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ANALYTICAL AND PRELIMINARY DESIGN STUDIES
OF NUCLEAR ROCKET PROPULSION SYSTEMS

VOLUME VI - COMPILATION OF HYDROGEN PROPERTIES

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AEROJET-GENERAL CORPORATION
Azusa, California
A Subsidiary of
The General Tire & Rubber Company

Aerojet-General
Report No. 1999 (Final)

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JUNE 1961

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ERRATA
FOR
ANALYTICAL AND PRELIMINARY DESIGN STUDIES
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VOLUME VI - COMPILATION OF HYDROGEN PROPERTIES

1. Figure 9, Page 15, Compressibility Factor Hydrogen Gas at Low Temperatures.

Compressibility Factors .6, .7 and .8 should be moved down 1/2 of a major division. In other words, there should be 1 1/2 major divisions between each numeral in the ordinate.

2. Figure 15, Page 22, Specific Heat of Normal Gaseous Hydrogen.

In the 50 to 55 degree R range the 20 atm and 40 atm lines are reversed.

ABSTRACT

This volume presents a graphic compilation of the physical, thermodynamic, and transport properties of molecular hydrogen, including the effects of variations in the ratio of concentrations of orthohydrogen and parahydrogen. It provides a single source of correlated data in engineering units for the design of fluid systems utilizing hydrogen.

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THE HYDROGEN ATOM

The hydrogen atom is the simplest of all the chemical elements. Its nucleus consists of one proton, and it has one orbital electron. Hydrogen has two isotopes: deuterium, in which the nucleus consists of one proton and one neutron, and tritium, which has a nucleus of one proton and two neutrons. Deuterium is stable and has a natural abundance in hydrogen of 0.015 atom-%. Tritium is unstable, but has a long half-life (12.26 years). It decays into helium 3 by the emission of an 0.018-Mev beta particle (Ref. 29).

The atomic mass unit (amu) is defined as one-sixteenth the mass of an oxygen atom. The most abundant stable isotope of oxygen is O^{16} , but O^{17} and O^{18} are also stable. The atomic mass unit on the physical scale is one-sixteenth the mass of the O^{16} atom. Due to the presence of the O^{17} and O^{18} isotopes, the average atomic weight of oxygen is somewhat greater. The atomic mass unit on the chemical scale is one-sixteenth the average atomic weight of oxygen with the stable isotopes in their natural abundances.

Because of the natural occurrence of deuterium in hydrogen, the average atomic weight of hydrogen is slightly greater than the atomic weight of the hydrogen atom. It is this average atomic weight on the chemical scale that is used in thermodynamic calculations. The atomic weights of some particles and atoms are given in Table 1 (Ref. 30).

The thermal-neutron absorption cross section for hydrogen is 0.330 barn, and it follows the $(1/v)$ law with no resonance (Refs. 31 and 39). For neutrons of any energy, the absorption cross section (σ_a , in barns) can be determined by the relation

$$\sigma_a = \frac{0.0525}{\sqrt{E_n}}$$

where E_n is the neutron energy in electron volts.

The thermal-neutron scattering cross section for hydrogen, averaged over a Maxwell distribution of neutron energies, is 38 barns. The total cross section of hydrogen for neutron energies from 0.003 ev to 100 Mev is shown in Figure 1 (Ref. 31).

TABLE 1
ATOMIC WEIGHTS OF SOME PARTICLES AND ATOMS

	<u>Symbol</u>	<u>Atomic Weight, amu</u>	
		<u>Physical Scale*</u>	<u>Chemical Scale*</u>
Electron	e	0.000548760	0.000548609
Proton	p	1.007595	1.007318
Neutron	n	1.008982	1.008705
Hydrogen atom	H ¹	1.008144	1.007867
Deuterium atom	D ²	2.014739	2.014185
Av atomic wt of hydrogen	H	1.008295	1.008018
Oxygen 16 atom	O ¹⁶	16.000000	15.995601
Av atomic wt of oxygen	O	16.004400	16.000000

* 1 chemical amu = 1.000275 physical amu.

1 physical amu = 1.6598×10^{-24} gram.

