

Current Status of Thermodynamic Properties of Hydrogen

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Abstract This article presents a survey of the thermodynamic properties of normal hydrogen and parahydrogen and gives comparisons of properties calculated from the standard models to available experimental data. Motivated by the fact that the currently accepted standards for the thermodynamic properties of hydrogen were based on experimental and correlation work completed before the mid-1980s, an assessment of the accuracy of property values for analysis and design of new systems at high and low temperatures and pressures is provided. Property values measured and published after the completion of the current standards for both normal hydrogen and parahydrogen are included in the comparisons. Recommendations for new experimental data needed and for new thermodynamic property formulations for normal hydrogen and parahydrogen are included.

Keywords Equation of state · Hydrogen · Normal hydrogen · Parahydrogen · Thermodynamic properties

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1 Introduction

Hydrogen is currently used in the production of chemicals (particularly fertilizers), in petroleum refining, in metals treatment, and as a rocket fuel in significant quantities. In 2004, 50 million metric tons of hydrogen were produced globally, 48% of which was produced by steam reforming of natural gas. If the proposed future use of hydrogen is realized as a replacement for conventional fuels in transportation and other applications, accurate properties will be needed at states over a wide range of pressures and temperatures to support such a “hydrogen economy.”

In this article, the designation “standard” is used to refer to the equation of state published by Younglove [1]. This equation was published in 1982 based on work done during the 1970s through the early 1980s at the National Bureau of Standards (NBS), in Boulder, Colorado, now the National Institute of Standards and Technology (NIST). This article reviews the current status of the equations of state for normal hydrogen and parahydrogen and establishes priorities for the development of new models that extend the ranges of temperature and pressure.

2 Hydrogen Behavior

Unique problems in developing a new property formulation are presented by the chemistry of hydrogen. The hydrogen atom is arguably one of the most studied structures in chemistry, with almost every textbook on fundamental chemistry including a chapter or section on this subject. The hydrogen atom has a single proton as its nucleus and a single orbiting electron. The hydrogen molecule is composed of two hydrogen atoms, which occur in two forms or allotropes called orthohydrogen and parahydrogen.

The differentiating feature of orthohydrogen and parahydrogen is the relative orientation of the nuclear spin of the individual atoms. The nuclear spin is associated with the rotational motion of the nucleus about an axis perpendicular to the molecular axis (a line between the atoms), and the spins in the two atoms forming the hydrogen molecule may be in the same direction (parallel), or in opposite directions (anti-parallel). The spin orientation relative to the individual nuclei of the molecule has a significant influence on the rotational states of the molecule. There are quantum numbers for the spin and the corresponding energy levels. The molecules with anti-parallel nuclear spins, called parahydrogen, have even rotational quantum numbers ($J = 0, 2, \dots$) and are in the lowest energy states. The molecules with parallel nuclear spins, called orthohydrogen, have odd quantum numbers ($J = 1, 3, \dots$) and are predominant at higher temperatures [2].

Hydrogen may be thought of as a binary mixture of two different species of molecules with different physical properties. The equilibrium percentage of the orthohydrogen and parahydrogen concentrations in the mixture is temperature dependent. Equilibrium hydrogen is defined as a mixture that has achieved the equilibrium concentration ratio of orthohydrogen–parahydrogen at a given temperature. At a temperature of 80 K, the equilibrium concentration is approximately 50% orthohydrogen and 50% parahydrogen, while at room temperature the equilibrium mixture is about 75% orthohydrogen and 25% parahydrogen, which is the concentration that is commonly

