

Current Status of Transport Properties of Hydrogen

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Abstract This article contains a survey of experimental data for the thermal conductivity and viscosity of hydrogen, which are needed for many applications in system analysis and design. It includes an analysis of the current standard models for thermal conductivity and viscosity of normal hydrogen and parahydrogen, which are based on measurement and correlation work done before the mid1980s. Properties calculated with these models are analyzed and compared to all available experimental data for normal hydrogen and parahydrogen. Finally, recommendations for future work, including new experimental measurements to enable the development of improved transport property formulations for hydrogen, are provided.

Keywords Hydrogen · Model · Normal hydrogen · Parahydrogen · Thermal conductivity · Transport properties · Viscosity

1 Introduction

A relatively large body of experimental data (over 3,000 individual data points) exists for the thermal conductivity and viscosity of hydrogen, dating back to the early 1900s.

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There are several sources of recommended values or correlations in the literature, including the studies of Vargaftik et al. [1], Stephan and Lucas [2], Assael et al. [3], Touloukian et al. [4,5], and NIST [6]; bibliographic information is available as well [7]. In this article, we will use the designation “standard” to refer to the transport property formulations selected for inclusion in the National Institute of Standards and Technology (NIST) database, REFPROP [6]. REFPROP uses the formulations given in the earlier NIST12 computer program [8], which are based primarily on the work of Diller [9] for viscosity and the work of Roder [10] for thermal conductivity. These formulations are based on the IPTS-68 temperature scale, and can be used at pressures up to 121 MPa and at temperatures to 400 K. We are not aware of any recent published efforts to evaluate and analyze the available data and modeling methods for wide-range representation of the viscosity and thermal conductivity of hydrogen, and the time is appropriate to consider a re-evaluation.

Transport properties are needed for engineering applications in systems for hydrogen transportation and storage. For instance, transport properties are needed up to 1,300 K for hydrogen generation using nuclear and coal gasification processes. If the projected future utilization of hydrogen for replacement of conventional fuels in transportation and other applications happens, accurate transport properties will be needed to support system design and analysis for the global “hydrogen economy.”

2 Background

The hydrogen molecule occurs in two allotropic modifications that differ in the relative orientation of the nuclear spins of the individual atoms in the molecule. The spin orientation relative to the individual nuclei of the molecule has a significant influence on the rotational states for the molecule. Molecules with parallel spins (same direction) are called orthohydrogen, have odd rotational quantum numbers ($J = 1, 3, \dots$), and are predominant at higher temperatures. Those with anti-parallel spins (opposite directions) are called parahydrogen, have even rotational quantum numbers, and allow the lowest energy state ($J = 0$). Normal hydrogen is a mixture of about 75% orthohydrogen and 25% parahydrogen. This paper is a companion to a parallel study on the thermodynamic properties of hydrogen [11], which contains information on hydrogen behavior.

In general, the largest differences in the thermophysical properties of orthohydrogen and parahydrogen occur in the properties where the rotational heat capacity is important. The thermal conductivity, in particular, exhibits significant differences. In the temperature range from 120 to 190 K, the thermal conductivity of parahydrogen can be 30–50% larger than that of orthohydrogen [12]. Figure 1 shows the differences in thermal conductivities of parahydrogen and normal hydrogen at temperatures up to 300 K, calculated by use of REFPROP [6].

The storage and transportation of large quantities of liquid and gaseous hydrogen present unique problems for engineering designs to monitor and control the ratio of orthohydrogen to parahydrogen in commercial systems. The energy of conversion of

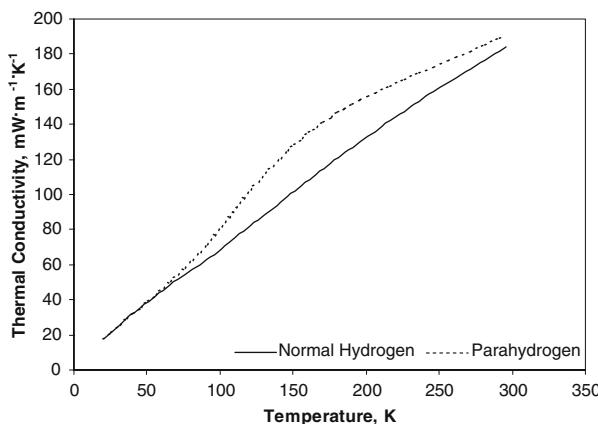


Fig. 1 Thermal conductivity of normal hydrogen and parahydrogen

orthohydrogen to parahydrogen is discussed by McCarty et al. [13]. There are few proven methods for measuring the ratio of orthohydrogen to parahydrogen in the liquefaction process or in storage and transportation. Those being investigated most recently use methods described by Zhou et al. [14] that are based on the differences between the thermal conductivities of the orthohydrogen and parahydrogen species.

3 Experimental Data for the Transport Properties of Hydrogen

Tables 1 and 2 list the available data for the viscosity and thermal conductivity of normal hydrogen and parahydrogen, while Figs. 2–5 graphically display the temperature and pressure regions where data are available. When possible, the original author's estimate of uncertainty (or in some cases, the precision, in italics) is also given. Although the tables summarize a relatively large amount of data, there are very few data sets that provide enough information (detailed description of experimental method, sample purity/composition, complete working equation, analysis of errors) to obtain a reliable uncertainty estimate; the estimates given by the original authors are often unrealistically low and the data sets do not agree with each other to within their stated uncertainties. (This will be demonstrated in the next section.) In addition, data at temperatures below 240 K require a specification of the orthohydrogen-parahydrogen concentration to be reproducible, as the concentration of a non-equilibrium sample at these temperatures changes over time. Data for these temperatures published before 1933 were taken before the existence of orthohydrogen and parahydrogen was established and are compared to normal hydrogen here, although the true concentration of the measured samples is uncertain. While most data sets after 1933 estimate a sample concentration prior to taking data, the only data set with clearly specified measured values of the orthohydrogen-parahydrogen concentration for each point is that of Roder [15].