FIGURE 1
NEUTRON CROSS SECTIONS OF HYDROGEN

THE HYDROGEN MOLECULE

Each nucleus (i.e., proton) of the diatomic hydrogen molecule has a mechanical momentum characterized as "nuclear spin." Molecules in which the spins of the two nuclei are parallel are called orthohydrogen (o-H₂), and those in which these spins are antiparallel are called parahydrogen (p-H₂). At temperatures above 0°F, hydrogen is 75% o-H₂ and 25% p-H₂; this composition is referred to as normal hydrogen (n-H₂). When the temperature is lowered, the o-H₂ molecules gradually convert into p-H₂ molecules. The equilibrium ortho-para composition of hydrogen as a function of temperature is shown in Figure 2. This equilibrium ortho-para ratio is not appreciably affected by pressure (Ref. 1).

The rotational energy levels of the o-H₂ molecules have odd quantum numbers; those of the p-H₂ molecules have even numbers. Thus, in their respective lowest energy states, the o-H₂ molecules have a rotational level of 1 and the p-H₂ molecules have a rotational level of 0. In the conversion of o-H₂ to p-H₂, this difference in rotational energy is released as heat. This heat of conversion (Ref. 33) is

$$H_c = 302.4 \text{ Btu/lb}$$

In the liquid, the conversion rate is independent of temperature and, in the absence of paramagnetic substances, is proportional to the square of the o-H₂ concentration. Letting o denote the fraction of liquid in o-H₂ form,

$$-\frac{do}{dt} = 0.0114 \text{ hr}^{-1} o^2$$

Integrating and letting the initial condition be the normal composition,

$$o = \frac{1}{4/3 + 0.0114 \text{ hr}^{-1} t}$$

The conversion, in the absence of molecular dissociation, is caused by a reversal of the nuclear spin of one proton, which requires the presence of a strong magnetic field that must be inhomogeneous on a scale of molecular dimensions. Because p-H₂ molecules have no net magnetic moment, the self conversion must occur by the interaction of two o-H₂ molecules. The p-H₂ concentration and rate of heat liberation with time are shown in Figure 3 for liquefied normal hydrogen.

In the solid state, the self conversion commences at a much faster rate than in the liquid:

$$-\frac{do}{dt} = 0.019 \text{ hr}^{-1} o^2$$

However, when the o-H_2 fraction becomes reduced to about 0.30, the reaction-rate constant begins to decrease and becomes even less than for the liquid. If the solid is melted and resolidified, the rate constant returns to its original high value and starts decreasing again. This phenomenon is explained by the small mobility of the molecules in the solid state. As the conversion proceeds, the o-H_2 molecules become isolated from each other and cannot interact. When the solid is melted, their mobility is restored and they have the opportunity to "find" each other, thus accelerating the reaction rate.

The conversion rate is increased when paramagnetic molecules (O_2 , NO , Fe(OH)_3 , etc.) are added to the liquid, because an o-H_2 molecule then no longer requires a second o-H_2 molecule to provide the magnetic field for the conversion. A surface catalysis is effected if the liquid is contained in a paramagnetic vessel.

FIGURE 2
EQUILIBRIUM CONCENTRATION OF PARAHYDROGEN

FIGURE 3

PARAHYDROGEN CONCENTRATION & RATE OF HEAT LIBERATION
FOR UNCATALYZED CONVERSION OF LIQUID
ORTHO- TO PARAHYDROGEN

GAS CONSTANT, CRITICAL POINT, AND TRIPLE POINTGas Constant

$$R = 1.98719 \text{ cal/mole-}^{\circ}\text{K} = 766.53 \text{ ft-lbf/lbm-}^{\circ}\text{R} \quad (\text{Refs. 3 and 48})$$

Critical Point

Normal hydrogen (n-H₂): 75% o-H₂, 25% p-H₂

$$T_c = 59.84 \text{ }^{\circ}\text{R} \quad (\text{Ref. 34, p. 11})$$

$$P_c = 188.12 \text{ psia}$$

Equilibrium hydrogen at 36.7 °R (e-H₂): 0.21% o-H₂, 99.79% p-H₂

$$T_c = 59.39 \text{ }^{\circ}\text{R} \quad (\text{Ref. 41})$$

$$P_c = 187.67 \text{ psia}$$

Triple Point

Normal hydrogen (n-H₂)

$$T_t = 25.13 \text{ }^{\circ}\text{R} \quad (\text{Ref. 53})$$

$$P_t = 1.044 \text{ psia}$$

Equilibrium hydrogen at 36.7 °R (e-H₂)

$$T_t = 24.9 \text{ }^{\circ}\text{R} \quad (\text{Ref. 40})$$

$$P_t = 1.02 \text{ psia}$$

Because equilibrium hydrogen is very nearly pure parahydrogen, its properties may be taken as those of pure parahydrogen with negligible error.

