高温高压水素の粘性係数推算式の提案

Prediction of Hydrogen Gas Viscosity at High Pressure and High Temperature

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本報では高温高压状態の水素の粘性係数を簡便かつ正確に求めるための実用的な推算式を提案した。従来、低密度気体に適用できる Chapman-Enskog 理論をベースにした式に、高密度補正を加える方法が提案されているが、本推算式は、低密度領域で Chapman-Enskog 式における分子間力定数を、高密度領域で Diller の密度補正式の定数を最適化することにより得られている。既存の実測値と比較した結果、本推算式は高温・高圧力下の水素の粘性係数を偏差 2~4% 以内で予測出来ることが確かめられた。本推算式の適用範囲は、圧力 0.1MPa で温度範囲 40~2130K、および 0.1~220MPa で、100~990K である。

キーワード: 粘性係数, 高压水素ガス, Chapman-Enskog 理論, 推算式

Accurate knowledge of hydrogen gas viscosity is extremely important for developing a hydrogen economy. There is a need for recommendations relating to the best available viscosity data and the best available correlations for practical purposes. The purpose of this work is to analyze the various possibilities for viscosity correlation to estimate hydrogen gas viscosity in the high pressure and high temperature region for practical industrial uses. We recommend suitable force constants and a collision integral for the Chapman-Enskog solution to estimate viscosity in the limit of zero density. At high density, modification of Diller's extrapolation equation for excess viscosity gives good agreement with the available experimental data. A combination of the Chapman-Enskog solution and modification of Diller's excess viscosity gives an estimation of hydrogen gas viscosity within 2 to 4 % deviation from the existing experimental data for the high-temperature and high-pressure region.

[Keywords: Chapman-Enskog, correlation, gas, high pressure, high temperature, hydrogen, viscosity]

1. Introduction

Recently, interest in hydrogen gas has been increasing rapidly because of its potential to contribute towards resolving issues related to energy resourcing, energy security, global warming, and sustainable energy generation. One of the generation-IV reactors nuclear projects (HTGRs, HTR, VHTR) is designed to be coupled with a hydrogen producing power plant to produce pure hydrogen at high temperature in the process [1]. Presently, hydrogen is industrially produced from hydrocarbons such as methane and is used widely in ammonia production. However, it is preferable to produce hydrogen from water using the

process of electrolysis where the electricity is from renewable sources. Hydrogen is an environmentally clean energy carrier for the user, and is particularly attractive for transportation applications because the release of pollutants is very low at the point of end use. Hydrogen is the most abundant of the chemical elements and accurate knowledge of the thermophysical properties of hydrogen gas is extremely important for developing a hydrogen society. The viscosity of hydrogen is very important and it is a basic property required for developing any device in which hydrogen gas flows. The purpose of this work is to analyze the various possibilities for viscosity correlation (semi-theoretical or empirical) to estimate hydrogen gas viscosity in the high temperature and high pressure region and then a recommendation for the most suitable correlation is proposed.

2. Existing Experimental Data

There are several experimental techniques available to obtain hydrogen gas viscosity data. Table 1 shows the selected experimental data used to compare with

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the present correlation. We selected only the published data sets which include high temperature and high pressure because we want to make a correlation particularly in this region. Unfortunately, the existing wide-range correlations at high temperature and high pressure have accuracies poorer than 5 %. The temperatures are from 40 to 2128 K and the pressures are from 0.0009 to 217 MPa. The highest pressure data was collected by Rudenko et al. [2]. The highest temperature data was collected by Guevara et al. [3]. The most recent data was collected by May et al. [4] (213 to 394 K, 0.1 MPa).