Table 1 Summary of transport property data for normal hydrogen

Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average deviation (%)	Original author's estimated uncertainty ^a (%)
Thermal conductivity						
Assael and Wakeham [16]	1980	12	307.1–307.7	2–9.2	1.19	0.2
Blais and Mann [17] ^b	1960	8	1200–2100	0.08	4.13	4.0
Chaitkin and Markovich [18]	1958	5	293–503	0.1	10.23	ns
Clerc et al. [19]	1977	16	298.2–373.2	0.1–60	3.49	1.0
Clifford et al. [20]	1981	41	310–385	1.9–23.5	0.58	0.2
Clifford et al. [21]	1980	30	299–301	2–36	0.86	0.2
Clifford et al. [22]	1975	2	77.6–283.2	0.1	1.84	2.6
Dickins [23]	1934	1	282.2	0.08	1.42	0.4
Eucken [24]	1911	4	90.4–373	0.1	2.89	ns
Eucken [25]	1913	4	20.7–273.1	0.1	16.95	ns
Geier and Schafer [26]	1961	13	273.2–1473.2	0.1	18.98	1–2
Golubev and Kalsina [27]	1964	195	78.2–298.2	0.1–49.0	2.77	ns
Gregory [28]	1935	9	295.1–593.4	0.1	3.98	ns
Gregory and Archer [29]	1926	16	273.2	0.0001–0.1	10.41	ns
Hannrin and Thodos [30]	1966	58	274.8–347.8	0.1–66.9	5.23	2.5
Hemminger [31]	1987	6	313–463	0.1	1.11	0.8
Johnston and Grilly [32]	1946	19	80–380	0.1	3.94	0.5
Kannuluuk and Martin [33]	1934	8	274.1	0.004–0.8	2.37	ns
Keyes [34]	1954	12	358.2–523.15	0–14.6	1.42	ns

Table 1 continued

Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average deviation (%)	Original author's estimated uncertainty ^a (%)
Lenoir and Comings [35]	1951	8	315.9	0.1–20.9	4.55	1.5–3
Powers et al. [36]	1954	12	16,81–24,29	0.03–0.3	14.23	2
Roder [15]	1984	689	103.0–300.8	0.6–69.7	0.56	1.5
Roder and Diller [37]	1970	111	17.0–198.0	0.1–11.8	3.35	2
Salceanu and Bojin [38]	1956	1	303.2	0.1	2.7	ns
Saxena and Saxena [39]	1970	32	373.2–1273.2	0.1–0.2	15.42	2
Schneider [40]	1926	6	280.3–315.0	0.002–0.06	1.19	0.4
Sherif [41]	1965	8	92.3–280.0	0.1	5.95	ns
Spencer-Gregory and Dock [42]	1938	11	94.6–280.4	0.1	1.85	ns
Srivastava and Srivastava [43]	1958	1	94.6	0.1	3.12	ns
Stolyarov et al. [44]	1950	23	291.2–575.2	0.1–49.0	7.71	ns
Timrot et al. [45]	1969	17	400–2000	0.1	20.95	ns
Ubbink [46]	1948	14	14.6–273.6	0.1	6.48	ns
van Dael and Cauwenbergh [47]	1968	1	296.8	0.1	0.79	0.5
Vargaftik and Perfenov [48]	1938	10	319.2–710.2	0.1	1.42	ns
Viscosity						
Adzumi [49]	1937	5	293.2–373.2	0.1	4.58	ns
Barua et al. [50]	1964	38	223.2–423.2	1.0–17.8	1.25	0.2
Boyd [51]	1930	56	303–343	7.2–19.4	5.77	ns
Breitenbach [52]	1901	5	252.5–575.2	0.1	1.84	ns

Table 1 continued

Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average deviation (%)	Original author's estimated uncertainty ^a (%)
Buddenberg and Wilke [53]	1951	7	293.2–301.2	0.1	0.53	ns
Chuang et al. [54]	1976	37	173.15–273.15	0.4–50.6	1.82	0.5
Clifford et al. [55]	1981	2	298.2–308.2	0.1	0.07	0.2
Coremans et al. [56]	1958	14	20.5–77.8	0.1	1.51	2–3
Diller [9]	1965	13	14.0–26.0	0.008–0.4	2.18	0.5
Gibson [57]	1933	20	298.2	1.1–29.9	0.52	0.4
Golubev and Petrov [58]	1970	64	298–523	0.1–81.6	1.44	ns
Golubev and Shepeleva [59]	1966	96	77–273	0.9–48.2	2.65	ns
Gracki et al. [60]	1969	42	173–298	0.4–17.1	1.32	0.1–0.2
Guevara et al. [61]	1969	23	1102–2128	0.1	4.61	0.4
Günther [62]	1924	16	15.2–273.1	0.02–0.1	19.13	ns
Ishida [63]	1923	1	321.2	0.1	0.84	0.3
Johns [64]	1939	21	19.0–20.7	0.07–0.1	3.33	1.1
Johnston and McCloskey [65]	1940	7	300	0.02–0.1	0.09	<0.3
Kammerlingh-Onnes et al. [66]	1913	17	20.0–293.9	0.03–0.06	2.17	ns
Keesom and Keesom [67]	1940	7	14.4–20.48	0.008–0.01	16.02	2
Keesom and MacWood [68]	1938	14	14.6–20.3	0.01–0.1	19.11	ns
Kestin and Leidenfrost [69]	1959	9	293.2	0.4–8.4	0.39	0.05
Kestin and Nagashima [70]	1963	13	293.2–303.2	0.1–0.7	0.15	0.2
Kestin and Pilarczyk [71]	1954	1	294.2	0.1	0.07	0.2

Table 1 continued

Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average deviation (%)	Original author's estimated uncertainty ^a (%)
Kestin and Wang [72]	1958	10	298.2	0.1–10.1	0.18	0.2
Kestin and Yata [73]	1968	12	293.2–303.2	0.1–2.4	0.21	0.05
Kestin et al. [74]	1971	3	295.6–308.4	0.1	0.12	0.1
Klemenc and Remi [75]	1923	1	273	0.1	1.13	ns
Kompaneets [76]	1953	7	284–873	0.1	0.66	ns
Kuss [77]	1952	27	292.2–348.2	0.1–49.0	1.83	2
Lukin et al. [78]	1983	23	76.5–293.2	0.1	0.69	0.3
Mal'tsev et al. [79]	2004	3	500–1100	0.3	2.85	3
Menabde [80]	1965	11	77.4–299.7	0.006	0.91	2
Michels et al. [81]	1953	95	298.2–398.2	2.6–186.3	1.26	ns
Nabizadeh and Mayinger [82]	1999	76	295.6–399.2	0.1–5.8	0.91	0.5–1
Powers et al. [36]	1954	12	16.8–24.3	0.1	12.53	2
Rietveld and van Itterbeek [83]	1957	7	14.4–293.2	0.0009–0.005	3.33	ns
Rietveld et al. [84]	1959	14	14.4–293.1	0.0005–0.005	3.16	1–3%
Rudenko and Konareva [85]	1963	8	14.5–20.4	0.01–0.1	4.95	1.3
Rudenko and Slyusar [86]	1968	91	33.2–300.0	0.1–217.1	7.68	4–6
Sutherland and Maas [87]	1932	10	74.8–293.9	0.1	0.85	<0.4
Trautz and Baumann [88]	1929	12	195.8–525.2	0.1	1.15	ns
Trautz and Binkley [89]	1929	6	298.2–523.2	0.1	0.73	ns
Trautz and Heberling [90]	1931	4	300.0–551.3	0.1	0.64	ns

