Hydrogen Transport Property Correlations

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IMPROVED and more extensive experimental data and more detailed calculations of hydrogen transport properties warrant the generation of new correlations. Particularly useful correlations in the form of equations readily adaptable to machine computations have been developed for both thermal conductivity and viscosity from cryogenic temperatures to 5000° K. and essentially 1-atm. pressure to whatever upper pressure has been recorded in the literature. Several forms of equations have been used to represent the data with good accuracy and to account for the peculiar characteristics of the data in different temperature ranges.

The forms of the correlating equations developed have been determined primarily for their ability to represent the data and not because they have theoretical significance. The assumption has been made throughout that the data could be represented by some function of the form $y = f(\bar{x}, \bar{a})$ where $\bar{x}$ is a vector denoting the set of independent variables, $\bar{a}$ is a vector denoting the parameters of the function, and y is the dependent variable. The method of least squares was used to obtain values for the parameters of the function. Also the value

$$s_f = \left\{ \frac{\sum_{i=1}^{n} [y - f(\bar{x}, \bar{a})]^2}{n - k} \right\}^{1/2}$$

was computed where n is the number of data points and k is the number of parameters. This number will be referred to as the standard deviation of the least squares equation. Since several functions were tried for each set of data, the function reported is the one that gave the smallest s_f. A more detailed discussion of the least squares method used may be found (*48*). Graphical correlations of hydrogen data over more limited ranges of conditions have been undertaken by Schaeffer and Thodos (*56*) and by Codegone (*10*). The former point out the limitations of applying generalized correlations developed by others to the transport properties of hydrogen.

To include the effect of pressure the present work develops empirical equations in the form of the residual transport property as a function of density after the theoretical approach of Predvoditelev (*52*) and Abas-zade (*1*). Other correlating equations patterned after Enskog (*13, 23*) and Keyes (*41*) did not seem to be particularly convenient or advantageous to use in the manner of this development.

The literature was very throughly evaluated. Only original tabulated numerical results have been included. This accounts for omission, in a few cases, of references and data where either graphically presented results of low resolution have been given, or reproduced results of prior investigations have been repeated by authors as a basis for calibration of experimental equipment. Arbitrary criteria for removing inconsistent data for the final correlations were based on trends observed in prior sequential machine computations. These are discussed below and limitations of the equations are presented.

THERMAL CONDUCTIVITY

Normal Gaseous Hydrogen. Thermal conductivity of normal gaseous hydrogen at 1-atm. pressure was correlated with Equation 1, applicable from 0° to 2080° K.

$$k_n^* = [1.8052\,T - 1.4829\,(10)^{-3}\,T^2 + 1.2481\,(10)^{-6}\,T^3 - 4.195\,(10)^{-10}\,T^4 + 4.6302\,(10)^{-14}\,T^5]\,10^{-6} \qquad (1)$$

From 25 sources 115 data points were used (*2, 3, 6, 12, 14, 18, 20, 22, 24, 25, 33, 36, 42, 45, 49, 55–58, 59, 73–75, 78*). More were originally available but all those above 65° K. whose absolute deviation from the first fitted equation was more than 5% were discarded. Some lower temperature data points were also dropped as being considerably out of line. The final representation included 13 data points below 66° K. but gave deviations of −7.3 to 10.4%. Twelve points had positive deviations; the low temperature data of Ubbink and others (*73, 74*) are somewhat high. The data of Wassiljewa (*77*) did not meet the 5% criteria. The data of Gregory and Archer (*19*) as corrected by Hercus and Laby (*24*) were used. Fifty-seven data points had absolute deviations less than 2%, 79 less than 3%, and only one point above 66° K., at 295.1° K., had an absolute deviation from the final fitted equation greater than 5%. The standard deviation for the equation is 11.

