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Range 75-307 K and 2-20 kbar**

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THERMODYNAMIC PROPERTIES OF FLUID $n\text{-H}_2$ IN THE RANGE 75-307 K and 2-20 kbar

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ABSTRACT

An empirical equation of state (EOS) is developed which reproduces our simultaneous measurements of P , V , T , and sound velocity v_s in fluid $n\text{-H}_2$ from 75 to 307 K and 2 to 20 kbar. We then derive various thermodynamic properties from the EOS and give values in tabular form at rounded kbar pressures and 25 K temperature intervals over the experimental range. Included are the isobaric thermal expansion coefficient α_p , heat capacity at constant pressure C_p , isothermal compressibility coefficient χ_T , heat capacity ratio γ , and molar entropy S . Characteristics of the EOS at high pressure and temperature are examined and compared with several theoretical and phenomenological equations of state.

I. INTRODUCTION

Hydrogen isotopes play an important role in various energy technologies. Knowledge of the properties of hydrogen is basic to an understanding of the initiation and control of nuclear fusion where high densities and temperatures are required. In the fusion of small hydrogen targets by laser pulses, the energetics of hydrogen compression and heating must be accurately known since lasers of only marginal power are presently available. Chemical processes utilizing hydrogen, such as combustion in modified automobile engines and oxidation in fuel cells, have stimulated studies of a hydrogen economy that would involve production, storage, transmission, and distribution of the gas and liquid.

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Cooling of electrical transmission lines by slush hydrogen is a current application which draws on the properties of hydrogen in the limited region where measurements exist.

At sufficiently high pressure, hydrogen is expected to become a metal and possibly remain so when pressure is released. Although the generation of metallic hydrogen is a challenge to our present technology, a clear demonstration of its existence would contribute to our basic understanding of matter. The utility of the metal as a high-energy fuel and perhaps as a high-temperature superconductor has stirred new interest in hydrogen at superpressures. High-density hydrogen may exist in the planetary interiors of Jupiter and Saturn and accurate knowledge of the thermodynamics of hydrogen would improve astrophysical models of these planets.

In spite of considerable interest in the properties of hydrogen at high density, little experimental work has been done. In particular the equation of state, (the relationship between P , V , and T) is known in only a very limited region. While the low-temperature equation of state of solid hydrogen has been explored at pressures to 25 kbar,^{1,2} most studies of the gas near room temperature have been restricted to a few kilobars. A primary reason for this experimental limit is the deleterious effect that hydrogen has on many high-strength steels, especially at high temperatures. At room temperature there are accurate measurements by Michels³ up to 3 kbar, recent data by Tsiklis *et al.*⁴ to 6 kbar, and the crude pioneering measurements by Bridgman⁵ to 13 kbar.

At Los Alamos we have established a facility to make P - V - T studies at pressures above 3 kbar on various light molecules including the hydrogen isotopes. An apparatus and technique were developed for studies up to 21 kbar in the temperature range 75-320 K. Earlier measurements on argon⁶ and nitrogen^{7,8} were reported and recently we completed⁹ (Mills, Liebenberg, Bronson, and Schmidt hereafter called I) measurements of pressure, volume, temperature, and sound velocity on fluid normal hydrogen over the range of temperatures 75-307 K and pressures 2-20 kbar. In the present report we will review these measurements and the equation-of-state development based on our use of P - V - T and v_s data in a double-process least-squares fitting program that was used with all 1952 sets of data points. From the equation of state other thermodynamic properties may be derived and these will be presented in more detail than was possible in I. Comparison with other measurements was made in I, but recently Ashurst¹⁰ calculated properties based on an intermolecular potential model for H_2 and we

compare his results to ours. An earlier model for deuterium was given by G. Kerley¹¹ and, based on information he supplied recently for hydrogen, we make comparisons.

II. EXPERIMENTAL

The measurements were made in a thermostated piston-cylinder apparatus as described earlier.⁶ Bridgman unsupported-area closures were used to seal a movable piston and fixed plug in the belted, tungsten-carbide cylinder. The unsupported area at the fixed plug served as a mounting for an ultrasonic crystal used as transmitter and receiver. Pulses at ambient pressure were propagated through the plug, which served as a buffer rod, into the sample in a fixed-path cell and were reflected out again. At 30-MHz modulation frequency, no dispersion effects were detected in transmission through hydrogen. Volumes were determined from the piston position by a differential measurement using an external dial gauge. Force in the hydraulic press was generated with a rotating-piston dead-weight tester and pressures were calculated from the ratio of ram to cell area. Cell temperatures were computed from two external thermocouples calibrated in situ with an internal thermocouple at low pressure. Further details are available in I.

Errors in the measurements have been assessed in I. The corrected pressures are considered accurate to a few tenths of 1% at the lowest pressures and about 0.5% at the highest. Errors in volume average about 0.4% although some individual measurements, farthest in the P-T plane from normalization at 3 kbar and 295 K, could deviate by up to 1%. Temperatures at 75 K and ambient are known to ± 0.02 K but at intermediate temperatures the accuracy is estimated to be ± 0.5 K. Sound velocities were found to scatter from a smooth line by $\pm 0.5\%$, while measured timing accuracy and cell length errors would suggest an uncertainty of 0.4%.

A total of 1952 complete sets of $V(P,T)$ and corresponding $v_s(P,T)$ data points were obtained along 60 different isotherms in $n\text{-H}_2$ from 75 to 307 K and at pressures up to 20 kbar. These data sets are cataloged in a separate report for use as a data base in further equation of state development.¹² An additional number of separate $V(P,T)$ or $v_s(P,T)$ points were obtained and used as checks and in smoothing v_s data as discussed below.

We showed earlier for argon⁶ that the volume and sound velocity along a particular isotherm are well fitted at the higher pressures and densities by an equation of the form

$$V, v_s = A P^n, \quad (1)$$

where n is positive for the sound velocity and negative for the volume. The magnitude in each case is $n \approx 0.34$. Figures 1, 2, 3, and 4 show log-log plots of V, v_s data at two widely spaced temperatures. In order to generate a data set that was more manageable than the 1952 data points, we used Eq. (1) where the temperature dependence is introduced as $A = a_0 + a_1 T + a_2 T^2$, $n = n_0 + n_1 T + n_2 T^2$ to fit and smooth the $v_s(P, T)$ data. For the volumetric data a graphical linear interpolation of $\log V$ vs $\log P$ was used to obtain V in 1-kbar steps from 2 to 10 kbar and 2-kbar steps above 10 kbar. These interpolated points were plotted as 14 isobars on a $\log V$ vs T plot and smooth curves were drawn through the points. The average deviation of points from the smooth curves was less than $\pm 0.5\%$. Data to form a 14×10 matrix in P by T were obtained for both V

Fig. 1.

Plot of $\log v_s$ vs $\log P$ in fluid $n\text{-H}_2$ of 294 K. Symbols indicate different runs carried out over a period of a year. Circle, July 1976; triangle, May 1975; plus, June 1975. The smooth line is from a least-squares fit.

Fig. 2.
Plot of $\log V$ vs $\log P$ in fluid $n\text{-H}_2$ at 294 K.

Fig. 3.
Plot of $\log v_s$ vs $\log P$ in fluid $n\text{-H}_2$ at 146 K.

Fig. 4.
Plot of $\log V$ vs $\log P$ in fluid $n\text{-H}_2$ at 146 K.

and v_g at 25-K intervals. The actual number of points was 114 rather than 140 because the freezing curve truncates the fluid domain at temperatures less than 170 K.

III. EQUATION-OF-STATE DEVELOPMENT

For ease of manipulation we have considered an equation of state (EOS) of the form,

$$V = f(P, T). \quad (2)$$

Many types of closed-form expressions for the EOS of fluids have been given.^{13,14} Benedict¹⁵ found that an equation of the form

$$V = \sum_{m=1}^3 \sum_{n=-2}^2 A_{nm} T^{n/2} P^{-m/3} \quad (3)$$

could be simplified, when highly correlated terms were eliminated, to give the following equation which he used for nitrogen:

$$V = (A+BT+CT^{-1/2})P^{-1/3} + (D+ET)P^{-2/3} + (F+GT+HT^{-1/2} + JT^{-1})P^{-1} \quad (4)$$

We used this equation for our hydrogen data, all of which were obtained well into the supercritical fluid region.¹⁶ The equation has a form that is amenable to use with the double-process least-squares fitting method.