Table 1 continued

Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average deviation (%)	Original author's estimated uncertainty ^a (%)
Trautz and Kurz [91]	1931	12	301.3–551.2	0.1	0.71	ns
Trautz and Ludewigs [92]	1929	20	288.7–525.2	0.1	1.04	ns
Trautz and Melster [93]	1930	8	298.2–550.1	0.1	0.90	ns
Trautz and Sorg [94]	1931	12	292.7–523.2	0.1	0.62	ns
Trautz and Stauf [95]	1929	15	192.4–524.9	0.1	0.93	ns
Trautz and Zimmerman [96]	1935	1	363.2	0.1	175.2	ns
Trautz and Zink [97]	1930	8	289.9–1098.2	0.1	0.87	ns
Tsederberg et al. [98]	1965	14	288.7–644.2	4.4–50.55	1.21	3
van Cleave and Maass [99]	1935	1	295	0.1	0.75	0.2
van Iterbeek and Claes [100]	1938	11	14.0–292.9	0.0007–0.02	3.80	ns
van Iterbeek and Claes [101]	1938	8	14.1–293	0.008–0.1	3.72	ns
van Iterbeek and van Paemel [102]	1941	5	14.7–20.4	0.01–0.1	4.09	ns
van Iterbeek and van Paemel [103]	1940	34	14.9–68.9	0.00004–0.1	126.8	ns
van Iterbeek and van Paemel [104]	1940	7	12.7–298.2	0.01–0.1	3.58	ns
Verschaffelt and Nicaise [105]	1917	1	20.4	0.1	-15	ns
Vogel [106]	1914	7	21.0–273.1	0.02–0.1	5.68	
Wobser and Müller [107]	1941	5	298.2–371.2	0.1	0.38	1–4
Yen [108]	1919	1	295.2	0.1	0.60	ns

^a ns, not specified^b Only graphical results presented. Graph was digitized for data comparisons

Table 2 Summary of transport property data for parahydrogen

Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average deviation (%)	Original author's estimated uncertainty (%)
Thermal conductivity						
Dwyer et al. [109]	1966	12	15.1–21.4	0.02–21.9	53.35	58
Powers et al. [36]	1954	7	16.8–23.2	0.1	15.22	2
Roder [15]	1984	269	98.8–273.6	0.9–12.4	0.43	1.5
Roder and Diller [37]	1970	136	17.1–153.0	0.02–18.4	7.79	2.0
Diller [9]	1965	327	14.0–100.0	0.65–34.5	1.18	0.5

4 Comparisons of Calculated Transport Properties of Hydrogen with Experimental Data

Comparisons of calculated properties to measurements can provide a basis for the assessment of the quality of the representation of the transport properties model and of the variations among the data sets. In Tables 1 and 2, we list the absolute average deviations (AAD) of transport properties calculated with the standard model, with the definitions:

$$AAD = \frac{1}{n} \sum_{i=1}^n |\% \Delta X_i|, \quad \text{where} \quad \% \Delta X = 100 \left(\frac{X_{\text{data}} - X_{\text{calc}}}{X_{\text{data}}} \right). \quad (1)$$

X is any property, n is the number of data points in the data set, and i is the index for each data point. Even though the range of applicability of the formulations is exceeded, comparisons are included to data above 400 K, to illustrate the extrapolation behavior of the formulations.

4.1 Normal Hydrogen

Figures 6 and 7 compare the thermal conductivity of normal hydrogen calculated with REFPROP [6] to experimental data as functions of pressure and temperature, respectively. In Fig. 6 we see that the accurate wide-ranging data of Roder [15] are represented to within their uncertainty, 1.5%, except for pressures from 60 to 70 MPa, where the correlation begins to deviate from the data and reaches 5% deviations at 70 MPa. In order to meet the demands of high-pressure storage applications, pressures as high as 70 MPa are of interest [110]; therefore, a correlation should be developed that represents the data to within their uncertainty at least to 70 MPa. Although there is a large amount of scatter, most of the data sets are represented to within 5%, except for data at temperatures above 400 K (the limit of the correlation), as shown in Fig. 7. The deviations from these data sets increase as the temperature increases, approaching 30% at 1,000 K, demonstrating that the present correlation does not extrapolate well outside of its recommended temperature range. In order to support proposed nuclear hydrogen generation processes (above 900 K) [111], it is necessary to extend the range of the transport formulations to higher temperatures. In addition, Assael et al. [3] assessed the low pressure gas-phase data, and concluded that the only data of quality high enough to be suitable for correlation development are those from Roder [15], Clifford et al. [20, 21], and Assael and Wakeham [16], none of which cover temperatures above 400 K. Therefore, even though there are ten data sets for thermal conductivity of normal hydrogen in the temperature range 400–2000 K, additional measurements are recommended in this temperature range to support correlation development.

Figures 8 and 9 compare the viscosity of normal hydrogen calculated with REFPROP [6] to experimental data as a function of pressure and temperature, respectively. Within the stated range of validity of the correlation, up to 400 K and 121 MPa, there is considerable scatter, and most deviations are within 10%. Figure 8 shows that the correlation agrees with the wide-ranging data of Michels et al. to within 2%