Hydrogen should be considered "normal" if it is a freshly prepared uncatalyzed liquid or if it is in the process of condensing from a vapor. If liquid hydrogen has been stored for a long time, the equilibrium values are a better approximation.

DENSITY

Several sources are used in plotting the pressure-temperature-density graphs (Figures 4 through 6). In the vapor region, the data are interpolated from Refs. 1 and 3. The experimental data of Ref. 14 are used for the liquid. The only known source for solid hydrogen is Ref. 1, which gives the density corresponding to several pressures, mostly at 7.56°R.

Dissociation effects (see Figure 7) are negligible below 2000°R. There is no noticeable change in the density due to changes in the ortho-para ratio (Ref. 3, p. 254). There is a slight shift in the liquid-vapor boundary line, however.

At pressures below 0.2 psia, the perfect-gas equation can be used in the vapor region without introducing noticeable error. Figures 8 and 9 show compressibility factors plotted against temperature.

FIGURE 5
DENSITY OF GASEOUS HYDROGEN

FIGURE 6
DENSITY OF SOLID HYDROGEN

FIGURE 7
DISSOCIATION OF HYDROGEN GAS

FIGURE 8
COMPRESSIBILITY FACTOR
FOR HYDROGEN GAS

FIGURE 9
COMPRESSIBILITY FACTOR
HYDROGEN GAS
AT LOW TEMPERATURES

HEATS OF VAPORIZATION AND FUSION

Very little experimental data have been reported on the heat of vaporization of hydrogen (Figure 10). Reference 13 (1950) is probably the most complete article published. However, it covers only the temperature range of the data, and no comparison is made with the work of other researchers.

References 38 and 53 are articles on vapor pressure. Each computes the heat of vaporization at one point.

The experimental data used in Ref. 1 in calculating the heat of vaporization are those of F. Simon and F. Lange in 1923. The existence of the ortho and para forms of hydrogen was not known until 1928. Consequently, this early research does not consider the effect of the ortho-para ratio. It appears that Simon and Lange had a mixture between normal hydrogen and parahydrogen (Ref. 38). The data do not agree with the other sources and have been disregarded.

An estimated curve is shown between 25 and 37°R because there are no available data.

The only heat-of-fusion data reported are the following:

For normal hydrogen,

$$H_F = 28.0 \pm 0.15 \text{ cal/mole} = 25.0 \pm 0.13 \text{ Btu/lb} \quad (\text{Ref. 1, p. 467})$$

For parahydrogen,

$$H_F = 28.08 \pm 0.15 \text{ cal/mole} = 25.08 \pm 0.13 \text{ Btu/lb} \quad (\text{Ref. 38})$$

FIGURE 10
HEAT OF VAPORIZATION FOR
NORMAL HYDROGEN

SPECIFIC HEAT

Figures 11 through 16 present specific-heat data and heats of conversion for hydrogen (Figure 15).

The specific-heat data given for normal gaseous hydrogen above 33°K (59.4°R) are based on an expression given in Ref. 1 and the subsequent solutions in Refs. 3 and 6. Below 33°K, the experimental data of H. Gutsche (1939), as cited in Ref. 1, have been used.

Above 2000°R, hydrogen begins to dissociate and the specific heat increases tremendously, to peaks up to about 80 Btu/lb-°R, and then decreases. It is best in this area to use the enthalpy difference directly. The specific heat also has high peaks in the supercritical region. At temperatures below 100°R and at pressures greater than about 20 atmospheres, it is best to use the enthalpy difference.

Ref. 3 states that errors in the reported data may possibly be about 10% at room temperatures and may be considerably larger than this at low temperatures. Consequently, the values of the specific-heat ratio are considered to be reliable to within 10% of their departure from values for the ideal gas.

