The prediction of helium gas viscosity under high pressure and high temperature with the Chapman-Enskog solution and excess viscosity

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Abstract. The purpose of this work is to predict a helium gas viscosity under high pressure and high temperature for practical industrial uses. The suitable force constants and a collision integral for the Chapman-Enskog solution to estimate viscosity in the limit of zero density were recommended by the present author. At high density, modification of the Arp and McCarty extrapolation equation for excess viscosity was applied. A combination of the Chapman-Enskog solution and modification of the Arp and McCarty excess viscosity gives an estimation of helium gas viscosity within 2 to 5 % deviation from the existing experimental data under high-temperature and high-pressure region.

1. Introduction
Helium (Helium-4) is a noble gas that mostly uses in the industrial purpose. There is a need for recommendations relating to the best available viscosity data and the best available correlations for practical purposes under high pressure and high temperature for helium economy. Especially in a nuclear power plant generation IV, Helium is often used as a coolant in High Temperature Gas-cooled Reactors (HTGR), it is one among the other type of nuclear reactor for Generation IV (Gen-IV) of advance nuclear reactors system [1,2]. At atmospheric pressure, helium gas viscosity may be considered to be at the limit of zero density, \( \eta_o(T) \), which can be well represented by the Chapman-Enskog solution as a first approximation [3]. Besides the Chapman-Enskog solution, empirical correlations are also can be applied. However, the latest commonly represented in a complicated equation and not recommended. The present author has been succeded to predict hydrogen gas viscosity under high pressure and high temperature by the Chapman-Enskog solution and the excess viscosity within 2 to 4 % of deviations [4]. The same method with previous work will be applied for helium.

2. Existing experimental data
Helium is the second lightest after hydrogen. Moreover helium is a colorless, odorless and tasteless. It introduced difficulties to be measure in an accurate way due to leakage. There are several experimental techniques available to obtain helium gas viscosity data. Table 1 show the selected experimental data which are used to be compared with the present correlation. The highest
temperature of about 900 K obtained by Makavetskas with the capillary method. The highest pressure
of 100 MPa have done by team of Hydrogenius in Japan with the same method with Makavetskas.
Measuring viscosity at high pressure and temperature is often measured with capillary tube.

| Table 1. Selected experimental data set |
|----------------------------------------|
| References/Methods                    | Uncer./% | T/K    | P/MPa     |
| Seibt 2009 [5]/Vibrating-wire          | 0.25 to 0.3 | 293    | up to 30 |
| Ross (1957) [6]/Capillary tube         | 1        | 298 to 323 | 3.5 to 68.9 |
| Gracki (1969) [7]/Capillary tube       | 0.1 – 0.2 | -373 to 298 | 1.5 to 17.5 |
| Trauts (1930) [8]/Capillary tube       | 2        | 293 to 523 | 0.1      |
| Makavetskas (1963) [9]/Capillary tube  | -        | 283 to 918 | 5-51     |
| Enrique (1966) [10]/transpiration      | ns       | 373 to 473 | up to 82 |
| Kao&Kobayashi (1967) [11]/Capillary tube | 0.15       | 183 to 323 | 1 to 50  |
| Tsederberg (1969) [12]/Capillary tube  | 3        | 292     | 5 to 29  |
| Kestin&Leidenforst (1959) [13]/Oscillation | 0.2 – 0.5    | 298 to 510 | 0.1 to 13,7 |
| Evers (2002) [14]/Oscillating disk and oscillating wire | 0.6-1.0 | 233 to 523 | Up to 30 |
| Flynn (1963) [15]/Capillary flow       | 0.1      | 223 to 523 | Up to 30  |
| HYDROGENIUS [16]/Capillary method (2012) | 2        | 295-500 | Up to 100 |

3. The Enskoq theory

We can have assumptions that a gas can be treated as an ensemble of hard elastic spheres and multiple collisions can be neglected. $\psi$ is the collisional transfer of molecular property across an element of area $dS$ at point $r$. Then the collisional part of the vector of flow of $\psi$ is found to be

$$
\frac{1}{2} \chi \sigma^3 \int \int \int m(\psi - \psi) f f_i k(g,k) dk dc d c_1 + \frac{1}{4} \chi \sigma^4 \int \int \int (\psi - \psi) f f_i k. \nabla \ln(f/f_i) k(g,k) dk dc d c_1
$$

(1)

Where all the quantities are evaluated at the point $r$ since the second term of this already involves space-derivatives, $f$ and $f_i$ can here be replaced by $f^{(0)}$ and $f_i^{(0)}$. Consider first the transfer of momentum. Let

$$
\psi = mC = m(c - c_0)
$$

(2)