Normal Compared to Parahydrogen. Computed ratios of thermal conductivities of para to normal gaseous hydrogen are given by Johnson (*32*) for 10° to 300° K. These were extended to 550° K. using the equation of Farkas (*15*)

$$\frac{k_p}{k_n} = \frac{C_{vp} + 2.25\ R}{C_{vn} + 2.25\ R} \qquad (2)$$

Twenty-four points at 1-atm. pressure were fitted with a four-constant, skewed normal equation with a standard deviation of 0.0074 and a maximum absolute deviation for any single point being 0.013 at 120° K. The ratio is of the order of 1. The equation is

$$k_p/k_n - 1 = 0.19405 \exp[-1.4371\,(10)^{-4}\,(T - 169.78)^2]$$
$$[1 - 8.0139\,(10)^{-3}\,(T - 169.78) + 1.6243\,(10)^{-6}\,(T - 169.78)^3] \qquad (3)$$

The thermal conductivity of normal and parahydrogen is the same in the liquid phase (*51*).

Dense Phase. The theory of Predvoditelev (*52*) and Abas-zade (*1*) indicates that the pressure, or more properly density, variation of thermal conductivity of hydrogen should follow the form

$$k(\rho, T) = k(T) + a[\rho(P, T)]^b \qquad (4)$$

where a and b are constants. A least squares fit of the data (*42, 45, 51*) for thermal conductivity *vs.* density was made with this form of equation, and the result was slightly above the data for gaseous hydrogen with density less than 0.01 gram per cc. and slightly below the data from 0.01 to 0.015 gram per cc. Polynomial equations in both cartesian and log-log coordinate systems represented the thermal conductivity *vs.* gaseous density data rather well with almost identical results but did not conform reasonably to the form of Equation 4 in the region of the density data gap. No data exist over the gap from $\rho = 0.015$ to 0.065 gram per cc., the latter being for liquid. This covers the dense phase region of the vapor-liquid dome. The data that exist are badly scattered. The final empirical equation was a polynomial where smoothed data were made to agree with the polynomial equation in cartesian coordinates through the low density data and to the exponential form in the region of $\rho = 0.03$ to 0.074 gram per cc. with a smooth transition between these regions. The equation is

$$k_n - k_n^* = [1.84 + 1102.6\,\rho + 1.22648\,(10)^6\rho^2 - 1.15024\,(10)^8\rho^3 + 4.95228\,(10)^9\rho^4 - 1.16927\,(10)^{11}\rho^5 + 1.5678\,(10)^{12}\rho^6 - 1.12433\,(10)^{13}\rho^7 + 3.36150\,(10)^{13}\rho^8]\,10^{-6} \qquad (5)$$

The explicit temperature dependence of k_n is contained in k_n^* (Equation 1). The standard deviation of the least squares equation to the smoothed data is 1.18. The equation is applicable to both gas and liquid from $\rho = 0$ to $\rho = 0.074$ gram per cc. The high density data of Stolyarov and others (*59*) were not used, being internally inconsistent. Values for density were taken from Mathias, Crommelin, and Onnes (46) as corrected (*80*); Johnston, Keller, and Friedman (*34*); and Woolley, Scott, and Brickwedde (*80*). The plot of the equation and the data are shown in Figure 1.

Dissociation. No experimental data exist for the transport properties of hydrogen in the dissociating region. Most recent theoretical values have been computed by Clifton (*9*) and are used as a basis for empirical correlation. These data have been fitted with a combined normal and straight-line equation for each of several pressures as a function of temperature. Each equation was determined by 11 computed points from 1500° to 5000° K. The equation is

$$k = \left\{\frac{A_1}{A_2\,(2\pi)^{1/2}}\exp\left[-\tfrac{1}{2}\left(\frac{T-A_3}{A_2}\right)^2\right] + A_4 + A_5\,T\right\}10^{-7} \qquad (6)$$