To use P - V - T and v_s data together in a double-process least-squares fitting process, we write the thermodynamic relation

$$\frac{1}{v_s} = \left[\left(\frac{\partial p}{\partial P} \right)_s \right]^{1/2} = \frac{M}{V}^{1/2} \left\{ - \left(\frac{\partial v}{\partial P} \right)_T - \frac{T \left[\left(\frac{\partial v}{\partial T} \right)_P \right]^2}{C_p} \right\}^{1/2} \equiv g(P, T) \quad (5)$$

and, as discussed in I, minimize the sum S for n sets of points in the following equation:

$$S = \sum_n [v_{\text{exp}} - f(P, T)]^2 + \sum_n \left[\frac{1}{v_s} - g(P, T) \right]^2. \quad (6)$$

In addition to our own data we need the value of C_p at our normalization pressure over the full temperature range. The lower pressure data of Michels³ and recently tabulated Russian values¹⁷ were used to obtain $C_{p_0}(T)$ at our lowest pressure $P=2$ kbar over the 75-307 K temperature range. Then the heat capacity

$$C_p = C_{p_0}(T) - T \int_{P_0}^P \left(\frac{\partial^2 v}{\partial T^2} \right)_P dP \quad (7)$$

can be directly calculated for use in Eqs. (5) and (6). The expression is

$$C_{p_0}(T) = 1.1230T - 61.468T^{1/2} + 1259.3 \\ - 10512T^{-1/2} + 31638T^{-1} \text{ [J/mole-K]}. \quad (8)$$

From the smooth data set of 114 points we obtain values of the constants in Eq. (4) as shown in Table I. Shown for comparison are the values of the constants when all 1952 data points are used. In this latter case no attempt was made to

weight the points, although near 295 K there were about 500 data sets. We regard the smooth data set as more representative of the fit over the full temperature range. In view of the fewer points between 15-20 kbar, the smooth-data-set constants should be more appropriate for extrapolating Eq. (4) to high pressure. The standard error of estimate

$$S.E.E. = \left(\frac{\sum_{i=1}^N (X_{cal} - X_{exp})^2}{N} \right)^{1/2}, \quad (9)$$

where $X=V$ or v_s is given in Table I for each set of constants against the full 1952-point data set.¹⁸ There is little difference in the fit of these two sets of constants with the full data set although the S.E.E. is somewhat better, as expected, when the 1952-point constants are used with the full data set.

TABLE I

Constants of Eq. (4) as determined by a double-process least-square fit to the smooth-data set and the 1952-point data set.

	<u>Smoothed Data</u>		<u>1952-Point</u>
A	36.716		36.141
B	0.0033003		0.0006014
C	-22.479		-3.7094
D	-17.174		-21.952
E	-0.021393		-0.0023698
F	-8.9886		1.0750
G	0.11001		0.086622
H	69.233		-9.8085
J	-31.395		175.36
	<u>V</u>	<u>v_s</u>	<u>V</u> <u>v_s</u>
Std error of estimate	0.092	0.055	0.090 0.055

In the course of developing this equation of state for H_2 , other expressions were tried, such as the form we used earlier to fit nitrogen data over a more limited temperature range. We also determined that fitting only P-V-T data gave an EOS from which sound velocities could not be obtained to better than a few percent of measured values. Thus, the measurements of P, V, T, and v_s , together with the fitting procedure, have produced an EOS in a previously unexplored region of the P, T plane with an accuracy that we believe exceeds that of work done at lower pressures, insofar as deriving other thermodynamic quantities.

IV. DERIVED THERMODYNAMIC VALUES

We used standard thermodynamic relations and our smooth-data EOS of Eq. (4) to obtain the various thermodynamic quantities shown in Table II. The isobaric expansion coefficient is,

$$\alpha_p = \frac{1}{V} \left(\frac{\partial V}{\partial T} \right)_p. \quad (10)$$

The heat capacity at constant pressure is C_p , determined from Eq. (7) where $C_{p0}(T)$ is given by our fit to the low-pressure data expressed in Eq. (8). The isothermal compressibility is given by

$$\chi_T = - \frac{1}{V} \left(\frac{\partial V}{\partial P} \right)_T \quad (11)$$

and the specific heat ratio by

$$\gamma \equiv C_p/C_v = \chi_T/\chi_s. \quad (12)$$

However χ_s , the adiabatic compressibility, may be determined directly from the measurements

$$\chi_s = \frac{1}{\rho v_s^2}. \quad (13)$$

The value of γ can be conveniently used to obtain either C_v , the heat capacity at constant volume, or χ_s from Table II.

The molar entropy is obtained from the relation

$$S(P_0, T) - S(P, T) = \int_{P_0}^P \left(\frac{\partial V}{\partial T} \right)_P dP \quad (14)$$

A value of entropy $S(P_0, T)$ is obtained from the tables presented by Vargaftik.¹⁷ These extend to 1 kbar and were used with Eq. (14) to obtain values at 2 kbar. These in turn were fitted to the equation,

$$S(P_1, T) = 12.194 + 0.27442 T - 0.00030914 T^2 \text{ J/mole-K}, \quad (15)$$

which gives an average deviation from the Vargaftik values at 1 kbar of 0.10 J/mole-K or about 0.23%. This equation can not be extrapolated to high temperatures since the T^2 term completely dominates at 900 K. A simple power law fit to the data,

$$S(P_1, T) = -105.06 + 30.96 \ln (T + 13.0) \text{ J/mole-K} \quad (16)$$

yields entropies to better than 1% and extrapolates such that at 500 K the entropy is less than 1% from the Vargaftik value. Thus, Eq. (16) should be used when extrapolating our EOS.

The equations have been used in a FORTRAN program given in the Appendix to obtain the tabular listing of properties in Table II. These same values are graphically presented in Figs. 5-10, some of which duplicate the figures in I. The curves in these figures are drawn directly by a computer program and the special font is selected to indicate that these curves all refer to values calculated from our EOS.

V. COMPARISONS

We have discussed in I several direct comparisons between our data and those measured by others. Primarily, the molar-volume data of Michels *et al.*³ and Tsiklis⁴ are in excellent agreement, i.e., within less than 0.5% of our values. Unfortunately no measurements of sound velocity are available for comparison.