3. Existing Correlations

3.1 Low-Density Region

To estimate hydrogen gas viscosity, we need information about two regions of density i.e., the low density region and the high density region. At atmospheric pressure, hydrogen gas viscosity may be considered to be at the limit of zero density, $\eta_0(T)$, which can be well represented by the Chapman-Enskog solution as a first approximation [5].

$$\eta_0(T) = \frac{5}{16} \frac{\sqrt{\pi m k T}}{\pi \sigma^2 \Omega_\eta(T^*)} \quad (1)$$

where, m is the mass of one hydrogen molecule ($m = M/N_{\text{avo}} = 2.01594 \times 10^{-3} / 6.022 \times 10^{23}$ kg) and T is the temperature. This relation requires three parameters, i.e., collision diameter, σ , and collision integral, Ω_η , as a function of reduced temperature, $T^* = T/(\varepsilon/k)$. ε is a maximum energy of attraction and k is the Boltzmann constant ($k = 1.3806504 \times 10^{-23}$ J·K⁻¹). Table 2 shows various force constants and collision integrals for hydrogen gas. Reported values range in Table 2 is from $\sigma = 0.2695$ to 0.3337 nm and $\varepsilon/k = 29.2$ to 79.4 K. This range of values is the result of using different collision integrals and different data sets for hydrogen viscosity. Besides the Chapman-Enskog solution, other empirical correlations at low-density for hydrogen gas viscosity are also studied. We tabulated the correlations for the low-density region in Table 3.

3.2 High-Density Region

Correlations for hydrogen gas viscosity applied to high density are not sufficiently accurate at present. Therefore, we are interested in developing a new correlation for practical use. For industrial purposes, it is desirable to obtain a simple correlation with a high accuracy. To estimate hydrogen gas viscosity at high-density, the common approach is to add an extra term $\Delta\eta(T, \rho)$, known as the excess viscosity, to correct for the effect of the gas density as given in Eq. (2).

$$\eta(T, \rho) = \eta_0(T) + \Delta\eta(T, \rho) \quad (2)$$

For some gases, experimental data has been correlated successfully assuming that the excess viscosity is a

function of the density only (e.g. [6]). For lighter molecules such as hydrogen, however the temperature dependence of the excess viscosity is important. Diller [7] and McCarty et al. [8] use a temperature dependent excess viscosity relation of the form given in Eq. (3) for estimation hydrogen gas viscosity at high density.

$$\Delta\eta(T, \rho) = A(\rho) \exp[B(\rho)/T] \quad (3)$$

Diller mentioned in his paper that the assumption of the excess viscosity being independent of temperature is not adequate especially at densities greater than twice of the critical point value where the temperature dependence is much greater. However, Diller and McCarty correlations are limited to the low temperature region and the pressure up to 35 MPa.

As another alternative to the excess viscosity relation, the density dependence of viscosity can also be estimated using Enskog theory (Eq.4).

$$\eta(T, \rho) = b\rho\eta_0(T) [1 / (b\rho\chi) + 0.800 + 0.7614b\rho\chi] \quad (4)$$

b and χ can be derived either empirically or theoretically. Michels et al. [9] and Hanley et al. [10] compare Enskog theory with the experimental hydrogen data. Because Enskog theory is not valid in extreme density regions, Hanley modified the Enskog theory and named it Modified Enskog Theory (MET). However, Michels and Hanley correlations show deviations from the experimental data around 10 % at high density. We tabulated the correlations for the high-density region in Table 4.

4. New Correlation

4.1 Low-Density Region

At low-density we use the Chapman-Enskog solution. The Chapman-Enskog solution is a very simple equation as shown by Eq.1. However, the applicability of the solution strongly depends on the selected parameters for the collision integral and intermolecular force constants. The collision integral, Ω_η , empirically developed by Assael et al. [11] for the hydrogen gas case was used in the present correlation, Eq. 5. Assael used his primary data selected to evaluate the function Ω_η , over the temperature 20 - 2200 K at pressure 0.1 MPa. The force constants $\sigma = 0.2968$ nm and $\varepsilon/k = 33.3$ K were chosen. These force constants were derived from hydrogen viscosity experiments and classical formulae which are useful for rough calculations [5]. For the present correlation, we recommend a combination of force constants of 0.296 nm and 35 K. These force constants were chosen to fit with the existing available data set in Table 1.