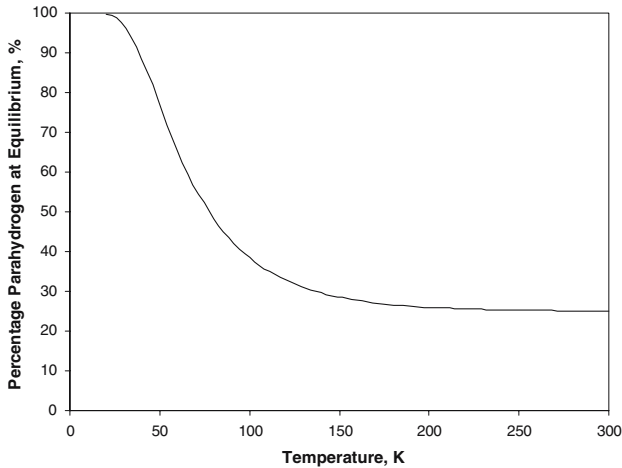


Fig. 1 Temperature dependence of orthohydrogen to parahydrogen at an equilibrium concentration

referred to as “normal hydrogen.” Figure 1, modified from NBS Monograph 168 [3], shows the equilibrium percentage of parahydrogen in this “mixture” as a function of temperature. At high temperatures (above 240 K), the equilibrium mixture is nearly constant at 75% orthohydrogen and 25% parahydrogen. At zero kelvin, all the molecules must be in a rotational ground state at equilibrium, meaning that all should be parahydrogen. In the literature, parahydrogen is usually defined as a mixture of 99.75% parahydrogen molecules and 0.25% orthohydrogen molecules, which is the equilibrium mixture at 19 K. Although nearly pure parahydrogen can be prepared at equilibrium at low temperatures, at higher temperatures the maximum possible concentration of orthohydrogen is 75% in an equilibrium state.

The self conversion of a non-equilibrium orthohydrogen–parahydrogen composition to an equilibrium composition is very slow in the absence of a catalyst. The main mechanism of natural orthohydrogen/parahydrogen transformation is the magnetic dipole interaction of the nuclear moments of orthohydrogen molecules [4]. The number of interactions between hydrogen molecules increases with density such that the orthohydrogen–parahydrogen conversion occurs slowly in the gaseous state, becoming more rapid as the density increases [2,4]. Conversion rates can be enhanced by using a paramagnetic catalyst that will reverse the nuclear spins without breaking the H–H bonds [5].

National Bureau of Standards Report 8812 summarizes the property differences of orthohydrogen and parahydrogen [6]. In general, larger differences in the properties of orthohydrogen and parahydrogen occur in properties related to specific heat. Properties such as enthalpy, entropy, and heat capacity can show large differences. Due to the large differences in thermal conductivity, measurements of this property are used to determine the ratio of orthohydrogen to parahydrogen in a mixture [7–9]. Away from the critical region (at temperatures over 60 K), differences in density are generally very small with orthohydrogen having larger densities at the same temperature and pressure. The critical points and triple points for normal hydrogen ($T_c = 33.19$ K,

$P_c = 1.315$ MPa, $T_{\text{trp}} = 13.957$ K, $P_{\text{trp}} = 0.0072$ MPa) and parahydrogen ($T_c = 32.976$ K, $P_c = 1.2928$ MPa, $T_{\text{trp}} = 13.803$ K, $P_{\text{trp}} = 0.00704$ MPa) are different, as are the saturation boundaries. These values are all taken from NBS Monograph 168 [3]. Temperatures are given on the NBS-55 temperature scale and no uncertainties are given except for ± 0.05 K on the parahydrogen critical point temperature.

3 Experimental Data for the Thermodynamic Properties of Hydrogen

Current data [10–77] for the thermodynamic properties of normal hydrogen and parahydrogen are listed in Tables 1 and 2. P - ρ - T and speed-of-sound data for normal hydrogen are illustrated in Figs. 2 and 3, and those for parahydrogen are illustrated in Figs. 4 and 5. Included are comparisons to data above 400 K (even though the range of applicability of the formulations is exceeded) to illustrate the extrapolation behavior of the formulations. Data at temperatures below 240 K require a specification of the orthohydrogen–parahydrogen concentration to characterize the sample because the equilibrium concentration differs from the “normal” 75–25 ratio, so that a sample in