TABLE II

HYDROGEN E O S OF MILLS, LIEBENBERG, BROWNSON, SCHMIDT T = 75.0 K												HYDROGEN E O S OF MILLS, LIEBENBERG, BROWNSON, SCHMIDT T = 150.0 K											
PRESS.	VOL.	V(S)	ALPHA	C(P)	GAMMA	BETA(S)	ENTROPY	PRESS.	VOL.	V(S)	ALPHA	C(P)	GAMMA	BETA(S)	ENTROPY								
KBAR	CC/MOLE	KM/S	/K	J/MOLE-K		/KBAR	J/MOLE-K	KBAR	CC/MOLE	KM/S	/K	J/MOLE-K		/KBAR	J/MOLE-K								
2.00	18.870	3.079	1.801E-03	19.215	1.242	9.872E-02	31.04	2.00	21.717	2.913	1.854E-03	27.543	1.320	1.269E-01	44.40								
2.50	17.865	3.350	1.593E-03	18.669	1.228	7.989E-02	29.49	2.50	20.183	3.160	1.603E-03	27.324	1.284	1.003E-01	43.12								
3.00	17.063	3.546	1.449E-03	18.339	1.217	6.741E-02	28.16	3.00	19.049	3.372	1.422E-03	27.187	1.256	8.307E-02	41.87								
3.50	16.445	3.735	1.342E-03	18.153	1.209	5.847E-02	26.99	3.50	18.150	3.500	1.287E-03	27.103	1.234	7.107E-02	40.77								
4.00	15.909	3.906	1.260E-03	18.069	1.203	5.173E-02	25.94	4.00	17.436	3.730	1.180E-03	27.058	1.217	6.217E-02	39.80								
4.50	15.448	4.061	1.195E-03	18.058	1.197	4.646E-02	24.98	4.50	16.829	3.884	1.095E-03	27.041	1.202	5.532E-02	38.92								
HYDROGEN E O S OF MILLS, LIEBENBERG, BROWNSON, SCHMIDT T = 100.0 K																							
2.00	18.605	3.057	1.888E-03	22.100	1.304	1.049E-01	36.54	2.00	21.717	2.913	1.854E-03	27.543	1.320	1.269E-01	44.40								
2.50	18.605	3.302	1.639E-03	21.724	1.272	8.463E-02	34.86	2.50	20.183	3.160	1.603E-03	27.324	1.284	1.003E-01	43.12								
3.00	17.718	3.512	1.465E-03	21.492	1.248	7.125E-02	33.46	3.00	19.049	3.372	1.422E-03	27.187	1.256	8.307E-02	41.87								
3.50	17.006	3.697	1.334E-03	21.356	1.230	6.170E-02	32.24	3.50	18.150	3.500	1.287E-03	27.103	1.234	7.107E-02	40.77								
4.00	16.413	3.864	1.234E-03	21.289	1.216	5.453E-02	31.17	4.00	17.436	3.730	1.180E-03	27.058	1.217	6.217E-02	39.80								
4.50	15.909	4.016	1.157E-03	21.271	1.205	4.892E-02	30.21	4.50	16.829	3.884	1.095E-03	27.041	1.202	5.532E-02	38.92								
5.00	15.470	4.157	1.092E-03	21.290	1.195	4.441E-02	29.35	5.00	16.309	4.027	1.024E-03	27.045	1.190	4.988E-02	38.12								
5.50	15.084	4.288	1.037E-03	21.340	1.187	4.070E-02	28.51	5.50	15.855	4.181	9.648E-04	27.066	1.180	4.543E-02	37.19								
6.00	14.739	4.410	9.910E-04	21.413	1.180	3.759E-02	27.76	6.00	15.455	4.285	9.142E-04	27.100	1.171	4.174E-02	36.12								
6.50	14.428	4.526	9.513E-04	21.504	1.174	3.493E-02	27.05	6.50	15.097	4.403	8.705E-04	27.145	1.164	3.843E-02	35.71								
7.00	14.145	4.636	9.148E-04	21.611	1.169	3.265E-02	26.38	7.00	14.774	4.515	8.348E-04	27.199	1.157	3.595E-02	35.07								
7.50	13.887	4.741	8.805E-04	21.732	1.164	3.063E-02	25.75	7.50	14.481	4.621	7.989E-04	27.260	1.151	3.364E-02	34.47								
HYDROGEN E O S OF MILLS, LIEBENBERG, BROWNSON, SCHMIDT T = 125.0 K																							
2.00	19.766	3.057	1.888E-03	22.100	1.304	1.049E-01	36.54	2.00	21.717	2.913	1.854E-03	27.543	1.320	1.269E-01	44.40								
2.50	18.605	3.302	1.639E-03	21.724	1.272	8.463E-02	34.86	2.50	20.183	3.160	1.603E-03	27.324	1.284	1.003E-01	43.12								
3.00	17.718	3.512	1.465E-03	21.492	1.248	7.125E-02	33.46	3.00	19.049	3.372	1.422E-03	27.187	1.256	8.307E-02	41.87								
3.50	17.006	3.697	1.334E-03	21.356	1.230	6.170E-02	32.24	3.50	18.150	3.500	1.287E-03	27.103	1.234	7.107E-02	40.77								
4.00	16.413	3.864	1.234E-03	21.289	1.216	5.453E-02	31.17	4.00	17.436	3.730	1.180E-03	27.058	1.217	6.217E-02	39.80								
4.50	15.909	4.016	1.157E-03	21.271	1.205	4.892E-02	30.21	4.50	16.829	3.884	1.095E-03	27.041	1.202	5.532E-02	38.92								
5.00	15.470	4.157	1.092E-03	21.290	1.195	4.441E-02	29.35	5.00	16.309	4.027	1.024E-03	27.045	1.190	4.988E-02	38.12								
5.50	15.084	4.288	1.037E-03	21.340	1.187	4.070E-02	28.51	5.50	15.855	4.181	9.648E-04	27.066	1.180	4.543E-02	37.12								
6.00	14.739	4.410	9.910E-04	21.413	1.180	3.759E-02	27.76	6.00	15.455	4.285	9.142E-04	27.100	1.171	4.174E-02	36.12								
6.50	14.428	4.526	9.513E-04	21.504	1.174	3.493E-02	27.05	6.50	15.097	4.403	8.705E-04	27.145	1.164	3.843E-02	35.71								
7.00	14.145	4.636	9.148E-04	21.611	1.169	3.265E-02	26.38	7.00	14.774	4.515	8.348E-04	27.199	1.157	3.595E-02	35.07								
7.50	13.887	4.741	8.805E-04	21.732	1.164	3.063E-02	25.75	7.50	14.481	4.621	7.989E-04	27.260	1.151	3.364E-02	34.47								
HYDROGEN E O S OF MILLS, LIEBENBERG, BROWNSON, SCHMIDT T = 150.0 K																							
2.00	20.723	2.980	1.889E-03	25.324	1.315	1.157E-01	41.67	2.00	21.717	2.913	1.854E-03	27.543	1.320	1.269E-01	44.40								
2.50	19.383	3.226	1.632E-03	25.044	1.279	9.236E-02	39.91	2.50	20.183	3.160	1.603E-03	27.324	1.284	1.003E-01	43.12								
3.00	18.377	3.437	1.451E-03	24.870	1.252	7.714E-02	38.46	3.00	19.049	3.372	1.422E-03	27.187	1.256	8.307E-02	41.87								
3.50	17.579	3.624	1.315E-03	24.766	1.231	6.640E-02	37.22	3.50	18.150	3.500	1.287E-03	27.103	1.234	7.107E-02	40.77								
4.00	16.923	3.792	1.209E-03	24.711	1.214	5.839E-02	36.14	4.00	17.436	3.730	1.180E-03	27.058	1.217	6.217E-02	39.80								
4.50	16.369	3.945	1.124E-03	24.693	1.201	5.218E-02	35.17	4.50	16.829	3.884	1.095E-03	27.041	1.202	5.532E-02	38.92								
5.00	15.891	4.086	1.055E-03	24.702	1.190	4.721E-02	34.29	5.00	16.309	4.027	1.024E-03	27.045	1.190	4.988E-02	38.12								
5.50	15.472	4.218	9.966E-04	24.733	1.180	4.314E-02	33.48	5.50	15.855	4.181	9.648E-04	27.066	1.180	4.543E-02	37.12								
6.00	15.100	4.341	9.471E-04	24.781	1.172	3.974E-02	32.74	6.00	14.766	4.458	9.045E-04	27.842	1.165	3.685E-02	32.05								
6.50	14.766	4.458	9.045E-04	24.842	1.165	3.685E-02	32.05	6.50	14.464	4.569	8.673E-04	24.916	1.159	3.437E-02	31.40								
7.00	14.464	4.569	8.673E-04	24.916	1.159	3.437E-02	31.40	7.00	14.188	4.674	8.346E-04	24.998	1.153	3.222E-02	30.80								
7.50	14.188	4.674	8.346E-04	24.998	1.153	3.222E-02	30.80	7.50	13.935	4.774	8.054E-04	25.080	1.149	3.033E-02	30.22								
8.00	13.935	4.774	8.054E-04	25.080	1.149	3.033E-02	30.22	8.00	13.702	4.870	7.797E-04	25.166	1.144	2.865E-02	29.67								
8.50	13.702	4.870	7.797E-04	25.166	1.144	2.865E-02	29.67	8.50	13.485	4.963	7.564E-04	25.260	1.140	2.716E-02	29.15								
9.00	13.485	4.963	7.564E-04	25.260	1.140	2.716E-02	29.15	9.00	13.284	5.052	7.354E-04	25.367	1.137	2.582E-02	28.65								
9.50	13.284	5.052	7.354E-04	25.367	1.137	2.582E-02	28.65	9.50	13.095	5.138	7.162E-04	25.510	1.134	2.461E-02	28.17								
10.00	13.095	5.138	7.162E-04	25.510	1.134	2.461E-02	28.17	10.00	12.918	5.221	6.987E-04	25.625	1.131	2.351E-02	27.71								
10.50	12.918	5.221	6.987E-04	25.625	1.131	2.351E-02	27.71	10.50	12.751	5.301	6.827E-04	25.744	1.128	2.251E-02	27.27								
11.00	12.751	5.301	6.827E-04	25.744	1.128	2.251E-02	27.27	11.00	12.594	5.379	6.679E-04	25.866	1.126	2.159E-02	26.84								

TABLE II (contd)