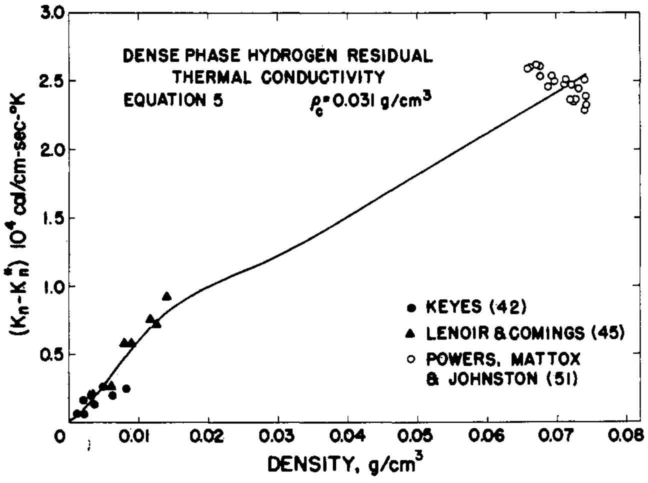

Figure 1. Residual thermal conductivity vs. density

Table I gives the values of the A's for several pressures, the maximum and median percentage deviations (absolute values), and the standard deviation of the fitted equation. Correction terms of −1974 and 3.690 have been added to A_4 and A_5 to adjust the equations correlating theoretical points of Clifton (*9*) to experimental data fit by Equation 1. This was necessary because the absolute values and the slope for the undissociated gas-computed data did not closely approximate experimental results (*6*) in the region of 1500° to 2080° K. An interpolation scheme must be devised for obtaining the thermal conductivity at other pressures. The table clearly shows the improved fit with higher pressures.

VISCOSITY

Normal Gaseous Hydrogen. Correlation to 1 atm.-experimental data (*4, 7, 8, 11, 22, 26-28, 30, 35, 40, 43, 44, 47, 50, 53, 54, 60-67, 69, 70, 72, 76, 79, 81*) for gaseous hydrogen from 13.97° to 1280° K. is as follows:

$$\mu^* = [2.105 + 0.4774\,T - 9.9784\,(10)^{-4}\,T^2 + 1.61827\,(10)^{-6}\,T^3 - 1.28024\,(10)^{-9}\,T^4 + 3.8164\,(10)^{-13}\,T^5]\,10^{-6} \qquad (7)$$

Data of Trautz and Narath (*68*) and Trautz and Weizel (*71*) were inadvertently omitted but would not noticeably alter the result. Two hundred and two data points were used. The two uppermost in temperature data points used were from Guevara and Wageman (*21*). Data for this correlation resulted from a previous fit of 230 data points from which data below 21.6° K. not within 10% and above 21.6° K. not within 5% of the correlation had been eliminated. All the data of Keesom and MacWood (*38*) were eliminated; all values except one were outside the above criteria. The standard deviation of the equation is 1.1.

Normal Compared to Parahydrogen. The work of Becker and Stehl (*5*) shows no significant contribution to the viscosity as a function of ortho-para concentration.

Dense Phase. The density variation of viscosity was correlated as the excess viscosity over that for 1-atm. pressure for both the gas and liquid. Data from eight sources were used (*16, 17, 29, 31, 37, 39, 44, 45*). Because of the double valued property of the residual viscosity in the neighborhood of 0.071 gram per ml. due to the difference in the liquid and gaseous phases, three empirical equations were evolved. Figure 2 graphically depicts Equations 8, 9, and 10 and gives the retained data points. Numbers associated with individual points in the figure represent weighting as the graphical resolution is too low to reveal several closely coinciding values. The basic assumption was made that the critical point condition of $\rho = 0.031$ gram per cc. and $\mu - \mu^* = 13.0\,(10)^{-6}$ g./cm.-sec. for the gaseous equation applied to the liquid phase. The equation used for the liquid data was extrapolated monotonically downwards from 0.070 gram per cc. through