Table 1 Summary of thermodynamic property data for normal hydrogen

Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average deviation (%)
P-ρ-T					
Bartlett [10]	1927	8	273.2	5.1–101.3	0.37
Bartlett et al. [11]	1928	43	273.2–673.0	5.1–101.3	0.29
Bartlett et al. [12]	1930	54	203–293	2.6–102.7	0.38
David and Hamann [13]	1953	12	65–79	30.4–126.7	0.53
Golubev and Dobrovolskii [14]	1964	31	33.2–77.4	4.8–47.6	1.03
Holborn and Otto [15]	1925	30	65.25–223.1	2.0–10.0	0.17
Jaeschke and Humphreys [16]					
Gasunie	1990	68	273.2–353	0.2–26.3	0.04
Ruhrgas	1990	221	273.2–353	0.5–28.1	0.04
Johnston et al. [17]	1954	62	20.3–32.6	0.9–11.9	0.68
Johnston et al. [18]	1953	227	33–300	0.5–20.6	0.69
Liebenberg et al. [19]*	1978	19	75.0–163.9	473.3–1871	–
Liebenberg et al. [20]	1977	1953	75–307	200–2,000	4.78
Machado et al. [21]	1988	60	130–159	1.2–105.5	5.16
Michels and Goudekot [22]	1941	283	273–423	0.9–300.9	0.13
Michels et al. [23]	1959	482	98.2–423.2	0.7–299.2	0.11
Scott [24]	1929	18	298	0.10–17.2	0.15
Townend and Bhatt [25]	1931	40	273–298	0.1–60.8	0.11
Tsiklis et al. [26]	1975	45	298.1–423.1	50–650	0.52

Table 1 continued

Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average deviation (%)
Verschoyle [27]	1926	25	273–293	5.0–21.0	0.15
Wiebe and Gaddy [28]	1938	47	273–573	2.5–101.3	0.08
Isobaric heat capacity					
Gutsche [29]	1939	73	16.5–37.5	1.0–9.7	7.95
Speed of sound					
Guesewell et al. [30]	1970	7	25–31	0.1	6.37
Liebenberg et al. [19]*	1978	19	75.0–163.9	473.3–1871	–
Liebenberg et al. [20]	1977	1953	75–307	200–2,000	9.30
Matsuishi et al. [31]*	2003	42	293–526	1,190–10,840	–
van Dael et al. [32]	1965	175	22.2–33	0.2–24.8	1.82
van Itterbeek et al. [33]	1961	42	14.1–20.4	0.009–0.1	1.69
van Itterbeek et al. [34]	1963	110	15.1–20.5	0.02–23.5	6.37
Vapor pressure					
Aston et al. [35]	1935	4	18.0–20.7	Sat	4.65
Barber [36]	1964	1	13.816	Sat	3.02
Grilly [37]	1951	8	19.3–24.5	Sat	3.53
Henning [38]	1926	25	14.0–20.5	Sat	6.78
Henning and Otto [39]	1936	19	13.93–20.38	Sat	13.11
Hiza [40]	1981	12	20.0–30.0	Sat	2.40
Keesom et al. [41]	1931	31	17.2–20.5	Sat	4.25
Traver and Jaquero [42]	1902	9	14.9–20.4	Sat	3.53
van Itterbeek et al. [43]	1964	42	20.6–32.3	Sat	3.54
White et al. [44]	1950	17	20.9–33.1	Sat	3.79
White et al. [45]	1950	6	33.08–33.25	Sat	3.33
Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average difference ($\text{cm}^3 \cdot \text{mol}^{-1}$)
Second virial coefficient					
Bartlett et al. [11]	1928	5	273.2–572.3		0.52
Beenakker et al. [46]	1959	1	20.4		10.02
Cottrell et al. [47]	1956	1	303.2		1.04
Dehaas [48]	1912	3	289.1–293.7		15.99
El Hadi et al. [49]	1969	7	19.3–26.3		1.48
Gibby et al. [50]	1929	7	298.2–448.2		0.48
Holborn and Otto [15]	1925	8	90.2–473.2		3.05
Holborn and Otto [51]	1926	9	65.3–473.2		11.12
Johnston et al. [18]	1953	18	35.1–300		0.20
Kerl [52]	1982	1	293.1		–