HYDROGEN E O S OF MILLS, LIEBENBERG, BRUNSON, SCHWIDT															
T = 175.0 K								T = 200.0 K							
PRESS.	VOL.	V(S)	ALPHA	C(P)	GAMMA	DELTA(S)	ENTROPY	PRESS.	VOL.	V(S)	ALPHA	C(P)	GAMMA	DELTA(S)	ENTROPY
KBAR	CC/MOLE	KW/S	/K	J/MOLE-K	/KBAR	J/MOLE-K	J/MOLE-K	KBAR	CC/MOLE	KW/S	/K	J/MOLE-K	/KBAR	J/MOLE-K	J/MOLE-K
2.00	22.734	2.865	1.805E-03	28.836	1.327	1.374E-01	50.75	2.00	23.767	2.831	1.749E-03	29.491	1.335	1.471E-01	54.71
2.50	20.998	3.110	1.563E-03	28.557	1.291	1.077E-01	48.92	2.50	21.823	3.073	1.520E-03	29.343	1.300	1.146E-01	52.86
3.00	19.730	3.322	1.388E-03	28.478	1.263	8.848E-02	47.42	3.00	20.418	3.285	1.352E-03	29.250	1.272	9.387E-02	51.35
3.50	18.747	3.511	1.256E-03	28.440	1.241	7.545E-02	46.15	3.50	19.337	3.473	1.224E-03	29.192	1.250	7.952E-02	50.07
4.00	17.952	3.681	1.151E-03	28.440	1.223	6.572E-02	45.08	4.00	18.469	3.643	1.122E-03	29.159	1.231	6.901E-02	48.97
4.50	17.290	3.837	1.067E-03	28.425	1.208	5.826E-02	44.08	4.50	17.751	3.800	1.040E-03	29.145	1.216	6.099E-02	47.99
5.00	16.726	3.981	9.965E-04	28.426	1.195	5.235E-02	43.20	5.00	17.142	3.944	9.711E-04	29.166	1.203	5.466E-02	47.11
5.50	16.237	4.115	9.375E-04	28.441	1.185	4.756E-02	42.40	5.50	16.617	4.079	9.131E-04	29.157	1.192	4.954E-02	46.32
6.00	15.807	4.241	8.872E-04	28.467	1.175	4.359E-02	41.67	6.00	16.156	4.206	8.634E-04	29.177	1.182	4.531E-02	45.56
6.50	15.424	4.360	8.434E-04	28.501	1.167	4.025E-02	41.00	6.50	15.743	4.325	8.202E-04	29.204	1.174	4.174E-02	44.92
7.00	15.080	4.472	8.052E-04	28.543	1.160	3.740E-02	40.37	7.00	15.382	4.458	7.824E-04	29.237	1.166	3.873E-02	44.30
7.50	14.768	4.579	7.715E-04	28.590	1.154	3.493E-02	39.78	7.50	15.051	4.546	7.490E-04	29.275	1.160	3.612E-02	43.71
8.00	14.483	4.682	7.415E-04	28.643	1.148	3.278E-02	39.23	8.00	14.749	4.649	7.192E-04	29.318	1.154	3.385E-02	43.17
8.50	14.221	4.780	7.146E-04	28.699	1.143	3.088E-02	38.71	8.50	14.473	4.748	6.924E-04	29.363	1.148	3.165E-02	42.65
9.00	13.980	4.874	6.901E-04	28.759	1.139	2.919E-02	38.21	9.00	14.219	4.842	6.683E-04	29.412	1.144	2.950E-02	42.16
9.50	13.756	4.964	6.684E-04	28.822	1.135	2.769E-02	37.74	9.50	13.983	4.933	6.464E-04	29.463	1.139	2.808E-02	41.70
10.00	13.547	5.052	6.484E-04	28.880	1.131	2.633E-02	37.30	10.00	13.764	5.021	6.264E-04	29.516	1.135	2.708E-02	41.26
10.50	13.352	5.136	6.301E-04	28.936	1.128	2.511E-02	36.86	10.50	13.568	5.106	6.081E-04	29.571	1.131	2.580E-02	40.84
11.00	13.168	5.218	6.131E-04	29.002	1.122	2.399E-02	36.45	11.00	13.388	5.188	5.913E-04	29.628	1.128	2.463E-02	40.43
11.50	12.996	5.297	5.977E-04	29.078	1.119	2.298E-02	36.05	11.50	13.215	5.265	5.750E-04	29.687	1.125	2.357E-02	40.05
12.00	12.833	5.376	5.834E-04	29.171	1.116	2.199E-02	35.67	12.00	13.057	5.345	5.600E-04	29.746	1.122	2.260E-02	39.67
12.50	12.679	5.448	5.701E-04	29.246	1.114	2.104E-02	35.31	12.50	12.905	5.421	5.460E-04	29.807	1.119	2.171E-02	39.32
13.00	12.533	5.521	5.577E-04	29.322	1.112	2.016E-02	34.95	13.00	12.765	5.494	5.354E-04	29.868	1.117	2.085E-02	38.97
13.50	12.394	5.592	5.461E-04	29.399	1.110	1.946E-02	34.61	13.50	12.631	5.565	5.240E-04	29.931	1.115	2.001E-02	38.64
14.00	12.262	5.661	5.353E-04	29.477	1.108	1.880E-02	34.27	14.00	12.423	5.634	5.131E-04	29.994	1.112	1.941E-02	38.31
14.50	12.136	5.728	5.251E-04	29.555	1.106	1.815E-02	33.95	14.50	12.292	5.702	5.029E-04	30.058	1.110	1.875E-02	38.00
15.00	12.015	5.794	5.154E-04	29.635	1.104	1.755E-02	33.64	15.00	12.167	5.768	4.933E-04	30.123	1.108	1.814E-02	37.69
15.50	11.900	5.858	5.066E-04	29.716	1.103	1.700E-02	33.33	15.50	12.048	5.833	4.843E-04	30.188	1.107	1.754E-02	37.40
16.00	11.789	5.921	4.981E-04	29.795	1.102	1.648E-02	33.03	16.00	11.933	5.897	4.757E-04	30.253	1.105	1.702E-02	37.11
16.50	11.680	5.983	4.901E-04	29.876	1.100	1.599E-02	32.74	16.50	11.823	5.959	4.672E-04	30.319	1.103	1.652E-02	36.83
17.00	11.580	6.043	4.825E-04	29.957	1.099	1.552E-02	32.46	17.00	11.717	6.019	4.588E-04	30.386	1.102	1.604E-02	36.56
17.50	11.482	6.103	4.753E-04	30.039	1.099	1.529E-02	32.18	17.50	11.616	6.079	4.508E-04	30.452	1.100	1.559E-02	36.29
18.00	11.387	6.163	4.685E-04	30.121	1.096	1.488E-02	31.91	18.00	11.518	6.137	4.439E-04	30.519	1.099	1.517E-02	36.03
18.50	11.296	6.218	4.620E-04	30.204	1.096	1.449E-02	31.65	18.50	11.424	6.195	4.373E-04	30.586	1.098	1.477E-02	35.78
19.00	11.208	6.274	4.558E-04	30.286	1.095	1.412E-02	31.39	19.00	11.333	6.251	4.312E-04	30.654	1.096	1.439E-02	35.53
19.50	11.123	6.329	4.499E-04	30.369	1.094	1.377E-02	31.14	19.50	11.245	6.306	4.252E-04	30.721	1.095	1.402E-02	35.29
20.00	11.040	6.383	4.443E-04	30.452	1.093	1.344E-02	30.89	20.00	11.160	6.361	4.213E-04	30.789	1.094	1.368E-02	35.04
20.50	10.960	6.436	4.399E-04	30.535	1.092	1.312E-02	30.65	20.50	11.078	6.414	4.169E-04	30.857	1.093	1.334E-02	34.82
								21.00	10.998	6.467	4.109E-04	30.925	1.092	1.304E-02	34.59
								21.50	10.921	6.519	4.059E-04	30.993	1.091	1.275E-02	34.36
								22.00	10.846	6.570	4.011E-04	31.061	1.090	1.247E-02	34.14
								22.50	10.774	6.620	3.966E-04	31.129	1.089	1.219E-02	33.93

TABLE II (contd)