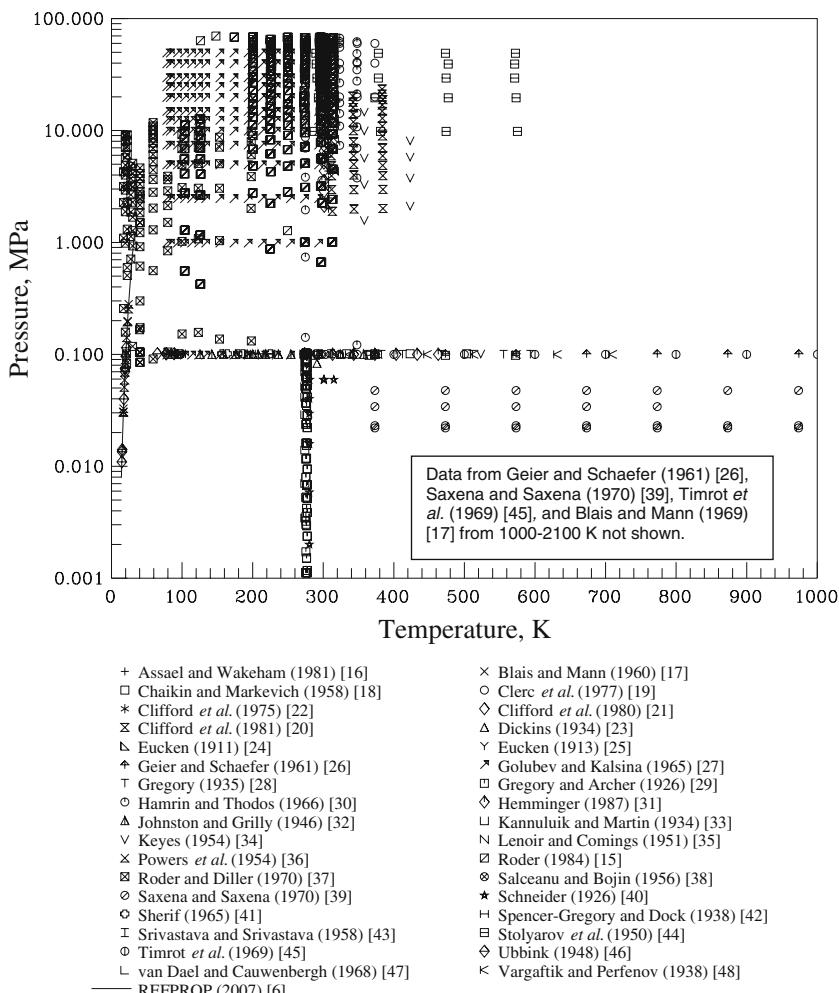


Fig. 2 Thermal conductivity data for normal hydrogen

up to 100 MPa. The data of Rudenko and Slyusar [86] and the data of Golubev and Shepeleva [59] are both wide-ranging, comprehensive data sets, but they show very different trends below 100 K. Thus there is a need for more measurements in the region from about 40 K to 100 K that extend to 100 MPa. Figure 9 shows that the correlation represents the data to within about 3% from 400 K to about 1,000 K, indicating that the extrapolation behavior is reasonable in this range. There are 14 data sets available for the viscosity of normal hydrogen at temperatures over 400 K ranging up to 2,128 K. Large systematic deviations up to 12% occur above 1,000 K with the data of Guevara et al. [61] that may indicate limitations in the extrapolation of the correlation to temperatures above 1,000 K. Measurements of both the thermal conductivity and viscosity