Table 1 continued

Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average deviation (%)
Knaap et al. [53]	1962	23	20.5–65.0		5.69
Long and Brown [54]	1937	7	20.9–46.5		1.83
Lopatinskii et al. [55]	1991	2	293.2		0.43
Michels and Goudekete [22]	1941	20	273.2–423.2		1.14
Michels et al. [23]	1959	17	98.2–423.2		0.77
Mihara et al. [56]	1977	3	298.2–348.2		0.57
Mueller et al. [57]	1961	6	73.2–323.2		8.85
Nijhoff and Keesom [58]	1927	8	24.84–373.15		1.54
Perez et al. [59]	1980	5	300–500		0.75
Schramm et al. [60]	1991	1	296.2		0.92
Scott [24]	1929	1	298.2		0.45
Townend and Bhatt [25]	1931	2	273.2–298.2		0.60
van Agt and Kamerlingh Onnes [61]	1925	9	14.6–90.3		3.90
Varekamp and Beenakker [62]	1959	8	14.0–21.0		8.87
Verschoye [27]	1926	2	273.2–293.2		0.44
Wiebe and Gaddy [28]	1938	6	273.2–573.2		0.39
Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average difference (cm ⁶ ·mol ⁻²)
Third virial coefficient					
Holborn and Otto [15]	1925	5	90.2–273.2		0.99
Johnston et al. [18]	1953	18	35.1–300.0		0.11
Michels and Goudekete [22]	1941	20	273.2–423.2		0.53
Michels et al. [23]	1959	17	98.2–423.2		0.14
Mihara et al. [56]	1977	3	298.2–348.2		0.12
Scott [24]	1929	1	298.2		0.20
Townend and Bhatt [25]	1931	2	273.2–298.2		0.12
Verschoye [27]	1926	2	273.2–293.2		0.03

* Some of the data in these data sets crossed the current solid phase boundary and has been omitted from comparisons

equilibrium at room temperature would change composition over time. Data published before 1933 were measured before the existence of orthohydrogen and parahydrogen had been established and are compared here to the normal hydrogen equation of state even though the true concentration of the measured samples is uncertain. While most data sets after 1933 estimate a sample concentration prior to taking data, the concentration of each data point for hydrogen that is not at equilibrium is generally not given and can vary throughout the measurements.

Table 2 Summary of thermodynamic property data for parahydrogen

Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average deviation (%)
P- ρ -T					
Goodwin et al. [63]	1963	1,234	15–100	1.5–35.5	0.87
Goodwin et al. [64]	1961	17	17.0–33.0	Sat	0.02
Hoge and Lassiter [65]	1951	46	32.9–33.3	1.3–1.4	4.41
Roder et al. [66]	1963	46	33.0–40.0	1.3–2.8	2.33
Isochoric heat capacity					
Younglove and Diller [67]	1962	162	19.9–90.4	1.1–63.3	1.24
Isobaric heat capacity					
Medvedev et al. [68]	1971	319	20.9–50.3	0.2–3.0	4.56
Speed of sound					
Younglove [69]	1965	251	14.5–100.0	Sat–32.0	1.58
van Itterbeek et al. [33]	1961	48	14.1–20.4	Sat	2.88
van Itterbeek et al. [34]	1963	116	15.1–20.5	Sat–23.5	1.52
van Dael et al. [70]	1965	23	20.3–32.0	Sat	4.25
Vapor pressure					
Barber and Horsford [71]	1963	10	13.8–20.3	0.007–1.2	0.20
Hoge and Arnold [72]	1951	45	15.8–32.9	0.02–1.2	0.16
Keesom et al. [41]	1931	31	17.2–20.5	0.1	1.12
van Itterbeek et al. [43]	1964	42	20.6–32.3	0.1–1.2	0.86
Weber et al. [73]	1962	32	20.3–32.7	0.1–1.2	0.32
Saturation heat capacity					
Brouwer et al. [74]	1970	12	24.5–30.0	Sat	0.73
Smith et al. [75]	1954	8	18.3–31.5	Sat	1.04
Johnston et al. [76]	1950	16	12.7–19.0	Sat	2.45
Younglove and Diller [67]	1962	32	14.8–31.5	Sat	2.73
Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average difference (cm ³ ·mol ⁻¹)
Second virial coefficient					
Goodwin et al. [77]	1964	58	15.0–423.2		0.78
Author	Year	Number of points	Temperature range (K)	Pressure range (MPa)	Absolute average difference (cm ⁶ ·mol ⁻²)
Third virial coefficient					
Goodwin et al. [77]	1964	52	20.0–423.2		0.29

