^{1/2}$$

$$\frac{\pi^{1/2}}{2} \frac{\beta m}{2}^{3/2} \quad 15 \frac{\pi^{1/2}}{8} \frac{\beta m}{2}^{7/2}$$

$$10 m^5 \beta^4 n \frac{m \beta}{2 \pi} \tau^5 \pi^{1/2} \frac{\beta m}{2}^{1/2}$$

$$\frac{3 \pi^{1/2}}{4} \frac{\beta m}{2}^{5/2} \quad 15 \frac{\pi^{1/2}}{8} \frac{\beta m}{2}^{7/2}$$

$$m^6 \beta^5 n \frac{m \beta}{2 \pi} \tau^5 \pi^{1/2} \frac{\beta m}{2}^{1/2}$$

$$\frac{15 \pi^{1/2}}{8} \frac{\beta m}{2}^{3/2} \quad 15 \frac{\pi^{1/2}}{8} \frac{\beta m}{2}^{7/2} \quad 0$$

Finally, of all expressions [III.1] to [III.4], expression [III.4] constitute the only higher-order non-vanishing term, and therefore is explicitly taken into account in the series development of P_{zx} .