Where $c_0$ is evaluated at the special point $r$ under consideration, and does not vary with the position of the molecule, the collisional part becomes

$$
\frac{1}{2} \chi \sigma^3 \int \int \int m(C - C) f f_i^{(0)} k(g,k) dk dc d c_1 + \frac{1}{4} \chi \sigma^4 \int \int \int (C - C) f f_i^{(0)} k. \nabla \ln(f^{(0)}/f_i^{(0)}) k(g,k) dk dc d c_1
$$

(3)

We use the relation $f = f^{(0)} \equiv n \left( \frac{m}{2 \pi kT} \right)^{3/2} e^{-m c^2/2 k T}$, after integration with respect to $k$, using

$$
\int \left( C - C \right) k(g,k) dk = \int k k(g,k)^2 dk = \frac{2c}{15} (2g + g^2 U)
$$

(4)

$$
\int (C - C) k k(v,k)(g,k) dk = \int k k(v,k)(g,k)^2 dk = \frac{R}{15} \left[ v. g \left( \frac{(g g + g^2 U)}{g} \right) + g(v g + g v) \right]
$$

(5)

The expression becomes
\[
\frac{\pi}{15} \chi \sigma^3 m \iint f_1 (2gg + Ug^2) d\xi d\eta + \frac{\pi}{48} \chi^4 \sigma m \iint f_1^{(0)} f_1^{(0)} \left[ \left\{ g \nabla \ln \left( \frac{f_1^{(0)} f_1^{(0)}}{\xi^4} \right) \frac{(gg + g^2u)}{g} + g \nabla \ln \left( \frac{f_1^{(0)}}{\xi^4} \right) \right\} d\xi d\eta \right]
\]

In the first integral, since for any function \( \varphi \)
\[
\int f \phi d\xi = \int f_1 \phi_1 d\xi = n \left( \frac{m}{2 \pi kT} \right)^{3/2} \int \phi e^{-mC^2/2kT} dC = n \phi
\]
and \( g = C_1 - C \), when \( C_1 = 0, C = 0 \), The value of this integral is
\[
\frac{1}{5} \beta \rho^2 \chi (2 \overline{CC} + \overline{C^2}U)
\]

Where \( \beta = 2 \pi \sigma^3 / 3m \) and \( \rho = nm \). In the second integral, since
\[
\nabla \ln \left( \frac{f_1^{(0)}}{\xi^4} \right) = \frac{m}{2kT^2} (C^2 - C_1^2) \nabla T + \frac{m}{kT} (\nabla \xi_0)(C - C_1)
\]

In terms involving \( \nabla T \) yield odd functions of \( C, C_1 \), which vanish on integration; hence the integral is equal to
\[
- \frac{\pi}{48 \pi k} \chi^4 \sigma m \iint f_1^{(0)} f_1^{(0)} \left[ \left\{ (\nabla \xi_0 \cdot g) \frac{(gg + g^2u)}{g} + g(\nabla \xi_0 \cdot g) + g(\nabla \xi_0 \cdot g) \right\} d\xi d\eta \right]
\]

This can readily be evaluated by charging the variable of integration from \( c \) and \( c_1 \) to \( G_0 \) and \( g \), where \( G_0 = \frac{1}{2} (C + C_1) \). Its value is found to be
\[
- \overline{\omega} \left\{ \frac{\overline{\nabla \xi_0}}{5} + UV \cdot c_0 \right\} = - \overline{\omega} \left\{ \frac{2\overline{e}}{5} + UV \cdot c_0 \right\}
\]

Where
\[
\overline{\omega} = \frac{1}{9} \pi^2 \chi n^2 \sigma^4 \sqrt{mkT} = \frac{\beta \rho^2 x \sqrt{mkT}}{\pi^3 \sigma^2}
\]

Alternatively, in term of the coefficient of viscosity at ordinary pressure namely
\[
\overline{\mu} = \frac{1016 \times 5 \sqrt{mkT}}{16 \sqrt{\pi} \sigma^2}
\]

\( \overline{\omega} \) is given by
\[
\overline{\omega} = 1.002 \mu \chi b^2 \rho^2
\]

Adding (8) and (11), we obtain for the value of (7)
\[
\frac{1}{5} \beta \rho^2 \chi (2 \overline{CC} + \overline{C^2}U) - \overline{\omega} \left\{ \frac{2\overline{e}}{5} + UV \cdot c_0 \right\}
\]