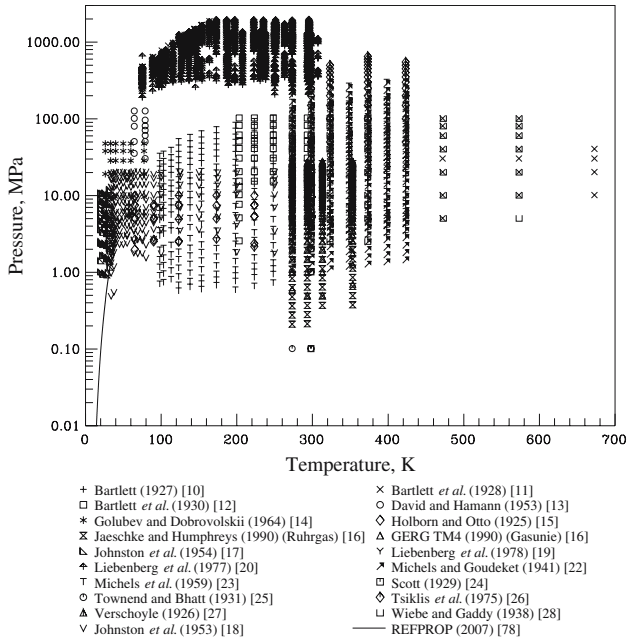


Fig. 2 P- ρ -T data for normal hydrogen

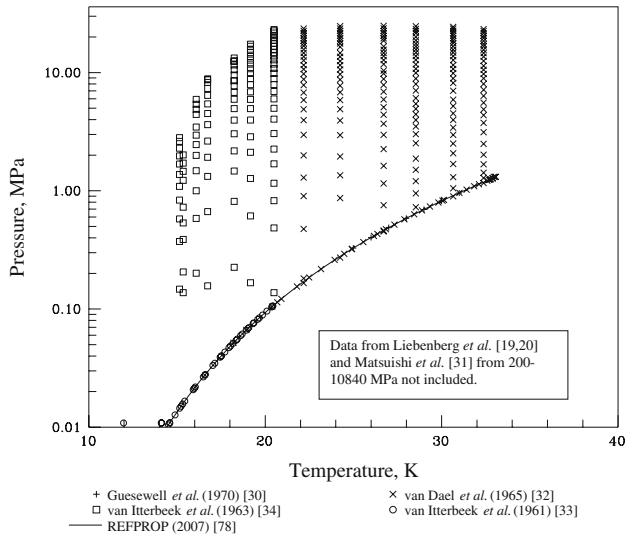


Fig. 3 Speed-of-sound data for normal hydrogen

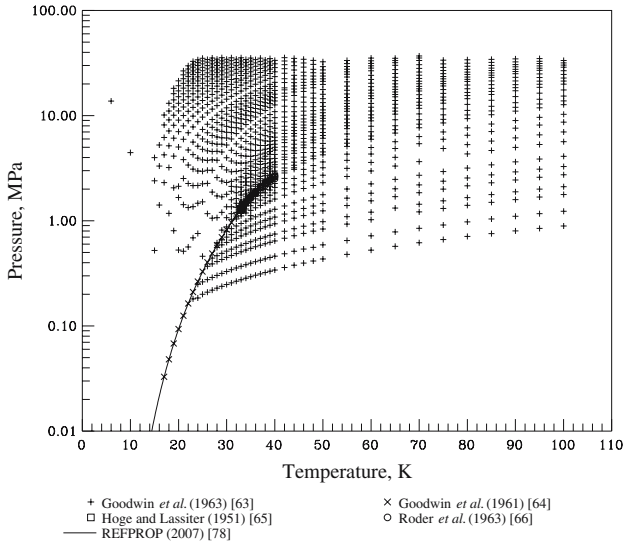


Fig. 4 P - ρ - T data for parahydrogen

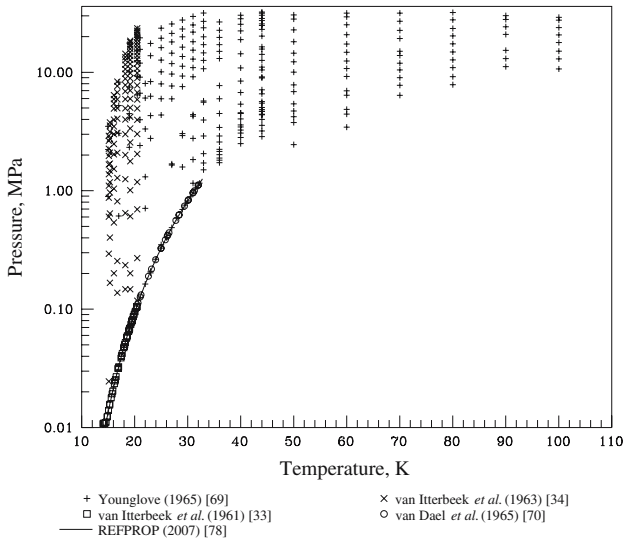


Fig. 5 Speed-of-sound data for parahydrogen

4 Comparisons of Calculated Thermodynamic Properties of Hydrogen to Data

Comparisons of calculated properties to measurements provide the basis for assessment of the accuracy and precision of the experimental data and the quality of the representation of the surface of state by the