The effect of p-H₂ concentration on specific heat is shown for the ideal gaseous state. Quoting from Ref. 6, "There is no reason to believe that a first-order difference exists between C_p normal and C_p para for the real gas other than the difference in the ideal gaseous state." The variation of specific heat with the ortho-para ratio is independent of pressure. At low temperatures, the variation with pressure is caused by action between the molecules, while the variation with differing p-H₂ concentrations is caused by actions within the molecule.

At absolute zero, the theoretical enthalpy of orthohydrogen is 302.4 Btu/lb greater than that of parahydrogen, while at high temperatures (above 700°R) the two enthalpies are equal. The heat of conversion is the difference of these enthalpies. Because specific heats are given in general by $C = dH/dt$, the changing enthalpies produce unusual effects in the specific heats of ortho-para mixtures. Of particular interest is the shape of the C_p curve of equilibrium hydrogen (mixture in equilibrium at every temperature). A complete theoretical discussion of the relationship between the heat of conversion and the specific heat of hydrogen is given in Ref. 18, pp. 3-19 and 33-38.

FIGURE 11
SPECIFIC HEAT OF SOLID & LIQUID HYDROGEN

FIGURE 12
SPECIFIC HEAT AT CONSTANT PRESSURE
OF HYDROGEN IN THE IDEAL GASEOUS STATE

FIGURE 13
SPECIFIC HEAT AT CONSTANT PRESSURE
OF HYDROGEN IN THE IDEAL GASEOUS STATE

FIGURE 14
SPECIFIC HEAT OF NORMAL GASEOUS HYDROGEN

FIGURE 15
HEAT OF CONVERSION FOR HYDROGEN
IN THE IDEAL GASEOUS STATE

FIGURE 16
SPECIFIC-HEAT RATIO
FOR GASEOUS NORMAL HYDROGEN

THERMAL CONDUCTIVITY

In the gas phase, the thermal conductivity of hydrogen is dependent on temperature, pressure, and the ortho-para ratio. The thermal conductivity of normal gaseous hydrogen up to 1500 psia and to 4000°R is shown in Figure 17. The curves for pressures above 1 atmosphere were determined from the correlation of Schaefer and Thodos (Ref. 82). Several references give data at atmospheric pressure; the data of Refs. 1, 3, 56, and 26 were used. The data of Ref. 45 were used above 2000°R.

The effect of para-concentration on the thermal conductivity of hydrogen is indicated by Farkas (Ref. 18, p. 20). The results of calculations based on the following equation are plotted as Figure 2 in Ref. 6:

$$K_{p-H_2} = K_{n-H_2} \left(\frac{C_{v,p-H_2} + 2.25R}{C_{v,n-H_2} + 2.25R} \right)$$

where p = para, n = normal, and R is the gas constant. This calculation assumes that the para-concentration effect on the real gas is the same as that on the ideal gas.

The ortho-para effect on thermal conductivity of low-pressure hydrogen is shown in Figure 18 of the present report.

Reference 20 reports experimental data for the thermal conductivity of liquid hydrogen. A straight line

$$K = (0.04117 + 0.0007488 T) \frac{\text{Btu}}{\text{hr ft}^2 \text{ } ^\circ\text{R/ft}}$$

with T in °R, is fitted to the data in Ref. 8 by the method of least squares and is shown in Figure 19. The thermal conductivity of liquid hydrogen is independent of the ortho-para composition.

FIGURE 17
THERMAL CONDUCTIVITY OF GASEOUS NORMAL HYDROGEN

FIGURE 18
ORTHO - PARA EFFECT ON THERMAL CONDUCTIVITY
OF LOW-PRESSURE HYDROGEN

FIGURE 19
THERMAL CONDUCTIVITY OF LIQUID HYDROGEN

PRANDTL NUMBER

The Prandtl number of gaseous hydrogen has been calculated from the thermodynamic properties (specific heat, viscosity, and thermal conductivity), using only properties of normal hydrogen. The Prandtl number at other para-concentrations can be calculated by using the modified properties of specific heat and thermal conductivity.

Because of large uncertainties in specific-heat and thermal-conductivity data, the Prandtl number has an uncertainty estimated between 10 and 15%, the larger value being more realistic at low temperatures.