Table I. Constants of Equation 6 and Deviations

Pressure, Atm.	A_1	A_2	A_3	A_4	A_5	Max. Dev., %	Med. Dev., %	Std. Dev., %
0.1	$2.9499(10)^8$	434.85	3233	1205	11.517	7.0	3.6	0.06
0.5	$3.0177(10)^8$	500.57	3556	−1609	13.250	3.2	2.0	0.03
1.0	$3.0109(10)^8$	532.07	3716	−3623	14.473	2.8	1.1	0.02
2.0	$3.1315(10)^8$	587.36	3902	−2409	13.637	2.5	1.1	0.02
10.0	$3.4259(10)^8$	732.61	4386	795	11.580	0.9	0.6	0.01
50.0	$3.7276(10)^8$	932.30	5001	2156	10.663	0.8	0.3	0.005
100.0	$3.6789(10)^8$	1013.91	5268	2143	10.672	0.9	0.4	0.006

the critical point condition with no rigid restriction applied to the slope. The difficulty of extrapolating over the region where there are no data is apparent from the slight undulating character of Equation 9 as shown in the figure. The dearth of viscosity data in the region of the vapor-liquid dome is comparable to that for thermal conductivity.

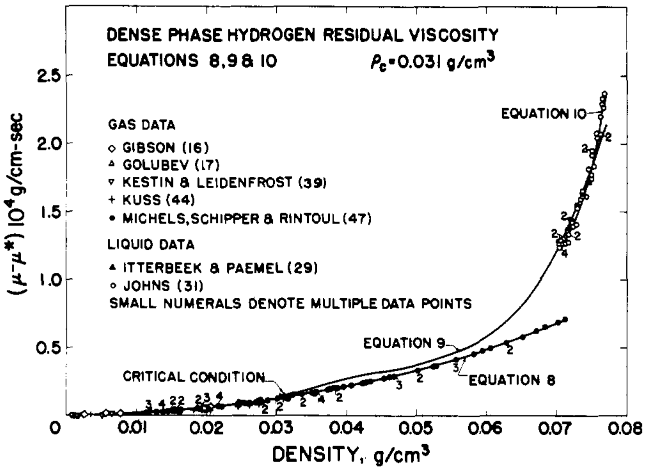

Figure 2. Residual viscosity vs. density

The equation for the gaseous phase for densities from near zero to 0.072 gram per cc. is

$$\mu - \mu^* = [0.27 + 46.6\rho + 13{,}450\rho^2 - 79{,}900\rho^3 + 1.10\,(10)^6\rho^4]\,10^{-6} \qquad (8)$$

Of 159 data points, 106 were retained and correlated with a maximum absolute deviation of 12%. Of these, 74 points deviated less than 5% from the least squares equation. The standard deviation of the equation is 0.054. Since the value of Equation 8 represents a correction term for pressure to μ^* as obtained from Equation 7, over-all percentage effect of this correlation to the final value of μ is considerably lessened. Further, the equation fits the data best for the higher $\mu - \mu^*$ values with no deviations in excess of 5% for densities greater than 0.0351 gram per cc. Also, most of the larger deviations correspond to low $\mu - \mu^*$ values which represent small differences in like numbers with all the inherent amplifications of experimental uncertainties. The equation is applicable to the gas phase from $\rho = 0.00093$ to 0.07109 gram per cc.