This gives the part of the pressure tensor which arises from the collision transport of momentum. To do this we must add \( \rho \overline{CC} \), which gives the rate of transport by molecular motions; then the whole pressure tensor is found to be
\[ P = \rho \left( 1 + \frac{2}{5} \rho \chi \right) \bar{C} C + \frac{1}{5} \rho^2 \chi \bar{C}^2 U - \sigma \left( \frac{1}{5} \bar{c} \dot{\varepsilon} + U \nabla \cdot c_0 \right) \]  \hspace{1cm} (15)

on substituting in this the value of \( \rho \bar{C} C = k n T U \), Where \( p_0 = k n T (1 + b \rho \chi) \) it becomes

\[ P = (p_0 - \sigma \nabla \cdot c_0) U - \rho \left( 2 \mu \chi^{-1} \left( 1 + \frac{2}{5} \rho \chi \right)^2 + \frac{3}{5} \sigma \right) \dot{\varepsilon} \]  \hspace{1cm} (16)

For a gas in a uniform steady state, \( P \) reduces to a hydrostatic pressure \( p_0 \). A result already implicit. For a gas not in a uniform steady state, the hydrostatic pressure is \( p_0 - \sigma \nabla \cdot c_0 \). The extra term \(- \sigma \nabla \cdot c_0\) differs from zero whenever the density of the gas is varying; it represents a volume viscosity similar to that opposing expansion or contracting of the gas. The deviation of the pressure system from the hydrostatic is given in last term of (16), which represents the usual shear viscosity. Equating this term to \(-2 \mu' \sigma \), we see that in a dense gas the coefficient of viscosity \( \mu' \) is given in term \( \mu \), the viscosity at ordinary densities (at the same temperature), by the relation

\[ \mu' = \frac{\mu \chi^{-1} \left( 1 + \frac{2}{5} b \rho \chi \right)^2 + \frac{3}{5} \sigma}{b \rho x} \]  \hspace{1cm} (17)

The function \( \eta / \rho \) has a minimum value for \( b \rho x = 1.146 \). The values \( b \rho \) and \( b \rho x \) are obtained from the comparison of the experimental \( P - V - T \) data with Enskog equation of state

\[ P + a \rho^2 = \frac{R \rho T (1 + b \rho x)}{M} \]  \hspace{1cm} (18)

Enskog suggested \( x \) and \( b \) to be independent of temperature. The first assumption is maintained, the latter however disregarded. With the substitutions \( v_A = 1 / \rho_A, \rho_A = \rho \rho_0, a' = a \rho_0^2 \), \( b' = b \rho_0 \), Equation (18) reads

\[ P v_A = \frac{R \rho_0 T}{M} + \frac{R \rho_0 T b' \rho_A x}{M} - a' \rho_A \]  \hspace{1cm} (19)

This equation (19) can be compared with the virial equation of state

\[ P v_A = A + B \rho_A + C \rho_A^2 + \ldots \]  \hspace{1cm} (20)

gives

\[ P v_A = A + b' \rho_A x - a' \rho_A \]  \hspace{1cm} (21)

Since \( x \) approaches unity if the density approaches zero, resulting in the relations

\[ A = \frac{R \rho_0 T}{M} \]  \hspace{1cm} (22)

\[ b' - a' = B \]  \hspace{1cm} (23)

And a possible transcription of (19) as

\[ P v_A = A + B \rho_A + b' (x - 1) \rho_A \]  \hspace{1cm} (24)
With the calculation of the values \( b \rho x \) from equation (14), the coefficients of viscosity can be determined with equation (8) and the experimental value for \( \mu_0 \). Finally it is possible to calculate a value for the diameter of the molecule with the expression, given by Enskog [17,18,19,20]

\[
\sigma = \frac{1}{0.995} \sqrt{\frac{M}{RT(\eta/\rho)_{\text{min}}}}
\]

(25)

To estimate helium gas viscosity, we can predict at the limit of zero density, which can be well represented by the Chapman-Enskog solution as a first approximation

\[
\eta_0(T) = \frac{5}{16} \frac{\sqrt{\pi m k T}}{\pi \sigma^2 \Omega_\eta(T^*)}
\]

(26)

The relation need three parameters i.e. collision diameter, collision integral as a function of reduced temperature. Table 2 shows various force constants and collision integrals for helium gas.

| Table 2. Intermolecular force constants for helium gas |
|------------------------------------------------------|
| Reference | Force constant |
|  | \( \sigma \) (nm) | \( \epsilon/k \) (K) |
| Mason (Exp-six)[21] | 0.3135 | 9.16 |
| Mason (L-J) | 0.2869 | 10.22 |
| Saxena (L-J)[22] | 0.2556 | 10.22 |
| Hirschfelder et al [23] | 0.270 | 6.03 |
| Present correlation | 0.279 | 6.03 |

4. Recommendation Equation

The helium gas viscosity in the present correlation was predict within the Eq. (16).

\[
\eta(T, \rho) = \eta_0(T) + \Delta \eta(T, \rho)
\]

(27)