HYDROGEN E O S OF MILLS, LIEBERBERG, BROWSON, SCHWIDT T = 225.0 K				HYDROGEN E O S OF MILLS, LIEBERBERG, BROWSON, SCHWIDT T = 250.0 K			
PRESS.	VOL.	V(S)	ALPHA	C(P)	GAMMA	BETA(S)	ENTROPY
KBAR	CC/MOLE	KW/S	/K	J/MOLE-K		/KBAR	J/MOLE-K
2.00	24.811	2.809	1.692E-03	29.748	1.344	1.559E-01	58.29
3.50	22.656	3.048	1.475E-03	29.643	1.309	1.210E-01	54.42
5.00	21.110	3.247	1.315E-03	29.563	1.282	9.870E-02	54.90
7.50	19.950	3.444	1.192E-03	29.516	1.259	8.333E-02	53.62
10.00	18.988	3.616	1.094E-03	29.485	1.241	7.210E-02	52.51
12.50	18.213	3.770	1.014E-03	29.473	1.225	6.355E-02	51.53
15.00	17.558	3.915	9.474E-04	29.472	1.212	5.683E-02	50.65
17.50	16.996	4.050	8.908E-04	29.481	1.200	5.140E-02	49.86
20.00	16.504	4.177	8.421E-04	29.497	1.190	4.693E-02	49.13
22.50	16.070	4.297	7.998E-04	29.520	1.181	4.318E-02	48.46
25.00	15.682	4.410	7.626E-04	29.547	1.174	3.999E-02	47.84
27.50	15.331	4.519	7.296E-04	29.578	1.167	3.725E-02	47.27
30.00	15.013	4.622	7.002E-04	29.613	1.160	3.484E-02	46.72
32.50	14.723	4.721	6.738E-04	29.651	1.155	3.277E-02	46.21
35.00	14.455	4.816	6.500E-04	29.691	1.150	3.091E-02	45.73
37.50	14.208	4.908	6.283E-04	29.734	1.145	2.926E-02	45.27
40.00	13.978	5.001	6.085E-04	29.778	1.141	2.778E-02	44.84
42.50	13.764	5.091	5.903E-04	29.824	1.137	2.644E-02	44.42
45.00	13.564	5.164	5.736E-04	29.871	1.133	2.523E-02	44.02
47.50	13.376	5.244	5.582E-04	29.920	1.130	2.413E-02	43.64
50.00	13.199	5.322	5.439E-04	29.969	1.127	2.312E-02	43.28
52.50	13.032	5.397	5.306E-04	30.020	1.124	2.219E-02	42.93
55.00	12.874	5.471	5.182E-04	30.071	1.121	2.134E-02	42.59
57.50	12.724	5.542	5.066E-04	30.123	1.119	2.055E-02	42.26
60.00	12.581	5.612	4.958E-04	30.176	1.116	1.981E-02	41.94
62.50	12.445	5.680	4.856E-04	30.229	1.114	1.913E-02	41.63
65.00	12.316	5.747	4.760E-04	30.283	1.112	1.850E-02	41.34
67.50	12.192	5.812	4.670E-04	30.337	1.110	1.790E-02	41.05
70.00	12.073	5.876	4.585E-04	30.392	1.108	1.735E-02	40.77
72.50	11.959	5.938	4.504E-04	30.447	1.107	1.682E-02	40.49
75.00	11.850	5.999	4.428E-04	30.503	1.105	1.633E-02	40.23
77.50	11.746	6.059	4.355E-04	30.558	1.103	1.587E-02	39.97
80.00	11.645	6.118	4.286E-04	30.614	1.102	1.543E-02	39.72
82.50	11.547	6.176	4.221E-04	30.670	1.100	1.502E-02	39.47
85.00	11.454	6.232	4.158E-04	30.726	1.099	1.463E-02	39.23
87.50	11.363	6.286	4.099E-04	30.783	1.098	1.426E-02	38.99
90.00	11.276	6.343	4.042E-04	30.839	1.097	1.390E-02	38.76
92.50	11.191	6.396	3.988E-04	30.896	1.096	1.357E-02	38.54
95.00	11.109	6.449	3.936E-04	30.953	1.094	1.325E-02	38.32
97.50	11.030	6.501	3.886E-04	31.010	1.093	1.294E-02	38.10
100.00	10.953	6.552	3.838E-04	31.067	1.092	1.265E-02	37.89
102.50	10.879	6.603	3.792E-04	31.123	1.091	1.238E-02	37.68
105.00	10.806	6.653	3.748E-04	31.179	1.090	1.213E-02	37.47
107.50	10.734	6.702	3.705E-04	31.235	1.089	1.189E-02	37.26
110.00	10.662	6.750	3.663E-04	31.291	1.088	1.166E-02	37.05
112.50	10.591	6.797	3.622E-04	31.347	1.087	1.144E-02	36.84
115.00	10.520	6.844	3.582E-04	31.403	1.086	1.122E-02	36.63
117.50	10.450	6.890	3.543E-04	31.459	1.085	1.101E-02	36.42
120.00	10.380	6.936	3.505E-04	31.515	1.084	1.080E-02	36.21
122.50	10.311	6.981	3.468E-04	31.571	1.083	1.060E-02	36.00
125.00	10.242	7.026	3.432E-04	31.627	1.082	1.040E-02	35.79
127.50	10.174	7.070	3.397E-04	31.683	1.081	1.020E-02	35.58
130.00	10.106	7.114	3.363E-04	31.739	1.080	1.000E-02	35.37
132.50	10.039	7.157	3.330E-04	31.795	1.079	9.800E-03	35.16
135.00	9.972	7.200	3.298E-04	31.851	1.078	9.600E-03	34.95
137.50	9.906	7.242	3.267E-04	31.907	1.077	9.400E-03	34.74
140.00	9.840	7.284	3.237E-04	31.963	1.076	9.200E-03	34.53
142.50	9.775	7.325	3.208E-04	32.019	1.075	9.000E-03	34.32
145.00	9.710	7.366	3.180E-04	32.075	1.074	8.800E-03	34.11
147.50	9.645	7.406	3.153E-04	32.131	1.073	8.600E-03	33.90
150.00	9.580	7.446	3.127E-04	32.187	1.072	8.400E-03	33.69
152.50	9.515	7.485	3.102E-04	32.243	1.071	8.200E-03	33.48
155.00	9.450	7.524	3.078E-04	32.299	1.070	8.000E-03	33.27
157.50	9.385	7.562	3.055E-04	32.355	1.069	7.800E-03	33.06
160.00	9.320	7.600	3.033E-04	32.411	1.068	7.600E-03	32.85
162.50	9.255	7.637	3.012E-04	32.467	1.067	7.400E-03	32.64
165.00	9.190	7.674	2.992E-04	32.523	1.066	7.200E-03	32.43
167.50	9.125	7.710	2.973E-04	32.579	1.065	7.000E-03	32.22
170.00	9.060	7.746	2.955E-04	32.635	1.064	6.800E-03	32.01
172.50	9.000	7.781	2.938E-04	32.691	1.063	6.600E-03	31.80
175.00	8.940	7.816	2.922E-04	32.747	1.062	6.400E-03	31.59
177.50	8.880	7.850	2.907E-04	32.803	1.061	6.200E-03	31.38
180.00	8.820	7.884	2.893E-04	32.859	1.060	6.000E-03	31.17
182.50	8.760	7.917	2.880E-04	32.915	1.059	5.800E-03	30.96
185.00	8.700	7.950	2.868E-04	32.971	1.058	5.600E-03	30.75
187.50	8.640	7.982	2.857E-04	33.027	1.057	5.400E-03	30.54
190.00	8.580	8.014	2.847E-04	33.083	1.056	5.200E-03	30.33
192.50	8.520	8.046	2.838E-04	33.139	1.055	5.000E-03	30.12
195.00	8.460	8.077	2.830E-04	33.195	1.054	4.800E-03	29.91
197.50	8.400	8.108	2.822E-04	33.251	1.053	4.600E-03	29.70
200.00	8.340	8.138	2.815E-04	33.307	1.052	4.400E-03	29.49
202.50	8.280	8.168	2.808E-04	33.363	1.051	4.200E-03	29.28
205.00	8.220	8.197	2.802E-04	33.419	1.050	4.000E-03	29.07
207.50	8.160	8.226	2.797E-04	33.475	1.049	3.800E-03	28.86
210.00	8.100	8.254	2.792E-04	33.531	1.048	3.600E-03	28.65
212.50	8.040	8.282	2.788E-04	33.587	1.047	3.400E-03	28.44
215.00	7.980	8.309	2.784E-04	33.643	1.046	3.200E-03	28.23
217.50	7.920	8.336	2.781E-04	33.699	1.045	3.000E-03	28.02
220.00	7.860	8.362	2.778E-04	33.755	1.044	2.800E-03	27.81
222.50	7.800	8.388	2.776E-04	33.811	1.043	2.600E-03	27.60
225.00	7.740	8.413	2.774E-04	33.867	1.042	2.400E-03	27.39
227.50	7.680	8.438	2.772E-04	33.923	1.041	2.200E-03	27.18
230.00	7.620	8.462	2.771E-04	33.979	1.040	2.000E-03	26.97
232.50	7.560	8.486	2.770E-04	34.035	1.039	1.800E-03	26.76
235.00	7.500	8.509	2.770E-04	34.091	1.038	1.600E-03	26.55
237.50	7.440	8.531	2.770E-04	34.147	1.037	1.400E-03	26.34
240.00	7.380	8.553	2.770E-04	34.203	1.036	1.200E-03	26.13
242.50	7.320	8.574	2.770E-04	34.259	1.035	1.000E-03	25.92
245.00	7.260	8.595	2.770E-04	34.315	1.034	8.000E-04	25.71
247.50	7.200	8.615	2.770E-04	34.371	1.033	6.000E-04	25.50
250.00	7.140	8.635	2.770E-04	34.427	1.032	4.000E-04	25.29
252.50	7.080	8.654	2.770E-04	34.483	1.031	2.000E-04	25.08
255.00	7.020	8.673	2.770E-04	34.539	1.030	1.000E-04	24.87
257.50	6.960	8.691	2.770E-04	34.595	1.029	5.000E-05	24.66
260.00	6.900	8.709	2.770E-04	34.651	1.028	2.000E-05	24.45
262.50	6.840	8.726	2.770E-04	34.707	1.027	1.000E-05	24.24
265.00	6.780	8.743	2.770E-04	34.763	1.026	5.000E-06	24.03
267.50	6.720	8.759	2.770E-04	34.819	1.025	2.000E-06	23.82
270.00	6.660	8.775	2.770E-04	34.875	1.024	1.000E-06	23.61
272.50	6.600	8.790	2.770E-04	34.931	1.023	5.000E-07	23.40
275.00	6.540	8.805	2.770E-04	34.987	1.022	2.000E-07	23.19
277.50	6.480	8.819	2.770E-04	35.043	1.021	1.000E-07	22.98
280.00	6.420	8.833	2.770E-04	35.099	1.020	5.000E-08	22.77
282.50	6.360	8.846	2.770E-04	35.155	1.019	2.000E-08	22.56
285.00	6.300	8.859	2.770E-04	35.211	1.018	1.000E-08	22.35
287.50	6.240	8.871	2.770E-04	35.267	1.017	5.000E-09	22.14
290.00	6.180	8.883	2.770E-04	35.323	1.016	2.000E-09	21.93
292.50	6.120	8.894	2.770E-04	35.379	1.015	1.000E-09	21.72
295.00	6.060	8.905	2.770E-04	35.435	1.014	5.000E-10	21.51
297.50	6.000	8.915	2.770E-04	35.491	1.013	2.000E-10	21.30
300.00	5.940	8.925	2.770E-04	35.547	1.012	1.000E-10	21.09
302.50	5.880	8.934	2.770E-04	35.603	1.011	5.000E-11	20.88
305.00	5.820	8.943	2.770E-04	35.659	1.010	2.000E-11	20.67
307.50	5.760	8.951	2.770E-04	35.715	1.009	1.000E-11	20.46
310.00	5.700	8.959	2.770E-04	35.771	1.008	5.000E-12	20.25
312.50	5.640	8.966	2.770E-04	35.827	1.007	2.000E-12	20.04
315.00	5.580	8.973	2.770E-04	35.883	1.006		