$$\begin{aligned} \Omega_\eta(T^*) = & \exp(0.354125 - 0.427581(\ln T^*)) \\ & + 0.149251 (\ln T^*)^2 - 0.037174 (\ln T)^3 \\ & + 0.003176 (\ln T^*)^4 \end{aligned} \quad (5)$$

Fig. 1 Collision integral versus reduced temperature

In the present analysis, hydrogen gas density was calculated from an equation of state derived by Leachman [16]. To determine Ω_η for the case of hydrogen, the Lennard-Jones potential is commonly used as an intermolecular potential energy function. However, the Lennard-Jones potential is an approximation [5]. Therefore, in the present correlation we use a collision integral developed by Assael et al. based directly on hydrogen viscosity data. Figure 1 shows the comparison of the collision integral for hydrogen and other gases versus reduced temperature.

For the Lennard-Jones potential, the collision integral data is conveniently available as an empirical equation developed by Neufeld et al. [17]. The hydrogen collision integral developed by Assael has a different trend to the other curves in that the rate of decrease with respect to the reduced temperature is faster at larger values of T^* . The collision integral for hydrogen is lower than the universal gas collision integrals, especially at high temperature. The other

collision integrals deviate from Assael's by as much as 13 % for $T^* > 30$ [18, 19].

Figure 2 shows the percent deviation in viscosity of dilute gas correlations tabulated in Table 3 from the present correlation. Mason et al. [20], Hanley et al. [21], Assael et al. and the present correlation used the Chapman-Enskog solution as a base equation at low-density. In spite of the different force constants, collision integral and experiment data selected, the discrepancy between these correlations and the present correlation is as much as 1% (for c and d) and up to 2% (for a and b). Assael's correlation gives a better agreement with the data of Guevara et al. [3] than the present correlation. However, it deviates more than 2% from the data of Trautz et al. [22] around 1000 K. Figure 2 shows that the force constants used in the present correlation give a better agreement in average with the existing experimental data than those used by Assael. Standard deviation of the existing data set in Fig.2 from the present correlation is as much as 0.10 $\mu\text{Pa}\cdot\text{s}$ and a little larger deviation 0.11 $\mu\text{Pa}\cdot\text{s}$ is obtained from Assael's correlation. Maitland's correlation [23] is a pure empirical relation for prediction of hydrogen viscosity at low-density. It also agrees with the present correlation within 1 %.

4.2 High-Density Region

At high density, we use an excess viscosity relation Eq. 3 rather than Enskog theory Eq. 4. The present excess viscosity expression is modification of the Diller's expression shown in Table 4 by changing the constant values in $A(\rho)$ and $B(\rho)$ as

$$A(\rho) = \exp(5.73 + \ln(\rho) + 65.0 \rho^{3/2} - 6.00 \times 10^{-6} \exp(135 \rho))$$

$$B(\rho) = T_0 \left\{ 10.0 + 8 \left[\left(\frac{\rho}{0.07} \right)^6 - \left(\frac{\rho}{0.07} \right)^3 \right] - 18 \exp \left[-59 \left(\frac{\rho}{0.07} \right)^3 \right] \right\} \quad (6)$$

Fig. 2 Hydrogen viscosity deviation vs temperature at low density. (a, b, c, d, correspond to correlations in Table 3)

Figure 3 shows the effect of pressure on hydrogen gas viscosity at 25°C for existing correlations and the present correlation. This isotherm was selected since there are a number of experimental data by various authors at high pressure. The viscosity correlation derived by Diller is slightly higher than the experimental data at high pressure. However, the good agreement with the experimental data is surprising since Diller's expression was developed for data at low temperature below 100 K. Therefore we conclude that we still can use Diller's expression for excess viscosity with a slight modification in the higher temperature and higher density region. The applicable ranges of temperature and pressure of the present correlation are 40 to 2130 K for dilute gas and 100 to 990 K for 0.1 to 220 MPa, respectively. The present correlation, as can be seen in the Fig.3, gives a good agreement with respect to the existing experimental data. The Enskog theory of Michels's with parameters χ and b ,

Fig.3 Viscosity comparison at 25 °C (e, f, g, and h correspond to the correlations in Table 4)

REFPROP [24] and Diller's correlation give reasonably good agreement with the present correlation up to 30 MPa. The Modified Enskog Theory (MET) derived by Hanley's and McCarty's correlation give lower estimations for viscosity at high density above 50 MPa.