At low density region, the Chapman-Enskog solution was used (Eq. (26)). This equation is quite simple and accurate to predict a helium gas viscosity at low pressure. This equation depends on the selected parameters for the collision integral and intermolecular force constants. The collision integral correlation used in the present correlation obtained by Assael et. al. for universal gas [24]as shown in Eq. (28).

\[
\Omega_\eta(T^*) = \exp(0.46649 - 0.57015(\ln T^*) + 0.19164(\ln T^*)^2
\]

\[
- 0.03708(\ln T^*)^3 + 0.00241(\ln T^*)^4)
\]

(28)

For high density, the exces viscosity is used by Arp & McCarty correlation as shown in Eq. (29) [25].

\[
\Delta \eta(T, \rho) = \rho B(T) + \rho^2 C(T) + \rho^3 D(T)
\]

(29)

The B(T), C(T) and D(T) are constanta. The constant used in the present correlation is shown in Eq. (30).
\[ B(T) = -47.525/x + 87.679 - 42.074x + 0.331x^2 - 0.587x^3 \]
\[ C(T) = 547.305/x - 904.487 + 431.404x - 81.45x^2 + 5.369x^3 \]
\[ D(T) = -1684.388/x + 3331.086 - 1632.191x + 308.804x^2 - 20.191x^3 \]

(30)

Figure 1. Viscosity prediction at 0.1 MPa based on the intermolecular force constant used in Table 1. Present correlation and Arp & McCarty correlation give a difference about 2-3%.

Figure 2. The present correlation compared with the existing experiment data set and correlations for Helium gas viscosity. RUN 1 to RUN 3 is the experiment conducted by HYDROGENIUS [16]. The range is 0.1 to 100 MPa and 25 to 226 C.

Figure 1 shows a simulation of helium gas viscosity derived from various intermolecular force constant tabulated in Table 1. The discrepancy among them is up to 40%. The present correlation gives a difference about 2-3% respect to the Arp & McCarty correlation. The tendency of the present...
works agree well compared with the data obtained by the experiment (HYDROGENIUS) about 2%, and the other researchers up to 5% (Fig. 2). The present correlation is also have been compared with respect to the commercial database developed by NIST (REFPROP)[26]. The deviation is up to ±5 % compared with the present equation. Figure 3 shows a helium gas viscosity prediction with the present correlation. The prediction has been compared with the experimental data set up from 0,1 to 100 MPa, and 25 to 226 C with the same apparatus described in the Ref. 27 for hydrogen. Based on the prediction shows that for temperature above 500 K, the trend is opposite for high pressure and low pressure. The present result give a motivation to measure helium at high pressure and temperature in more accurate. The main problem of the measurement so far is about leakage because the viscosity of helium is almost a similar coefficient to hydrogen. The methods to measure helium under extreme condition should be selected very well. The capillary method was not suggesting to measure because of apparatus geometry and limitation on the pressure drop transducer device under high pressure. The suitable method to measure the viscosity is vibrating method, either curve vibrating method or straight wire.

**Figure 3.** Helium gas viscosity prediction with the present correlation up to 100 MPa, 770 K.

**Conclusion**

A new correlation has been derived to predict helium gas viscosity at high pressure and temperature. The correlation has been compared with the experimental data set up to 500 K for the temperature and up to 100 MPa for the pressure. The correlation based on the Chapman-Enskog solution and the excess viscosity relation, therefore it is simple and convenient for practical use. The force constants use in the present correlation for the Chapman-Enskog solution are \(\sigma=0.279\) and \(\varepsilon/\kappa=6.03\), respectively. The excess viscosity in the present correlation use Arp & McCarty’s correlation with a few modification. The present correlation gives a difference about 2-3 % respect to the Arp & McCarty correlation.
Nomenclature

\( \mu \) = Viscosity at 1 atm

\( T^* \) = Reduced temperature (=\( T/(\varepsilon/k) \))

\( \varepsilon \) = A maximum energy of attraction

\( K \) = the Boltzmann constant

\( P \) = Density,

\( \Sigma \) = Diameter of the molecules

\( \Omega \) = force constants

\( N \) = Number-density

\( m \) = Mass of the molecules

\( X \) = Numerical factor used in Enskoq’s theory related to the collision probability

\( a \) = Temperature independent function of the molecular forces.

\( R \) = The gas constant

\( M \) = The molecular weight.

\( T \) = Absolute temperature

\( \psi \) = The collisional transfer of molecular property

\( C \) = Velocity

\( P \) = Pressure

\( \rho_0 \) = Hydrostatic pressure

\( q \) = Total flux-vector of energy

\( \rho_0 \) = Density of the gas at normal pressure and temperature

\( c_v \) = Specific heat per unit mass at constant volume

\( f \) = Maxwell’s velocity distribution function

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