TABLE II (contd)

HYDROGEN E O S OF MILLS, LIEBENBERG, SCHMIDT				T = 275.0 K				HYDROGEN E O S OF MILLS, LIEBENBERG, SCHMIDT				T = 300.0 K			
PRESS.	VOL.	V(S)	ALPHA	C(P)	GAMMA	BETA(S)	ENTROPY	PRESS.	VOL.	V(S)	ALPHA	C(P)	GAMMA	BETA(S)	ENTROPY
KBAR	CC/MOLE	KM/S	/K	J/MOLE-K		/KBAR	J/MOLE-K	KBAR	CC/MOLE	KM/S	/K	J/MOLE-K		/KBAR	J/MOLE-K
2.00	26.924	2.788	1.579E-03	29.943	1.359	1.719E-01	64.28	2.00	27.990	2.782	1.524E-03	30.092	1.362	1.794E-01	64.70
2.50	24.336	3.017	1.388E-03	29.849	1.326	1.326E-01	62.39	2.50	25.182	3.007	1.346E-03	30.009	1.330	1.581E-01	64.80
3.00	22.505	3.221	1.244E-03	29.788	1.299	1.076E-01	60.86	3.00	23.205	3.207	1.210E-03	29.955	1.304	1.119E-01	63.26
3.50	21.122	3.404	1.131E-03	29.750	1.276	9.040E-02	59.56	3.50	21.720	3.389	1.102E-03	29.921	1.282	9.382E-02	61.94
4.00	20.029	3.572	1.041E-03	29.728	1.258	7.787E-02	58.45	4.00	20.551	3.555	1.074E-03	29.901	1.264	8.048E-02	60.85
4.50	19.137	3.726	9.664E-04	29.716	1.242	6.834E-02	57.47	4.50	19.599	3.708	9.441E-04	29.892	1.248	7.071E-02	59.87
5.00	18.390	3.870	8.038E-04	29.716	1.228	6.090E-02	56.59	5.00	18.805	3.681	8.371E-04	29.890	1.234	6.290E-02	58.99
5.50	17.751	4.005	6.505E-04	29.722	1.216	5.491E-02	55.80	5.50	18.179	3.945	8.302E-04	29.895	1.222	5.643E-02	58.20
6.00	17.197	4.131	5.043E-04	29.733	1.206	4.998E-02	55.08	6.00	17.543	4.111	7.872E-04	29.905	1.212	5.148E-02	57.48
6.50	16.710	4.251	3.640E-04	29.749	1.197	4.596E-02	54.41	6.50	16.899	4.231	7.440E-04	29.915	1.202	4.719E-02	56.80
7.00	16.277	4.365	2.285E-04	29.769	1.188	4.237E-02	53.80	7.00	16.573	4.345	7.134E-04	29.925	1.194	4.355E-02	56.20
7.50	15.888	4.473	6.969E-04	29.791	1.181	3.938E-02	53.22	7.50	16.164	4.453	6.826E-04	29.955	1.187	4.052E-02	55.63
8.00	15.536	4.577	6.687E-04	29.816	1.174	3.678E-02	52.69	8.00	15.795	4.557	6.550E-04	29.977	1.180	3.773E-02	55.09
8.50	15.215	4.677	6.432E-04	29.844	1.168	3.451E-02	52.18	8.50	15.459	4.656	6.301E-04	30.001	1.174	3.537E-02	54.59
9.00	14.921	4.772	6.201E-04	29.873	1.163	3.250E-02	51.71	9.00	15.152	4.752	6.074E-04	30.026	1.168	3.328E-02	54.12
9.50	14.650	4.864	5.991E-04	29.904	1.157	3.071E-02	51.26	9.50	14.869	4.844	5.868E-04	30.053	1.163	3.143E-02	53.67
10.00	14.399	4.953	5.799E-04	29.936	1.153	2.911E-02	50.83	10.00	14.607	4.933	5.680E-04	30.081	1.158	2.977E-02	53.25
10.50	14.166	5.039	5.622E-04	29.970	1.148	2.767E-02	50.42	10.50	14.365	5.019	5.504E-04	30.111	1.153	2.828E-02	52.84
11.00	13.949	5.122	5.460E-04	30.005	1.145	2.637E-02	50.03	11.00	14.138	5.103	5.344E-04	30.141	1.149	2.693E-02	52.45
11.50	13.745	5.203	5.309E-04	30.040	1.141	2.518E-02	49.66	11.50	13.926	5.184	5.198E-04	30.172	1.146	2.571E-02	52.08
12.00	13.553	5.281	5.169E-04	30.076	1.137	2.410E-02	49.30	12.00	13.728	5.262	5.060E-04	30.204	1.142	2.459E-02	51.73
12.50	13.373	5.358	5.039E-04	30.114	1.134	2.311E-02	48.96	12.50	13.540	5.339	4.932E-04	30.236	1.136	2.357E-02	51.39
13.00	13.202	5.432	4.918E-04	30.151	1.131	2.220E-02	48.63	13.00	13.364	5.413	4.812E-04	30.269	1.130	2.262E-02	51.06
13.50	13.041	5.504	4.804E-04	30.189	1.128	2.135E-02	48.31	13.50	13.197	5.485	4.700E-04	30.303	1.125	2.176E-02	50.75
14.00	12.888	5.574	4.698E-04	30.228	1.126	2.057E-02	48.00	14.00	13.038	5.556	4.595E-04	30.336	1.120	2.095E-02	50.44
14.50	12.742	5.643	4.598E-04	30.267	1.123	1.985E-02	47.70	14.50	12.888	5.625	4.496E-04	30.371	1.117	2.020E-02	50.15
15.00	12.604	5.710	4.503E-04	30.307	1.121	1.917E-02	47.42	15.00	12.745	5.692	4.403E-04	30.405	1.115	1.951E-02	49.86
15.50	12.471	5.776	4.414E-04	30.347	1.119	1.854E-02	47.16	15.50	12.608	5.758	4.315E-04	30.440	1.113	1.886E-02	49.59
16.00	12.345	5.840	4.330E-04	30.387	1.117	1.795E-02	46.97	16.00	12.478	5.823	4.232E-04	30.475	1.110	1.825E-02	49.32
16.50	12.224	5.903	4.251E-04	30.427	1.115	1.740E-02	46.80	16.50	12.353	5.886	4.153E-04	30.511	1.108	1.769E-02	49.06
17.00	12.108	5.965	4.175E-04	30.468	1.113	1.688E-02	46.65	17.00	12.233	5.948	4.078E-04	30.546	1.106	1.715E-02	48.80
17.50	11.996	6.025	4.104E-04	30.509	1.111	1.639E-02	46.50	17.50	12.118	6.008	4.007E-04	30.582	1.105	1.665E-02	48.56
18.00	11.889	6.084	4.036E-04	30.550	1.109	1.593E-02	46.35	18.00	12.008	6.068	3.940E-04	30.618	1.103	1.618E-02	48.32
18.50	11.786	6.143	3.971E-04	30.591	1.108	1.549E-02	46.20	18.50	11.902	6.126	3.875E-04	30.654	1.101	1.573E-02	48.09
19.00	11.687	6.200	3.909E-04	30.633	1.106	1.508E-02	46.05	19.00	11.800	6.184	3.814E-04	30.691	1.100	1.531E-02	47.86
19.50	11.591	6.256	3.850E-04	30.674	1.105	1.469E-02	45.90	19.50	11.701	6.240	3.756E-04	30.727	1.098	1.491E-02	47.64
20.00	11.499	6.311	3.794E-04	30.716	1.103	1.432E-02	45.76	20.00	11.607	6.295	3.700E-04	30.763	1.097	1.453E-02	47.42
20.50	11.409	6.365	3.740E-04	30.758	1.102	1.397E-02	45.62	20.50	11.515	6.350	3.646E-04	30.800	1.096	1.417E-02	47.21
21.00	11.323	6.419	3.688E-04	30.800	1.101	1.363E-02	45.48	21.00	11.426	6.403	3.595E-04	30.837	1.095	1.382E-02	47.00
21.50	11.239	6.471	3.639E-04	30.841	1.100	1.331E-02	45.34	21.50	11.340	6.456	3.546E-04	30.875	1.094	1.350E-02	46.79
22.00	11.158	6.523	3.591E-04	30.883	1.098	1.301E-02	45.20	22.00	11.257	6.508	3.498E-04	30.910	1.093	1.318E-02	46.60
22.50	11.080	6.574	3.545E-04	30.925	1.097	1.272E-02	45.06	22.50	11.177	6.559	3.453E-04	30.947	1.092	1.289E-02	46.40

Fig. 5.