Figure 4 shows a comparison of the present excess viscosity correlations with respect to Diller's and McCarty's correlations. At low density they give a good agreement up to 23 kg·m⁻³. As for McCarty's correlation, increasing rate becomes low at 23 kg·m⁻³ and it reaches a maximum at around 65 kg·m⁻³. On the other hand, Diller's correlation increases more steeply than the present at higher densities.

Figure 5 shows the percent deviation in hydrogen viscosity experimental versus pressure. Diller's correlation (Table 4, f) at 100 MPa gives a larger discrepancy at high temperature region. Figure 6 shows the percent deviation in hydrogen viscosity experimental versus temperature. Tsederberg et al. measured (Ref. [23]) hydrogen gas at around 900 K at 50 MPa. He estimated the accuracy of his data set is 3 %. Totally, the discrepancy of the present correlation with respect to the existing hydrogen gas viscosity experimental data at high pressure and high temperature agree within 2 to 4 % (Figs.5 and 6).

5. Conclusion

A new correlation has been derived to estimate hydrogen gas viscosity at high pressure and high temperature. The correlation is very simple and convenient for practical use. It is based on the Chapman-Enskog solution and the excess viscosity relation. The discrepancy of the present correlation is as much as 2 to 4% with published data selected from the high-temperature and high-pressure region. Temperature and pressure ranges of the present correlation are 40 to 2130 K for dilute gas, and 100 to 990 K for 0.1 to 220 MPa, respectively.

Fig.4 Excess viscosity simulation (Eq. (3))

Fig. 5 Viscosity deviation vs. pressure

Fig.6 Viscosity deviation vs. pressure

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Table 1 Selected Experimental Data Set

References	Uncertainty, %	T/K	P/MPa	References	Uncertainty, %	T/K	P/MPa
May et al.	0.08	213.62-394.21	0.1	Kestin et al.*[30,31]	0.05-0.2	293.15-303.15	0.1-2.3
Maltsev et al.[25]	3	500,800,1100	0.1-1	Rudenko et al.	4-6	300	0.1-217
Nabizadeh et al.[26]	0.5-1	295.55-399.15	0.1-5.8	Tsederberg et al.[32]	3	288.65-989.65	4.4-50.92
Clifford et al.*	0.2	198.15-208.15	0.1	Barua et al.*[33]	0.2	223.15-423.15	1.01-20.26
Hongo et al. [27]	0.3	298.15-373.15	0.1-12.5	Golubev et al.[34]	ns	288.15-523.15	0.1-81.04
Chuang et al. [28]	0.5	173.15-273.15	0.4-50.6	Michels et al.*	ns	298.15-398.15	2.1-192.4
Guevara et al.*	0.4	1102-2128	0.1	Kuss [35]	2	298.15-348.15	0.1-50.66
Gracki et al.* [29]	0.1-0.2	173.15-298.15	15.2-25.3	Trautz et al.*[36-43]	ns	195.15-1098	0.1
Menabde et al.*	2	77-299	0.1	Rietveld et al.*[44]	ns	71-293	0.0009-0.005
Coreman et al.*	2-3	40-80	0.1	Itterbeek&Paemel*[45]	ns	77-293	0.1
Diller	0.5	100	0.6-34.5	Johnston&McCloskey [46]	<0.3	90-300	0.1

ns: not specified, * Reference data used by Assael et al.