available model(s). Absolute average deviations (AAD) of calculated properties of normal hydrogen are given in the right-hand column of Table 1. Similar comparisons for parahydrogen are given in the right-hand column

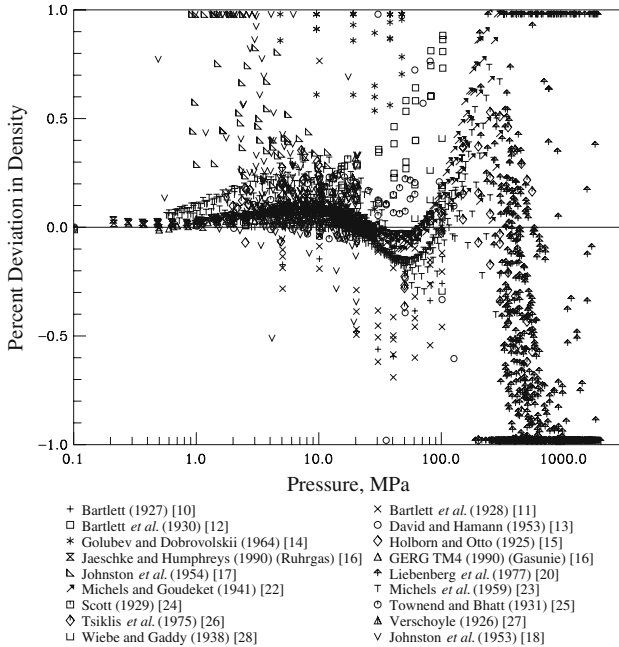


Fig. 6 Comparisons of density calculated by use of Younglove [1] with experimental normal hydrogen P- ρ -T data (% ΔX vs. Pressure)

of Table 2. The definitions for the statistical parameters are given below:

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

X is any property, n is the number of data points in the data set, and i is the index for each data point.