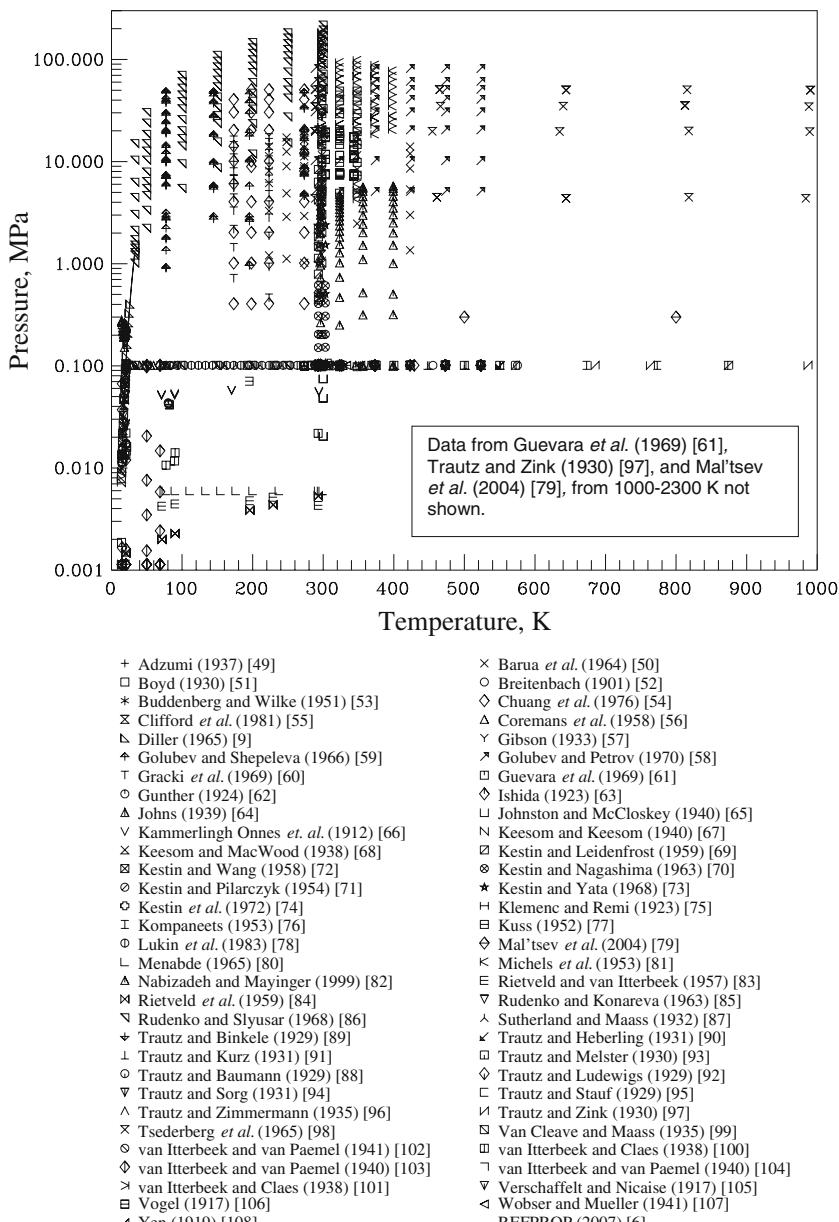


Fig. 3 Viscosity data for normal hydrogen

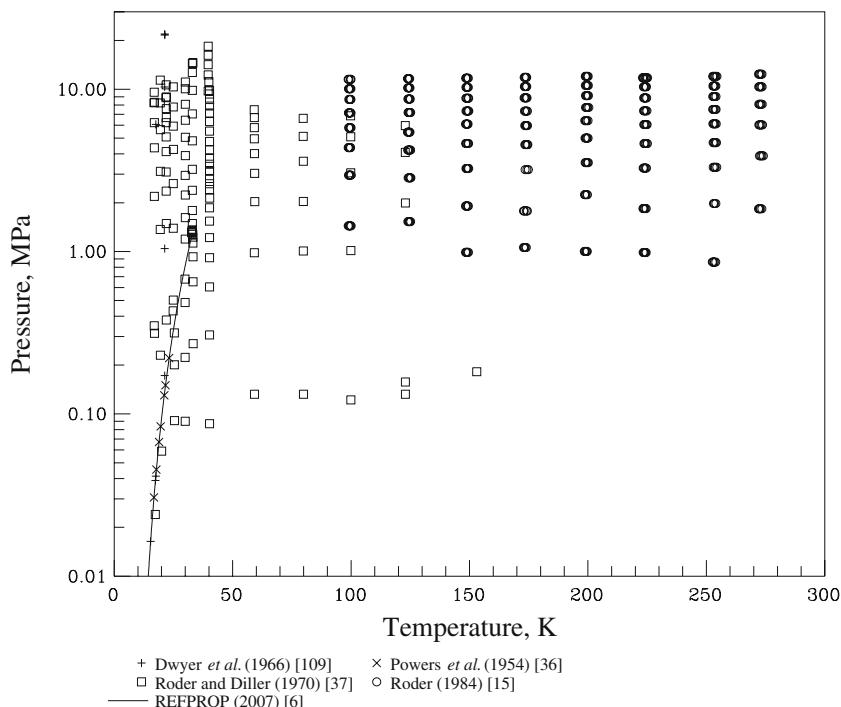


Fig. 4 Thermal conductivity data for parahydrogen

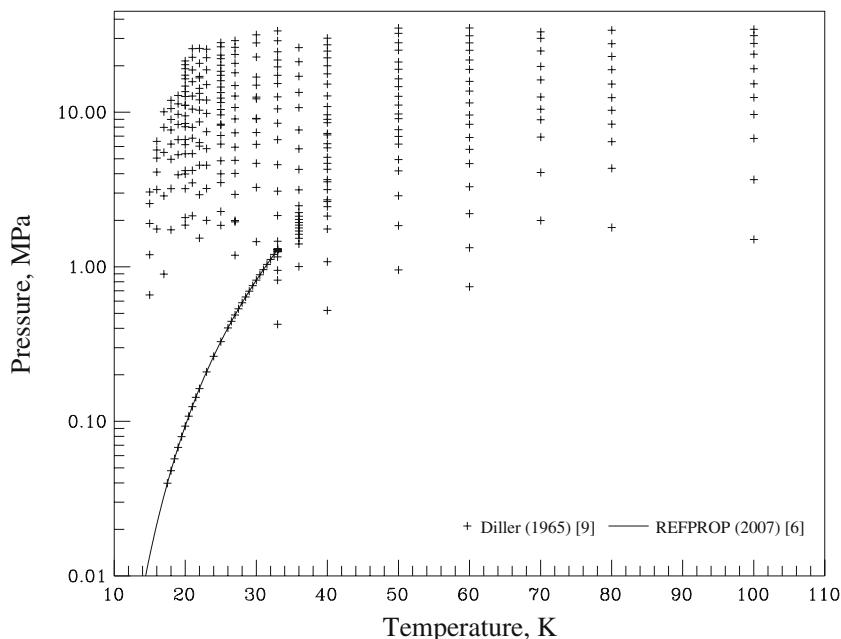


Fig. 5 Viscosity data for parahydrogen

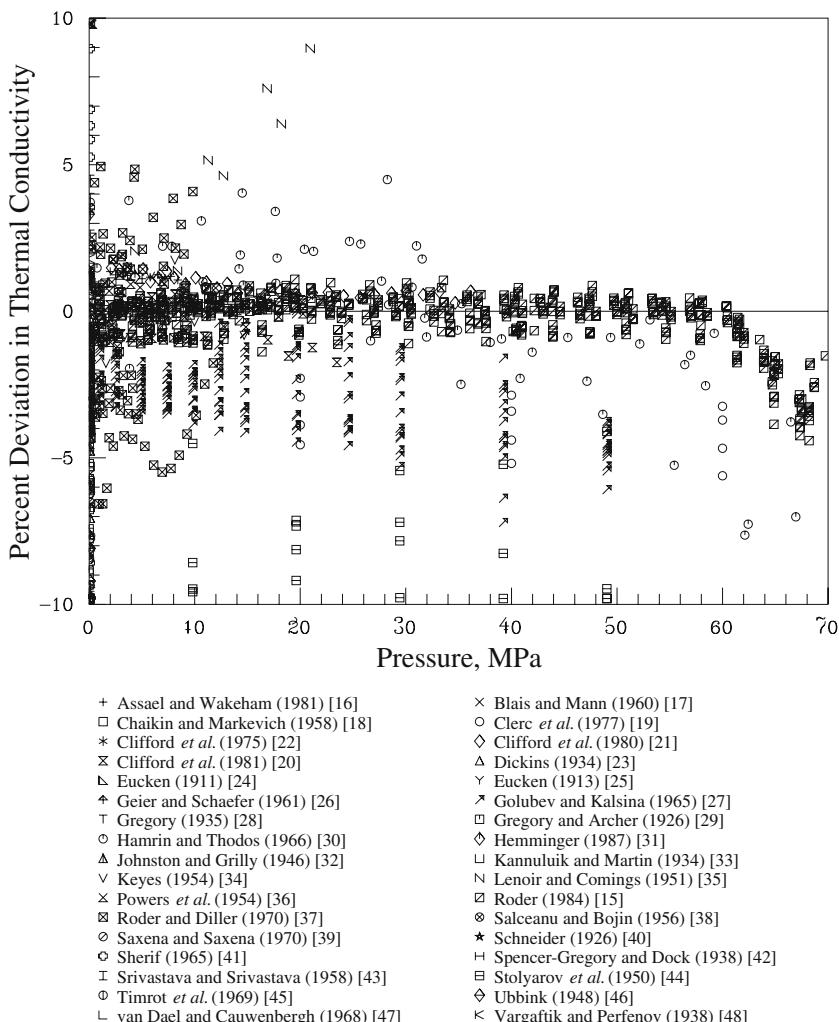


Fig. 6 Comparisons of thermal conductivity calculated by use of REFPROP [6] with experimental normal hydrogen data (% ΔX versus Pressure)

of normal hydrogen should be extended to 5.5 MPa and 1,300 K, consistent with coal gasification hydrogen generation processes [112] (Figs. 10–13).

4.2 Parahydrogen

Thermal conductivity data for parahydrogen range from the saturated liquid line to 273.6 K. The data should be extended to higher temperatures (350 K) consistent with engineering applications. The thermal conductivity formulation for parahydrogen fits the existing data of Roder and Diller [37] to within about 5%, and Roder [15] to within