Molar volume of fluid $n\text{-H}_2$ as a function of pressure along five isotherms computed from Eqs. (4) and (8) using the smoothed set of constants in Table I. Solid line, 300 K; dotted line, 250 K; dashed line, 200 K; short-dash line, 150 K; dash-dot line, 100 K.

Fig. 6.

Sound velocity in fluid $n\text{-H}_2$ as a function of pressure along five isotherms computed from Eqs. (4), (7), and (8). Solid line, 300 K; dotted line, 250 K; dashed line, 200 K; short-dash line, 150 K; dash-dot line, 100 K.

Fig. 7.

Isobaric thermal expansion coefficient of fluid $n\text{-H}_2$ as a function of pressure along five isotherms computed from Eqs. (4), (7), and (9). Solid line, 300 K; dotted line, 250 K; dashed line, 200 K, short-dash line, 150 K; dash-dot line, 100 K.

Fig. 8.

Heat capacity at constant pressure of fluid $n\text{-H}_2$ as a function of pressure along five isotherms computed from Eqs. (4), (7), and (8). Solid line, 300 K; dotted line, 250 K; dashed line, 200 K; short-dash line, 150 K; dash-dot line, 100 K.

Fig. 9.

Heat capacity at constant volume of fluid $n\text{-H}_2$ as a function of pressure along five isotherms computed from Eqs. (4), (7), and (8). Solid line, 300 K; dotted line, 250 K; dashed line, 200 K; short-dash line, 150 K; dash-dot line, 100 K.

Fig. 10.

Isothermal compressibility coefficient of fluid $n\text{-H}_2$ as a function of pressure along five isotherms computed from Eqs. (4), (7), and (8). Solid line, 300 K; dotted line, 250 K; dashed line, 200 K; short-dash line, 150 K; dash-dot line, 100 K.

The lower pressure values derived by Michels however, are well within the error of our extrapolated measurements.

Our EOS was extrapolated to high pressures and temperatures to compare values with calculations by Baker and Swift¹⁹ and Grigoriev et al.²⁰ as discussed in I. The molar volumes of Baker and Swift at $T > 1000$ K are larger by 10-20% at pressures from 20 to 100 kbar than those given by Eq. (4).

Recently Ashurst¹⁰ developed an EOS based on a pair-wise intermolecular interaction of the Lennard-Jones type and derived many thermodynamic properties. Ashurst generously prepared his data to match our format so that the comparison in Table III is possible. At room temperature and low pressure the agreement between measured and calculated values of V and v_s is quite good. However, at lower temperatures the volumes calculated by Ashurst are smaller than those we measure. At high pressures, throughout the temperature range, the sound velocities calculated are all larger than those measured. Thus, while the volumetric data are in moderately good agreement up to 5 kbar the sound velocity difference exceeds the experimental error by an order of magnitude and indicates the virtue of using simultaneous measurements of V and v_s . Since Ashurst's calculated sound velocities are in error, this implies that the other thermodynamic quantities will have limited accuracy and in general we find even larger differences.

G. I. Kerley¹¹ has calculated theoretically the various thermal properties of high-density hydrogen. As indicated in Fig. 11, the agreement between the calculated and measured molar volumes is excellent. In Fig. 12 similar data for the sound velocity are presented and the disagreement is only slightly larger than the experimental errors. Since the agreement between molar volumes and sound velocity data was quite good, specific heats at 2 kbar were determined by Kerley for our possible use in revising Eq. (8). Unfortunately the specific heat C_p shows large disagreements that we have not resolved, but are in part related to the lack of spin dependence used in the calculations.

Entropies calculated by Kerley were compared with values obtained from an expanded version of Eq. (16) that we know are within a few percent of the full EOS calculation. Along the 200 and 500 K isotherms the entropy values agree to within 10%, which we consider very good. Similar agreement is found with the tabular data of Vargaftik.¹⁷

Haar and Shenker²¹ used the Percus-Yevick relationship along with a temperature-dependent hard-sphere diameter to derive an EOS. In applying this formalism to our results for hydrogen, we find that the hard-sphere diameter

TABLE III

Comparison of Eq. (4) from Mills, Liebenberg, Bronson, and Schmidt, I with unpublished data of W. T. Ashurst in Ref. 19. In each case the value listed is obtained as $\{[Y(\text{MLBS}) - Y(A)]/Y(\text{MLBS})\} \times 100$, or percent deviation.

<u>T</u>	<u>P</u>	<u>Molar Volume Deviation</u>	<u>Sound Velocity Deviation</u>	<u>Specific Heat Deviation</u>	<u>Thermal Expansion Coeff. Deviation</u>
300	2	0.35	0.36	3.0	-1.14
	5	-1.01	-5.92	-0.35	-4.70
	10	-3.42	-11.5	-2.03	-8.93
	15	-5.42	-14.9	-2.55	-10.2
	20	-6.98	-17.2	-2.84	-10.9
250	2	0.31	0.36	1.70	-1.10
	5	-0.78	-6.22	-1.76	-7.18
	10	-3.17	-11.9	-3.72	-10.4
	15	-5.14	-15.3	-4.73	-11.6
	20	-6.76	-17.4	-6.52	-14.3
200	2	0.59	0	1.06	-4.92
	5	-0.35	-6.85	-4.04	-10.9
	10	-2.83	-12.6	-7.67	-13.3
	15	-4.77	-15.7	-11.8	-17.7
	20	-6.36	-17.4	-20.4	-31.7
150	2	1.34	-0.38	-8.77	-13.6
	5	0.37	-7.60	-18.2	-19.0
	10	-2.18	-12.9	-31.49	-27.0
	15*	*	*	*	*
100	2	3.69	-2.45	-5.71	-46.6
	5	2.13	-8.85	-112	-62.0
75	2	7.05	-11.3	-151	-122

*According to Ashurst the solid phase was entered.

Fig. 11.

Molar volume vs pressure. The full line is data from Kerley (Ref. 11) at $T=300$ K. Circles are our EOS values from Eqs. (4), (7), and (8) at 300 K. The dashed line from Kerley and triangles of our EOS are at $T=100$ K.

must also be pressure dependent to give a reasonable fit to the data up to 20 kbar along the 300 K isotherm. When the second virial coefficient is taken as density independent in the form given by Weber,²² then the hard-sphere radius decreases with increasing pressure. Sound velocities estimated with the virial EOS are in quite good agreement at 300 K but are much too steep a function of pressure at lower temperatures.

Since hydrogen at high densities exhibits liquid-like characteristics, such as the velocity of sound increasing with decreasing temperature, we have applied Rao's Law to the data.²³ Rao's Law states that

$$R_a = v_s^{1/3} \frac{M}{\rho} \quad (17)$$

The "constant" R_a is given the interpretation of a molar velocity of sound in analogy with molecular refraction. From our measurements we determine values

Fig. 12.

Sound velocity vs pressure. The full line is data from Kerley (Ref. 11) at $T=300$ K. Circles are EOS values from Eqs. (4), (7), and (8) at 300 K. The dashed line from Kerley and triangles of our EOS are at $T=100$ K.

of $V v_s^{1/3} \propto R_a$ given in Table IV. A large temperature variation in R_a is found. Thus, fluid hydrogen does not follow Rao's Law.

TABLE IV

Comparison of Rao's Law, a free volume model, and a parameterization by the present authors as discussed in the text.