Comparisons were made to the NIST Standard Reference Database Program REFPROP [78], which implements the parahydrogen equation of state of Younglove [1] that uses the IPTS-68 temperature scale, and the upper limits in pressure and temperature are 121 MPa and 400 K. The equation of state for normal hydrogen uses the real fluid portion of the parahydrogen equation of state coupled with the ideal-gas heat-capacity equation for normal hydrogen. Graphical comparisons of density and speed of sound for normal hydrogen are given in Figs. 6 and 7. Those for parahydrogen are given in Figs. 8 and 9.

4.1 Normal Hydrogen

The normal hydrogen and parahydrogen models calculate the same vapor pressure values. The large AAD of calculated vapor pressure for normal hydrogen suggest that the differences in vapor pressures between normal hydrogen and parahydrogen

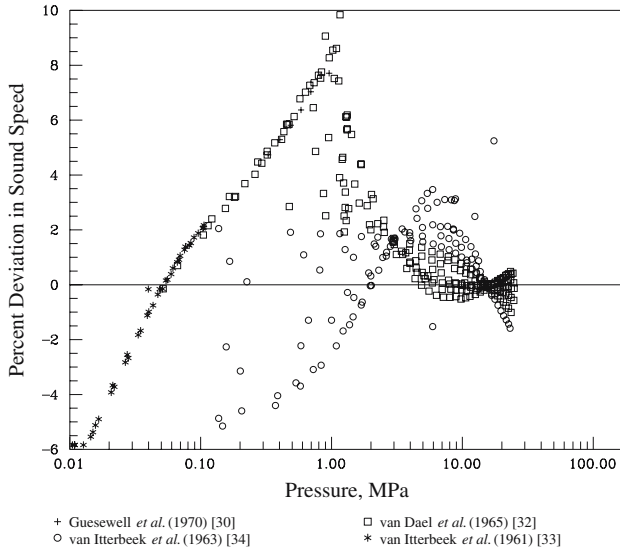


Fig. 7 Comparisons of speed of sound calculated by use of Younglove [1] with experimental normal hydrogen speed of sound data ($\% \Delta X$ vs. Pressure)

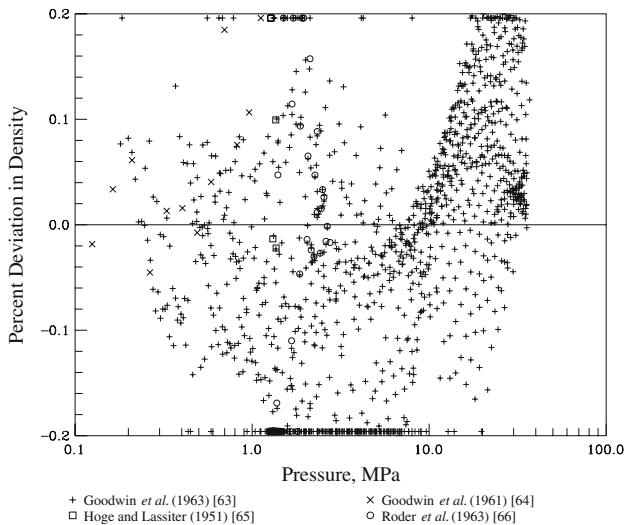


Fig. 8 Comparisons of density calculated by use of Younglove [1] with experimental parahydrogen P - ρ - T data ($\% \Delta X$ vs. Pressure)

should be factored into development of equations of state. There is one set of liquid-density measurements for normal hydrogen below 34 K. There are no vapor-density measurements below 34 K. These measurements are crucial in developing accurate equations of state. Additionally, substantial high accuracy measurements of the density in the critical region from 32 K to 50 K are vital in model development. Comparisons