The smoothed curves presented in Figures 20 and 21 are in fair agreement with those of Ref. 6. Above 800°R they differ by about 8% from the values tabulated in Ref. 3.

FIGURE 20

PRANDTL NUMBER OF GASEOUS NORMAL HYDROGENAT LOW TEMPERATURES

FIGURE 21
PRANDTL NUMBER OF GASEOUS NORMAL HYDROGEN

VISCOSITY

The viscosity of hydrogen is plotted as a function of temperature in Figures 22 through 24.

Very little information has been reported on the viscosity of hydrogen. Probably the best source of data is the 1956 report of Chelton, Macinko, and Dean (Ref. 6).

In the gaseous region, the viscosity of hydrogen is independent of the ortho-para ratio. It is only dependent on the temperature and the pressure. The curves plotted are very reliable between 100 and 550°R. Below 100°R, the curves are not uniform in shape and are plotted from few points. Above 550°R, the reported data are very meager and not in agreement.

Curves from two sources are shown in the liquid region. They are not in agreement but are within the spread of the experimental data.

FIGURE 22

VISCOSITY OF HYDROGEN AT LOW TEMPERATURES

FIGURE 23

VISCOSITY OF HYDROGEN

FIGURE 24

VISCOSITY OF HYDROGEN AT HIGH TEMPERATURES

SURFACE TENSION

Figure 25 is from Ref. 16, which used the basic data of H. Kamerlingh Onnes and H. A. Kuypers (Leiden Communication 142d) and A. Van Itterbeek (Physica, 7: 325).

An empirical equation was fitted to the data. The equation is not valid at high temperatures (near the critical) because the temperature corresponding to the vanishing of the surface tension calculated from it does not agree with the temperature of the critical point. In the region covered by the data, 26 to 39°R, the estimated uncertainty is 10%.

DIELECTRIC CONSTANT

Reference 16 credits several sources of data, all agreeing to within 0.3% in the temperature range from 25 to 43°R. An empirical equation,

$$\text{dielectric constant} = 1.296 - 0.001917 T$$

where T is in °R, has been fitted to the data and is presented in Figure 26.

SONIC VELOCITY

Reference 3 is the source for velocities in the vapor region (Figure 27); its data are from several observers. The values are considered very reliable except at the lowest temperatures and at elevated pressures. The uncertainty is probably less than 0.1% for pressures of 10 atmospheres or less. For 100 atmospheres, it is probably less than 0.6% up to 180°R, 0.2% up to 360°R, and 0.1% at higher temperatures.

For the liquid, Ref. 72 (1949) provides the only data reported, the accuracy of which is not known.

FIGURE 25
SURFACE TENSION OF LIQUID HYDROGEN

FIGURE 26
DIELECTRIC CONSTANT OF LIQUID HYDROGEN

FIGURE 27

SONIC VELOCITY AT LOW FREQUENCY IN HYDROGEN

ENTROPY

There are three known temperature-entropy charts for hydrogen. They differ significantly in the high-pressure, low-temperature region. The first (Figure 28) was prepared by the National Bureau of Standards (NBS) and is taken from Ref. 1. The second (in two parts, Figures 29 and 30) was prepared for the internal use of the National Advisory Committee for Aeronautics (NACA), which is now the National Aeronautics and Space Administration (NASA). It is classed (by NASA) as preliminary and has not been formally released for publication.

Except in the high-pressure area near the dome, the two charts appear to be in substantial agreement. The most obvious difference is in the shape of the isenthalpic lines. There appears to be no compelling reason for their directional discontinuity at the saturated-liquid line in the NBS chart.

The third chart (Figure 31) was prepared in Germany. Efforts to locate the publication in which it was issued have been unsuccessful.

The NBS has undertaken an experimental determination of hydrogen properties that should resolve the differences between these charts.

FIGURE 28

NBS TEMPERATURE-ENTROPY DIAGRAM

FIGURE 29

NACA TEMPERATURE-ENTROPY DIAGRAM, 25 to 100°R

FIGURE 30
NACA TEMPERATURE-ENTROPY DIAGRAM, 100 to 500°R

FIGURE 31
GERMAN TEMPERATURE-ENTROPY DIAGRAM

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