Rao rule $v_s^{1/3} V = \text{constant}$			
Pressure kbar	300 K	200 K	100 K
2	39.37	33.61	28.69
5	29.48	27.08	24.87
10	24.87	23.48	Freezing
15	22.76	21.82	
20	21.43	20.68	

Free Volume model $v_s^{1/3} V = \text{constant}$			
	300 K	200 K	100 K
2	8.45	8.14	8.27
5	10.24	10.17	10.36
10	12.06	12.03	Freezing
15	13.30	13.27	
20	14.25	14.21	

Liebenberg, Mills, Bronson model $v_s V = \text{constant}$			
	300 K	200 K	100 K
2	77.87	67.29	60.42
5	72.42	67.61	64.31
10	72.06	69.11	Freezing
15	72.54	70.18	
20	73.07	70.99	

Several different attempts have been made to calculate the propagation of sound waves using a free volume model.^{23,24} In one model the mechanism for propagation proposes a gas-kinetic velocity in the spaces between the molecules and an infinitely fast velocity inside the molecules. The actual velocity is

then the sum of these two velocities taken in proportion to the fractional volumes involved in the discontinuous propagation. Thus, in the model by Schaaffe, as discussed by van Dael and van Itterbeek,²⁴ $v_s = v_\infty r s$, where $r = B_0/V$, $s = b/B_0$, and $b = 2/3 \pi N \sigma^3$. B_0 is the geometrical volume of N molecules each with interaction distance σ , and v_∞ is a constant. In this case the sound velocity can be written $v_s = 2/3 \pi v_\infty / V^*$, where $V^* = V/V_0 = V/N\sigma^3$ and $r_0^6 = 2\sigma^6$. If the value of b given by Eyring is used (assuming a simple cubic lattice so the free volume is $8[(V/N)^{1/3} - \sigma]^3$ per molecule) then $b = N^{1/3} \sigma V^{2/3}$ resulting in

$$v_s = \frac{v_\infty N^{1/3} \sigma}{V^{1/3}} = \frac{v_\infty}{V^*^{1/3}} \quad (18)$$

and thus $v_s V^{1/3} = v_\infty N^{1/3} \sigma$, the rhs being treated as a constant. This relation is essentially different from the relation of Rao's rule, Eq. (17). Values of $v_s V^{1/3}$ are tabulated in Table IV.

From the data for high-density fluid hydrogen we have determined that Eq. (1) is valid, and since the exponents n for both volume and sound velocity are nearly equal and of opposite sign, we are led to the conclusion that

$$v_s V = \text{constant}. \quad (19)$$

In Table IV we compare the application of the high-density fluid-hydrogen data to Rao's rule, to the free-volume model with the Eyring approximation, and to our own relation Eq. (19). The hydrogen data show a temperature variation that is larger than in either the free-volume model or Rao's rule (which is temperature independent). In comparison with both the free-volume model and Rao's rule our own relation shows much less pressure dependence over the range of data.

Some further discussion of Eq. (1) is possible.²⁵ A Lennard-Jones 6, 12 potential is used in the high-density limit. Thus,

$$\phi(R) = 4\epsilon \left[\left(\frac{R_0}{R} \right)^{12} - \left(\frac{R_0}{R} \right)^6 \right] \quad (20)$$

and in terms of the volume at high density $\phi(V) = 4\epsilon (V_0/V)^4$. Then since pressure, $P = -(\partial F / \partial V)_T$, therefore $P = 16\epsilon V_0^4 / V^5 = A\rho^5$. The sound velocity is given

approximately as $v_s^2 = (\partial P / \partial \rho)_s \approx 5A\rho^4$. Thus, in terms of the pressure

$$v_s \propto P^{2/5}$$

and

$$V \propto P^{-1/5} \quad (21)$$

The product of v_s and V shows a weak pressure dependence $v_s V \propto P^{+1/5}$. However, if the repulsive part of the potential is taken to have the form $\phi(R) = 4\epsilon[(\frac{R_0}{R})^9 - (\frac{R_0}{R})^6]$ and the above arguments carried through, we find $v_s \propto P^{3/8}$, $V \propto P^{-1/3}$ and $v_s V \propto P^{1/24}$. In this latter case the determined exponents are in quite good agreement with the derived values. The purpose of this argument has been to indicate the plausibility of Eqs. (1) and (2).

In summary we have provided here a supplement to the published⁹ pressure, volume, temperature, and sound velocity measurements and the derived equation of state for $n\text{-H}_2$ fluid at pressures up to 20 kbar in the temperature range 75–307 K. Thermodynamic quantities of interest were calculated and presented in tabular form for convenience. These thermodynamic quantities include the thermal expansion coefficient, heat capacity, isothermal compressibility, heat capacity ratio, and molar entropy. We have examined the characteristics of our EOS at high pressure and compared our results with several theoretical models. In particular we found differences of 15% in values of V and v_s derived from a modified Lennard-Jones potential by Ashurst¹⁰ and even greater differences in derived thermodynamic quantities. The molar volumes obtained from a phenomenological model by Kerley¹¹ were in excellent agreement. Though the Kerley sound velocities were found to deviate by greater differences than the experimental errors, the agreement was better than in other models tested. A qualitative argument was presented for the observation that the product of the sound velocity and volume of high-density fluid hydrogen is approximately constant, in contrast to the Rao rule and the free-volume model.

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16. The critical temperature for H_2 is 33.2 K and the critical pressure is 13.2 bar.

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APPENDIX

This program will provide the formatted output of Table II, obtained from a solution of the EOS Eq. (4) with the smoothed data constants in Table I.

```

00100 PROGRAM H2BEN(OUTPUT)
00110*** CALCULATION OF THERMODYNAMIC PROPERTIES HYDROGEN
00120 DIMENSION VO(50), CP(50), VDO(50), AP(50), VS(50), VSS(50)
00130 DIMENSION CAM(50), BETAS(50), P(50)
00140 DIMENSION DENT(50)
00150 T=75.0
00160*** INPUT PRESSURE IN KBAR , TEMPERATURE IN KELVIN
00170 DO 6 K=1,10
00180 CP1=(1.123+T-61.468*(T**0.5)+1259.3-(10512.0/(T**0.5))
00190+ +(31638.0/T))/2.016E-7
00200 PRINT 14,T
00210 14 FORMAT( // 44H CALCULATED VALUES FROM EOS FOR H2 AT ,F5.1
00220+,3H K.)
00230*** THESE VALUES ARE NORMALIZED TO 2 KBAR THROUGH C(P)
00240 DO 5 I=1,42
00250 IF(I-1)1,1,2
00260 1 P(I)=2.0
00270 2 CONTINUE
00280 A=36.7157 $ B=0.00330027 $ C=-22.4789
00290 D=-17.1738 $ E=-0.0213930 $ F=-8.98863
00300 G=0.110013 $ H=69.2326 $ J=-31.3951
00310 B1=A+B*T+C/(T**0.5)
00320 B2=D+E*T
00330 S3=F+G*T + (H/(T**0.5)) + J/T
00340 AA=B-(0.5*C/(T**1.5))
00350 BB=G-(0.5*H/(T**1.5))-(J/(T**2.0))
00360 DENT2=(AA*1.5*(2.0**0.66667))+E*3.0*(2.0**0.33333)+(BB*ALOG(2.0))
00370 DENT2=DENT2+100.0
00380 DENT(I)=(AA*1.5*(P(I)**0.66667))+E*3.0*(P(I)**0.33333)+
00390+(BB*ALOG(P(I)))+100.0
00400 DENT(I)=DENT(I)-DENT2
00410 SENT=12.194+(0.27442*T)-(3.0914E-4*(T**2.0))
00420 DENT(I)=SENT-DENT(I)
00430 VO(I)=(B1/(P(I)**0.33333))+(B2/(P(I)**0.66667))+(S3/P(I))
00440 AP(I)=(AA/(P(I)**0.33333))+E/(P(I)**0.66667)+(BB/P(I))/VO(I)
00450*** EXPANSION COEFF. (K-1)
00460 CP(I)=(0.5+1.5*1.5*(C/(T**1.5)))+(P(I)**0.66667)-(2.0**0.66667))
00470 CP(I)=CP(I)+((0.5*1.5*H/(T**1.5))+(2.0*J/(T**2.0)))+(ALOG(P(I))
00480+ALOG(2.0))
00490 CP(I)=CP(I)+1.0E9/2.016
00500 CP(I)=CP1-CP(I)
00510 VSS(I)=(B1*0.33333/(P(I)**1.33333))+(B2*0.66667/(P(I)**1.66667))
00520+ +(B3/(P(I)**2.0)))+(2.016E-9/(VO(I)**2.0))
00530 VS(I)=VSS(I)-(T*(AP(I)**2.0)/CP(I))
00540 VS(I)=ABS(VS(I))
00550 VS(I)=VS(I)**0.5
00560 VS(I)=1.0/VS(I)
00570*** C(P) J/MOLE-K VS THERMAL CM/SEC
00580 P(I+1)=P(I)+0.5
00590 CAM(I)=VSS(I)*(VS(I)**2.0)
00600 BETAS(I)=(VO(I)+1.0E9)/(2.016*(VS(I)**2.0))
00610 VS(I)=1.0E-5*VS(I)
00620 CP(I)=CP(I)+2.016E-7
00630 5 CONTINUE
00640 PRINT 100
00650 PRINT 500
00660 100 FORMAT( // * PRESS. VOL. V(S) ALPHA C(P) GAMMA
00670+BETA(S) ENTROPY * )
00680 500 FORMAT( // * KBAR CC/MOLE CM/S /K J/MOLE-K
00690+KBAR J/MOLE-K * / )
00700 300 FORMAT( 0PF5.2,0P2F9.3,3X,1PE11.3,0PF9.3,2X,0PF7.3,1PE11.3,0PF7.2)
00710 PRINT 300, (P(I),VO(I),VS(I),AP(I),CP(I),CAM(I),BETAS(I),DENT(I),
00720+ I=1,42)
00730 T=T+25.0
00740 6 CONTINUE
00750 END

```