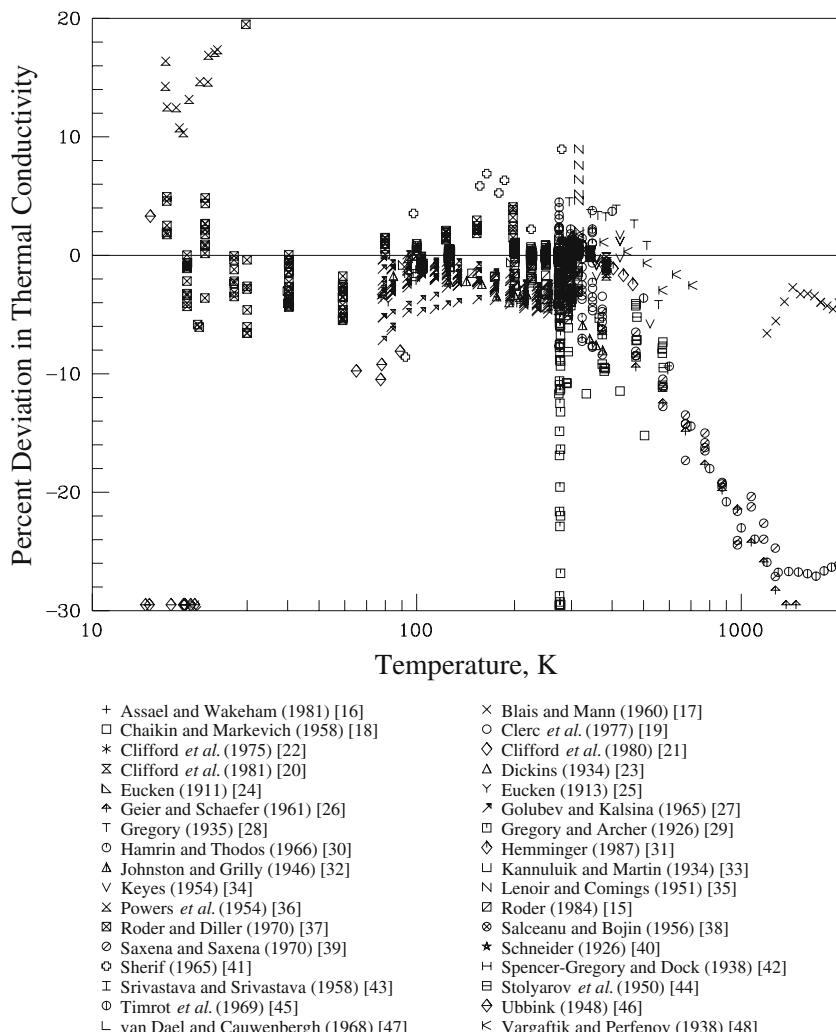


Fig. 7 Comparisons of thermal conductivity calculated by use of REFPROP [6] with experimental normal hydrogen data (% ΔX versus Temperature)

2% except near the critical region. The near-critical data at 33 K of Roder and Diller [37] have very large deviations, approaching 80%. The present correlation does not diverge at the critical point, leading to very large deviations in the critical region. Any future correlation should incorporate the correct behavior in the critical region.

Viscosity data for parahydrogen range from 15 to 100 K, and are reproduced by the standard formulation to within about 2%. This exceeds the reported uncertainty of the data set. Also, since there is only one source of data, we recommend that additional measurements be made to verify the existing data. Hanley *et al.* [113] stated that for the gas phase, the differences between the viscosities of normal hydrogen and parahydrogen are negligible. However, in the liquid region, differences have

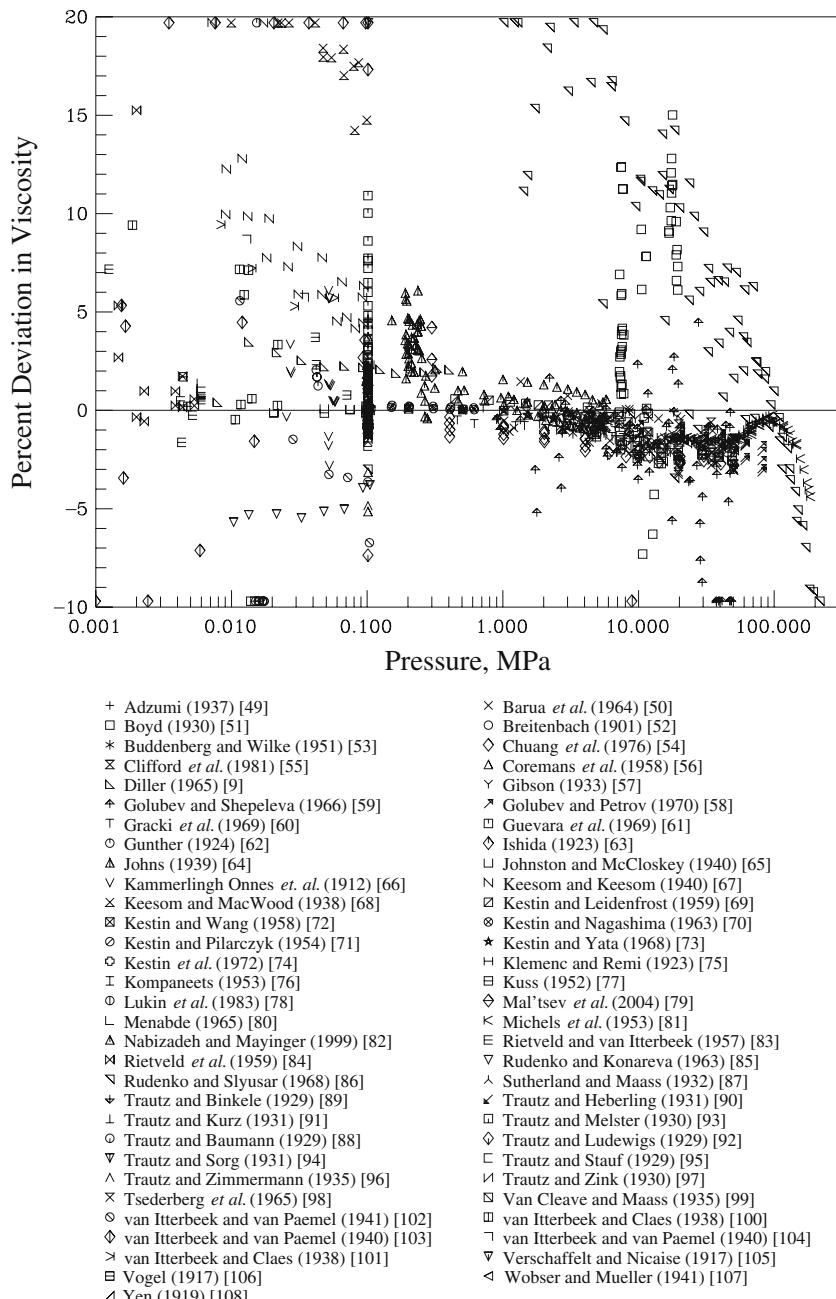


Fig. 8 Comparisons of viscosity calculated by use of REFPROP [6] with experimental normal hydrogen data (% ΔX versus Pressure)