The liquid phase equation from $\rho = 0.031$ to $\rho = 0.077$ gram per cc. is

$$\mu - \mu^* = [-181.85 + 0.60966 \exp(75.2\,\rho) + 10899\,\rho - 164480\,\rho^2 + 9.467\,(10)^6\rho^4]\,10^{-6} \qquad (9)$$

The explicit temperature dependence is included in μ^* from Equation 7. An initial 66 data points from the literature (*29, 31, 37*) were first correlated by a polynomial not extrapolated through the critical conditions This equation is

$$\mu - \mu^* = [-2302.1 + 2.2430\,(10)^5\rho - 4.96060\,(10)^6\rho^2 + 3.21592\,(10)^7\rho^3]\,10^{-6} \qquad (10)$$

which fits all the liquid data except one value within 7.8% and is applicable from $\rho = 0.070$ to $\rho = 0.077$ gram per cc. All data points (16) not within 5% of this latter equation were eliminated from the input which led to Equation 9. This former equation approximates 40 of the 50 remaining data points within 5% and all of the 50 data points within 11%. The standard deviation of the equation is 0.055. Equation 9 is useful for estimating the viscosity for the liquid phase in the region where no data exist—i.e., $\rho = 0.031$ to $\rho = 0.070$ gram per cc.—whereas Equation 10 is a better representation of the existing liquid data. Almost all the uncertainty in viscosity for the liquid phase as represented herein will be attributable to either Equations 9 or 10 as $\mu - \mu^*$ is usually large compared with μ^*.

Dissociation. An equation involving one dependent and two independent variables was used to approximate Clifton's (*9*) data. The functional form is:

$$\mu = [a_1 + a_2T + a_3P + a_4T^2 + a_5TP + a_6P^2 + a_7T^3 + a_8T^2P + a_9TP^2 + a_{10}P^3 + a_{11}T^4 + a_{12}T^3P + a_{13}T^2P^2 + a_{14}TP^3 + a_{15}P^4]\,10^{-6} \qquad (11)$$

where

$a_1 = 110.55$
$a_2 = 0.02321$
$a_3 = 9.0610$
$a_4 = 8.9787\,(10)^{-5}$
$a_5 = -8.806\,(10)^{-4}$
$a_6 = -1.156858$
$a_7 = -3.10378\,(10)^{-8}$
$a_8 = 1.07509\,(10)^{-6}$
$a_9 = -5.1082\,(10)^{-5}$
$a_{10} = 0.02970271$
$a_{11} = 3.0243\,(10)^{-12}$
$a_{12} = -1.23112\,(10)^{-10}$
$a_{13} = 1.4078\,(10)^{-9}$
$a_{14} = 2.5251\,(10)^{-7}$
$a_{15} = 1.873858\,(10)^{-4}$

This equation fits 86 of 88 theoretical values with temperatures from 1500° to 5000° K. and with pressures of 0.1, 0.2, 0.5, 1, 2, 10, 50, and 100 atm. to within 3.9%. The equation deviated from the other two data points used by 5 and 5.5%. The equation does not give good values for viscosity at all the intermediate pressures and interpolation schemes should be devised. For values of viscosity below 1500° K. down to 700° K., the pressure should be set equal to zero. The agreement with 1-atm. Equation 7 is within 3% in the region of 700° K. to 1000° K. The standard deviation of the least squares equation over the 1500° to 5000° K. range is 77.

In general, some caution should be exercised in going from one equation applicable in a given range to another equation covering a different range of conditions. In the region of overlap a double valued transport property will result despite the close agreement between the equations. In addition, whereas the data of Clifton (*9*) show no pressure dependence under some conditions, the nature of the equations is such that a small random pressure dependence is exhibited. Also, some caution should be used in taking derivatives of the equations herein which are polynomials whose slopes are not always well behaved. As an alternate, it is suggested that two values of the property be computed over a sufficient range covering the independent variables of interest and a straight line slope be taken. The derivatives of Equations 3 and 6 are well behaved.

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NOMENCLATURE

C = specific heat
k = thermal conductivity in cal./cm.-sec.-° K.
P = pressure in atm.
R = gas constant
T = ° K.

Greek Symbols

μ = viscosity in g./cm.-sec.
ρ = density in g./cc.

Subscripts

n = normal (room temperature, equilibrium ortho-para) hydrogen
p = parahydrogen
v = constant volume

Superscripts

* = 1 atm. pressure or equivalent

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