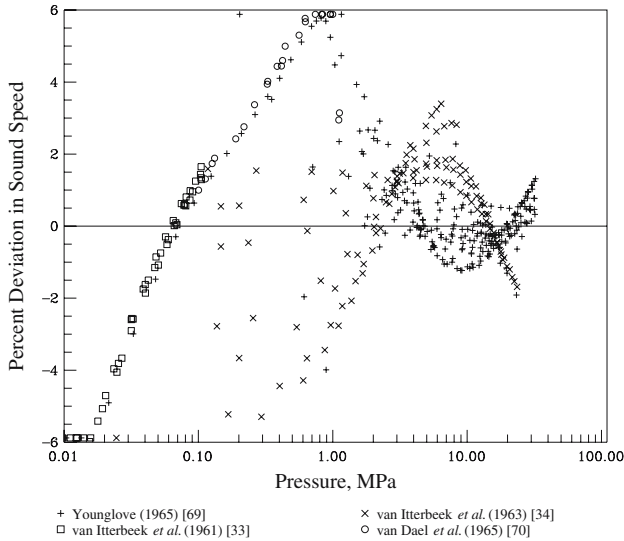


Fig. 9 Comparisons of speed of sound calculated by use of Younglove [1] with experimental parahydrogen speed of sound data ($\% \Delta X$ vs. Pressure)

from the equation to the only available data set in this region show a scatter of 2% or more in density. Additional measurements up to 250 K would resolve the discrepancy seen in the currently available measurements. Data above 400 K are scarce, and new measurements are needed at temperatures between 400 K and 1,200 K. Although the current model deviates significantly from the available density data between 250 K and 400 K, an updated equation of state will reduce these deviations to less than 0.05% in density over this temperature range. Additional sound-speed measurements are needed for vapor states at low pressures and moderate temperatures. There are insufficient caloric property data available for normal hydrogen. New measurement of caloric properties will ensure that the equation of state exhibits correct behavior.

4.2 Parahydrogen

Many of the experimental data needs for normal hydrogen also exist for parahydrogen. The few exceptions are: (a) measurements of the speed of sound are available from near the triple point in the liquid to 100 K, but none exist at higher temperatures; (b) heat capacities for the single phase and saturated liquid are available, which are fundamental to equation-of-state development; (c) a substantial amount of density data is available in the liquid phase, but the uncertainty in these data is higher than desired, especially in the critical region; and (d) density data above 100 K are not available. If necessary, a few high-accuracy data points above 100 K and at all pressures would allow the opportunity to transform the data for normal hydrogen to values that can be used in fitting the equation of state for parahydrogen.

5 Conclusions and Recommendations

In the years since the publication of the current standard formulation for hydrogen, there have been significant advances in both experimental measurements and in computer modeling.

The benefits of such advances suggest an improvement in the equations of state for hydrogen for use in engineering applications. There is a need for further measurements of thermodynamic equilibrium properties of hydrogen and for the revision of the thermodynamic property formulations used for engineering system design and analysis.

The temperature range should be increased consistent with proposed nuclear hydrogen generation processes (above 900 K), coal gasification processes (1,300 K), and revised to the ITS-90 temperature scale. The temperatures of calculated values should be updated to the ITS-90 temperature scale.

The standard formulation for normal hydrogen uses the parahydrogen equation of Younglove [1] in lieu of a separately published wide-range equation of state for normal hydrogen without introducing significant error above 250 K. The equation of state for parahydrogen has been considered accurate to about $\pm 0.2\%$ in density. The ideal-gas heat-capacity equation for parahydrogen in REFPROP [78] was modified because the original from Younglove [1] was a piecewise equation that would have required unique computer coding. Ideal-gas heat capacities for normal hydrogen were taken directly from McCarty et al. [3].

Recently, a truncated virial equation for use in fuel consumption applications was developed by Lemmon et al. [79]. This equation was based on the current parahydrogen equation of Younglove [1] and is limited to low-density vapor states. The density uncertainty for this equation is the same as that of Younglove [1], but it has fewer terms making it suitable for engineering calculations.

Hydrogen is most commonly used as a cryogenic liquid (99.75% parahydrogen) or as a compressed gas near 300 K (normal hydrogen). However, the liquefaction process involves orthohydrogen-to-parahydrogen conversion at intermediate temperatures near those of liquid nitrogen. To provide for engineering calculations of these mixture values, we have begun the formulation of an equation of state for mixtures of orthohydrogen and parahydrogen.

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