Table 2 Force Constants for Hydrogen Gas

References	Force Constants		Ω_η	References	Force Constants		Ω_η
	σ/nm	$\varepsilon/k/\text{K}$			σ/nm	$\varepsilon/k/\text{K}$	
Nabizadeh et al.	0.2695	79.4	Empirical[19]	Kestin & Nagashima	0.2892	46.2	L-J
Hanley et al.	0.2970	39.2	L-J [47]	Hirschfelder et al.	0.2968	33.3	L-J (Cl)
Hirschfelder et al.	0.2915	38.0	L-J (Cl)	De Boer[50]	0.2928	37.0	L-J (Qu)
Hirschfelder et al.	0.2870	29.2	L-J (Cl)	Mason and Rice	0.3287	37.0	L-J [51]
Michels et al. [48]	0.2958	36.7	L-J (Qu)[49]	Mason and Rice	0.3337	37.3	Exp-Six [52]

L-J:Lennard-Jones potential; Cl:Classical formulae; Qu:Quantum formulae

Table 3 Correlations at Low Density

No	References	Equations/method	Experiments
a	Mason and Rice n-hydrogen	$\eta_o(T) = \frac{266.93 f_\eta \sqrt{MT}}{\sigma^2 \Omega_\eta}$; $\sigma=0.3337 \text{ nm}$; $\varepsilon/k=37.3 \text{ K}$ M is molecular weight; f_η and Ω_η is dimensionless and tabulated in his paper; Ω_η calculated by the Exp-6 potential developed by Mason $T_{\text{range}}=200\text{-}1098 \text{ K}$	Johnston & McCloskey; Trautz et al.; Wobser & Muller [53]
b	Hanley et al. p-Hydrogen and n-Hydrogen	$\eta(T) = \eta_o \left\{ 1 + \frac{3}{196} \left[8 \frac{Q^{(2,3)}}{\Omega_\eta} - 7 \right]^2 \right\}$ $\eta_o(T) = \frac{5}{16} \frac{\sqrt{\pi m k T}}{\pi \sigma^2 \Omega_\eta}$; $(T_{\text{low}})\sigma=0.297 \text{ nm}$; $\varepsilon/k=39.2 \text{ K}$; $(T_{\text{high}})\sigma=0.306 \text{ nm}$; $\varepsilon/k=30.4 \text{ K}$; Ω_η calculated by the 9-6 potential; $T_{\text{range}}=15\text{-}5000 \text{ K}$	Guevara et al.; Barua et al.; Van Itterbeek et al.; Johnston & McCloskey; Wobser & Muller; Rietveld et al.; Kestin et al.; Gracki et al.; Sutherland & Maass [54]
c	Maitland et al. n-Hydrogen	$\ln(\eta_o/88.0[\mu P]) = 0.68720 \ln T - 0.61732/T - 111.49/T^2 - 3.9001$ $T_{\text{range}}=20\text{-}2200 \text{ K}$	Johnston & McCloskey; Kestin & Leidenfrost; Gracki et al.; Guevara et al.; Barua et al.; Coreman et al.; Yen [55]
d	Assael et al. n-Hydrogen	$\eta_o(T) = \frac{5}{16} \frac{\sqrt{\pi m k T}}{\pi \sigma^2 \Omega_\eta(T^*)}$ $\sigma=0.2968 \text{ nm}$; $\varepsilon/k=33.3 \text{ K}$; Ω_η calculated empirically from hydrogen viscosity experiment; $T_{\text{range}}=20\text{-}2200 \text{ K}$	Asterisk (*) symbol in Table 1