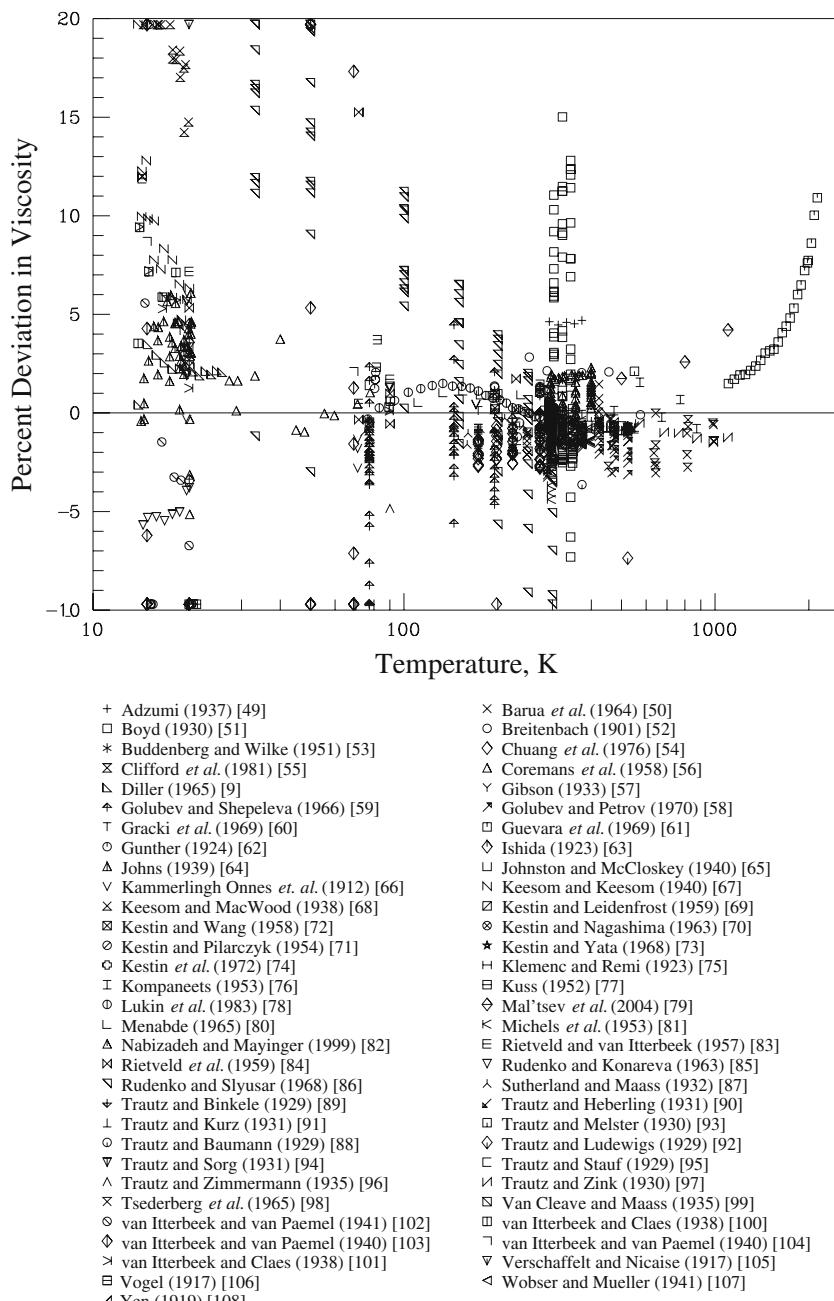


Fig. 9 Comparisons of viscosity calculated by use of REFPROP [6] with experimental normal hydrogen data (% ΔX versus Temperature)

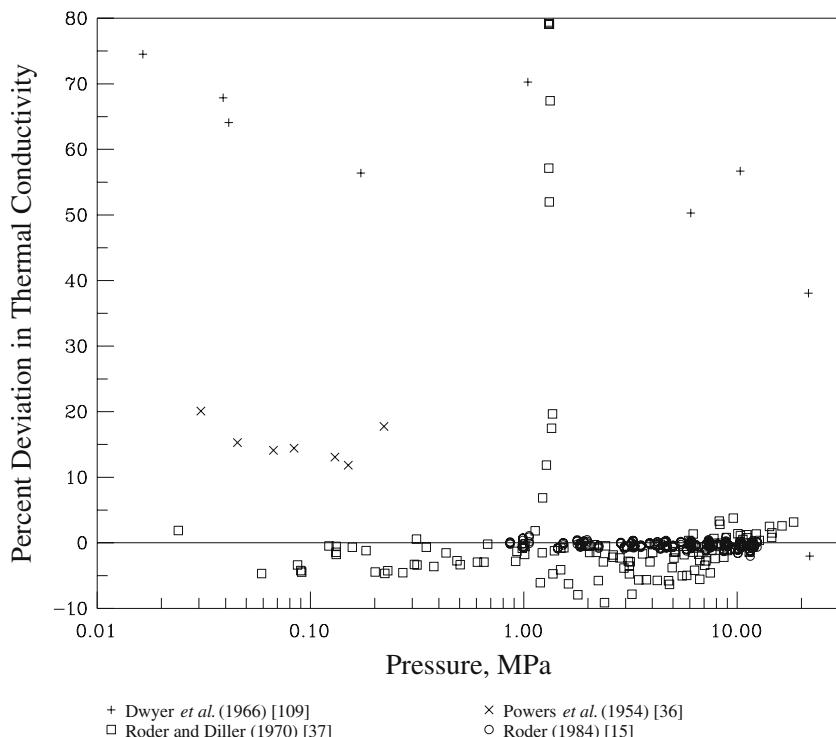


Fig. 10 Comparisons of thermal conductivity calculated by use of REFPROP [61] with experimental parahydrogen data (% ΔX versus Pressure)

been reported. Stephan and Lucas [2] found that their recommended values for liquid parahydrogen and liquid normal hydrogen differed by as much as 5–10% and sometimes more. Diller [9] reported differences in the measured viscosity of the saturated liquid of up to 5% between normal hydrogen and parahydrogen, which is greater than his reported uncertainty. Becker and Stehl [114, 115] also report differences between parahydrogen and normal hydrogen ranging from 1% at 90 K to 7% at 15 K. However, their reported uncertainty is relatively large, 5%, making it difficult to separate what is a true difference and what is measurement error. We recommend that additional low-uncertainty measurements of normal hydrogen and parahydrogen in the liquid region be made to help clarify whether there are significant differences (exceeding the level of uncertainty of the measurements) that should be accounted for in any future correlation efforts.