Table 4 Correlations at High Density and the Present Recommended Correlation

No	References	Equations/method	Experiments
e	Michels et al.	Enskog-theory $\eta(T, \rho) = \eta_o(T) b \rho [1/b \rho \chi + 0.800 + 0.7614 b \rho \chi]$ $\eta_o = \eta(\text{at } 0.1 \text{ MPa})$, $b = 2\pi\sigma^3/3m$ m is mass of hydrogen; $b \rho \chi$ is calculated from the PVT data	Michels et al. Ranges: T:298-398K P:0.1-200 MPa
f	Diller	Excess viscosity relation $\eta(T, \rho) = \eta_o(T) + \Delta\eta(T, \rho)$ $\eta_o = 8.5558[T^{3/2}/T + 19.55][(T + 650.39)/(T + 1175.9)] \times 10^{-6}$ $\Delta\eta(T, \rho) = A(\rho) \exp[B(\rho)/T]$ $A(\rho) = \exp\{5.7694 + \log_e(\rho) + 65.0\rho^{3/2} - 6.00 \times 10^{-6} \exp(127.2\rho)\}$ $B(\rho) = T_o \left\{ 10.0 + 7.2 \left[\left(\frac{\rho}{0.07} \right)^6 - \left(\frac{\rho}{0.07} \right)^{\frac{3}{2}} \right] - 17.63 \exp \left[-58.75 \left(\frac{\rho}{0.07} \right)^3 \right] \right\}$ note that $T_o = 1.0 \text{ K}$ and ρ is in $\text{g}\cdot\text{cm}^{-3}$.	Diller Ranges: T:14-100 K P:0.1-35 Mpa
g	Mc-Carty et al.	Excess viscosity relation $A(\rho) = \frac{(306.4636\rho - 3350.628\rho^2 + 3868.092\rho^3)}{(1.0 - 18.47830\rho + 110.915\rho^2 + 25.3524\rho^3)} \times 10^{-6}$ $B(\rho) = 10.0 + 7.2 \left[\left(\frac{\rho}{0.07} \right)^6 - \left(\frac{\rho}{0.07} \right)^{\frac{3}{2}} \right] - 17.63 \exp \left[-58.75 \left(\frac{\rho}{0.07} \right)^3 \right]$ McCarty modified Diller's correlation on excess viscosity at high density.	Diller Ranges: T<100 K P:0.1-35 Mpa
h	Hanley et al.	Modified Enskog Theory (MET) $\eta(T, \rho) = \eta_o(T) + \eta_1 \rho + \dots$ $\eta_o(T) = \frac{5}{16} \frac{\sqrt{\pi m k T}}{\pi \sigma^2 \Omega_\eta(T^*)}$; $\eta_1 = (-0.601(\pm 0.008) + 0.8)b \eta_o$ $b = 2\pi\sigma^3/3m$; $\sigma = 0.2970 \text{ nm}$; $\varepsilon/k = 39.2 \text{ K}$; Ω_η calculated by the Lennard-Jones potential derived by Munn et al.	Diller; Barua et al.; Michels et al. Ranges: T>100 K
i	Present correlation	Excess viscosity relation $\eta(T, \rho) = \eta_o(T) + \Delta\eta(T, \rho)$ $\eta_o(T) = \frac{5}{16} \frac{\sqrt{\pi m k T}}{\pi \sigma^2 \Omega_\eta(T^*)}$ $\Omega_\eta(T^*) = \exp(0.354125 - 0.427581(\ln T^*) + 0.149251(\ln T^*)^2 - 0.037174(\ln T^*)^3 + 0.003176(\ln T^*)^4)$ $\sigma = 0.296 \text{ nm}$; $\varepsilon/k = 35 \text{ K}$; $\Delta\eta(T, \rho) = A(\rho) \exp[B(\rho)/T]$ $A(\rho) = \exp\{5.73 + \log_e(\rho) + 65.0\rho^{3/2} - 6.00 \times 10^{-6} \exp(135\rho)\}$ $B(\rho) = T_o \left\{ 10.0 + 8 \left[\left(\frac{\rho}{0.07} \right)^6 - \left(\frac{\rho}{0.07} \right)^{\frac{3}{2}} \right] - 18 \exp \left[-59 \left(\frac{\rho}{0.07} \right)^3 \right] \right\}$	Table 1 Ranges: T:40 to 2130K for dilute gas and T:100 to 990 K for 0.1 to 220 Mpa

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