5 Conclusions

The goal of any reference quality correlation is the representation of the experimental data to within the experimental uncertainty. As shown in the previous section, there is clearly room for improvement in the correlations for viscosity and thermal

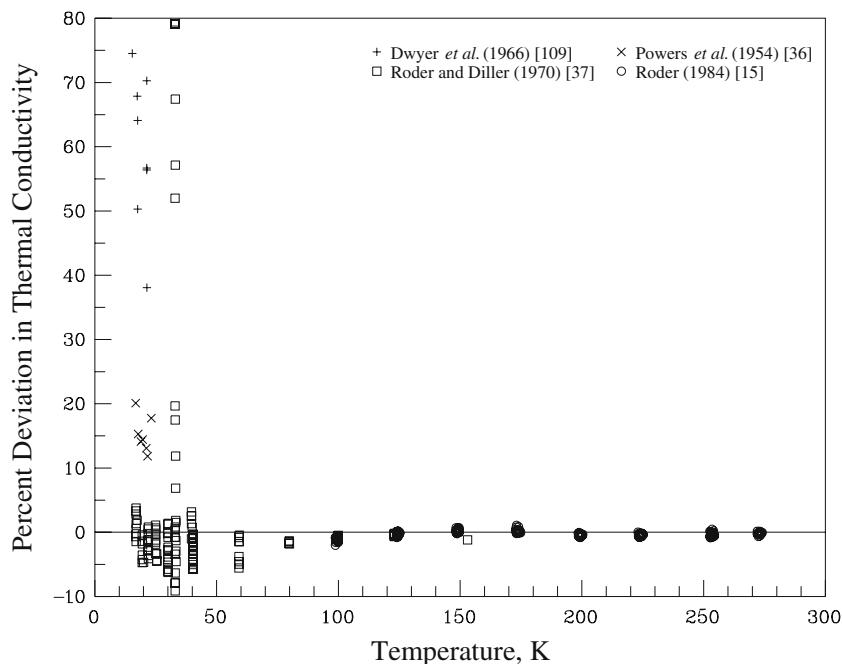


Fig. 11 Comparisons of thermal conductivity calculated by use of REFPROP [6] with experimental para-hydrogen data (% ΔX versus Temperature)

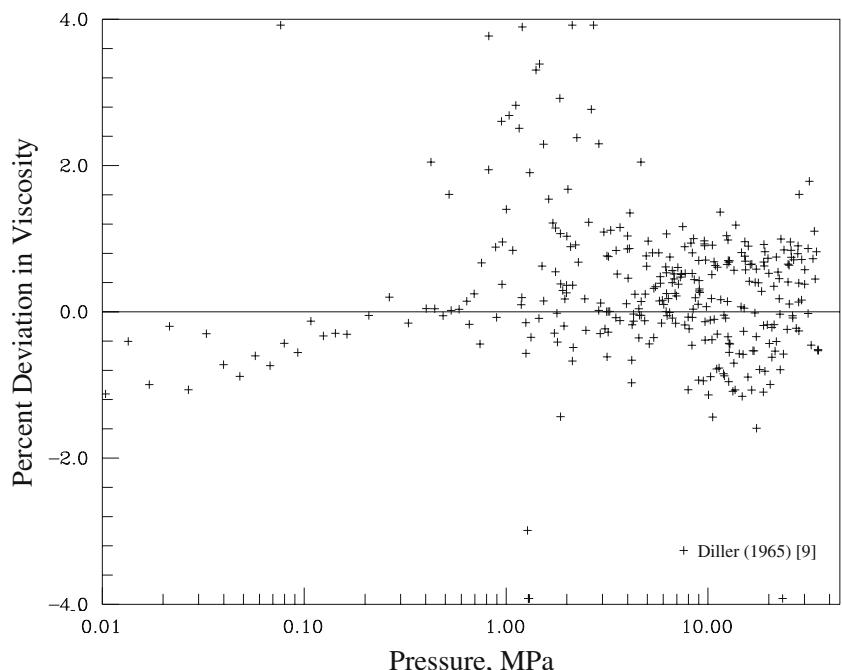


Fig. 12 Comparisons of viscosity calculated by use of REFPROP [6] with experimental parahydrogen data (% ΔX versus Pressure)

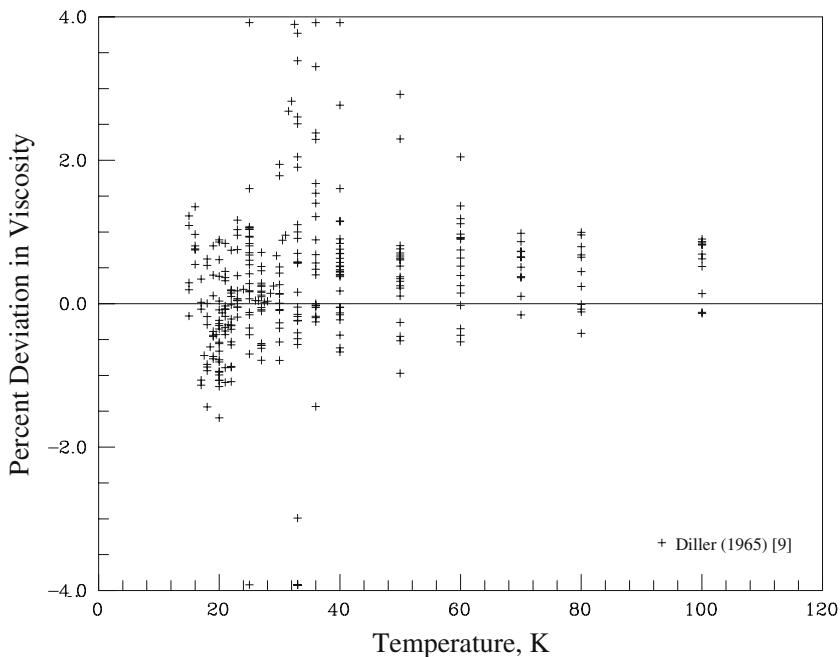


Fig. 13 Comparisons of viscosity calculated by use of REFPROP [6] with experimental parahydrogen data ($\% \Delta X$ versus Temperature)

conductivity of hydrogen, and revised correlations should be developed. We also noted that there are deficiencies in the existing experimental data and made recommendations on the need for additional measurements with low uncertainties. The state of the art in experimental techniques, regression methods, equation of state (EOS) development, and theory (especially in the critical region) have progressed since the publication of these correlations, and all of these factors can contribute to the development of improved formulations. Development of equations of state is especially important because correlations for viscosity and thermal conductivity are typically formulated in terms of density and temperature, while measurements are usually reported in terms of temperature and pressure. Thus, the analysis of the experimental measurements often requires density, which is not directly measured but rather obtained independently from an EOS or other correlation. As recently pointed out [11], there are deficiencies in the equation of state of Younglove [116] that is presently widely used. The development of an improved equation of state will allow improvement in the representation of the transport surfaces. In addition, any future correlations also should be revised to be consistent with the ITS-90 temperature scale. This is most important at temperatures below 20 K and above 500 K, where deviations between temperature scales become significant.

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