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Review of Hydrogen Isotope Permeability Through Materials

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Lawrence Livermore National Laboratory

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Abstract

This report is the first part of a comprehensive summary of the literature on hydrogen isotope permeability through materials that do not readily form hydrides. While we mainly focus on pure metals with low permeabilities because of their importance to tritium containment, we also give data on higher-permeability materials such as iron, nickel, steels, and glasses.

Introduction

This paper is the first part of a comprehensive review and summary of the literature on hydrogen isotope permeability through materials. It is the first review of its kind. Hydrogen isotope permeability is important to many scientific fields, such as metallurgy, nuclear reactor design, and tritium processing.

Our own research has frequently required hydrogen permeability data through many materials, only to be frustrated by an absence of data or by obscure data. This review is a result of that frustration and need. Initially, we selected for review those materials that resist hydrogen penetration because we are interested in improving tritium containment. We have also included many higher-permeability materials, however, such as iron, steels, nickel and copper alloys, and glasses, because of their importance to tritium technology. Hydride-forming metals have been ignored because of their high hydrogen solubility at moderate pressures and their narrow dilute-solution or α-phase regions.

Our intention is to continue to update this review and expand it to include ceramics, modified materials such as coated alloys, and organics. The updating and expansion will be done over several revisions of this report because of the enormity of the task.

The hydrogen permeation literature has several characteristics:

1. Most of it focuses on the higher-permeability materials mentioned above, because hydrogen permeates such materials relatively quickly, allowing ready measurement.

2. A corollary to point 1 is that most data are high-temperature, i.e., between 200°C and 1000°C or the melting point, an experimental approach that also facilitates easier measurement. Useful data at low temperatures do not exist, making extrapolation from high temperatures an error-prone affair.

3. Very few permeation data exist on tritium, probably because of its radioactivity and consequent restrictions on availability and use.

4. When permeability data are given as a function of temperature, the units vary widely, decreasing their utility. We have settled on a unit based on the SI system, which we hope will simplify calculations and make comparisons between materials easy.

5. In several reports the classic "time-lag" permeation method was used for measurement, but only the diffusion coefficient was calculated and reported; the directly measured permeability data were not given.

6. In many cases, diffusion and solubility data were available from different reports, sometimes from the same laboratory, but from different samples and techniques. We combined such data only when no other trustworthy permeability data were available, and still did so with great trepidation.

Definition of Permeability

Steady-state permeability of gases through solids is defined by a variation of Fick's first law,

$$J = -D \frac{\partial c}{\partial x} \quad , \tag{1}$$

where J is the flux of diffusing species, in units of mol/m$^2\cdot$s, diffusing out of the material; D is the diffusion coefficient in m^2/s; and $\partial c/\partial x$ is the concentration gradient across the solid. This solution is most easily envisioned in the semi-infinite plane sheet geometry of an isotropic solid.

Permeability becomes important in practical systems, when the surface concentrations of the gas are not known. In this case Henry's law,

$$S_H = \frac{C_{gas}}{P_{gas}} \quad , \tag{2}$$

is used, where S_H is a constant relating the vapor pressure of a nondissociative gas to its concentration in a liquid or solid at dilute concentration, i.e., the solution phase. The unit of S_H in Henry's law is mol/m$^3\cdot$Pa. C_{gas} is the concentration of gas in the solvent in mol/m^3, and P_{gas} is the pressure of the gas over the solvent in pascals. Since the inlet and outlet pressures of the gas can usually be measured, pressure is substituted for concentration into Fick's first law, via Henry's law.

The Henry's law constant is in fact our solubility constant S, which expresses concentration per unit of pressure. In the case of diatomic molecules, such as hydrogen, that dissociate prior to dissolution, a modification of Henry's law is required. This version is called Sieverts' law, in honor of an early scientist deeply involved in research on hydrogen solubility in metals. His work produced the observation that hydrogen solubility in metals was a function of the half-power of pressure, explained by dissociation of the molecule. Thus Sieverts' law,

$$S = \frac{C_H}{P_{H_2}^{1/2}} \quad , \tag{3}$$

may be used to convert Fick's first law to a usable form. The Sieverts' law constant S is also called solubility and is in units of mol/m$^3\cdot$Pa$^{1/2}$. The constant of Henry's and Sieverts' laws may often be given as inverts of the S expressed above, but the version given in Eq. (3) serves our discussion more clearly.

Substitution of rearranged Eq. (3) into Eq. (1) yields

$$J = -DS \frac{\partial P_{H_2}^{1/2}}{\partial x} \simeq -DS \frac{\Delta P_{H_2}^{1/2}}{\Delta x} \quad . \tag{4}$$

In this case, $\Delta P_{H_2}^{1/2}$ is the square root of the pressure difference of the diffusing gas across a membrane, and Δx is the thickness of the membrane. The negative sign in Eq. (4) represents the direction of diffusion and can be ignored in this discussion.

Equation (4) can be rearranged to correspond to the usual representation of the permeation data, where the total amount of gas that has permeated the membrane, Q (mol), is plotted as a function of time. Equation (4) then becomes

$$Q = JAt = DS \frac{A}{\Delta x} \Delta P_{H_2}^{1/2} \cdot t \quad , \tag{5}$$

where A is the slab area (m^2), and t is the time (s) since reaching steady state.

The slope of this line (dQ/dt), the steady-state flow rate, includes the constant DS, which is specific for a given membrane material. This product of D and S, which we call Φ (sometimes called K), is defined as the permeability, here in the units mol/m$\cdot$s$\cdot$Pa$^{1/2}$. We have, by differentiating Eq. (5) and substituting Φ:

$$\frac{dQ}{dt} = \Phi \frac{A}{\Delta x} \Delta P_{H_2}^{1/2} \quad . \tag{6}$$

By knowing the steady-state flow rate, the area and thickness of the sample, and the hydrogen pressure difference across the sample, the permeability Φ may be calculated. A plot of Q vs t should be a straight line at steady state, and the slope is obviously dQ/dt, from which Φ may be easily calculated.

The permeability, like the diffusivity and solubility, is temperature-dependent and may be represented by an Arrhenius equation,

$$\Phi = \Phi_0 \exp(-E_\Phi/RT) \quad , \tag{7}$$

where

$$\Phi_0 = D_0 S_0 \quad , \tag{8}$$

2

and

$$E_\Phi = E_D + E_S \quad . \tag{9}$$

The E's are activation energies for each process. In the following discussions, we have normalized R so that the activation energies are independent of energy units and given in degrees Kelvin only. This process can be reversed, of course, by multiplying our energies (K) by the value of R in the desired energy units.

Data Review

The permeation data are listed by metal. Where there is a large body of data for a metal, the data are given in tabular form. Tabular data are listed in chronological order. Because permeation data have been published in many units, we have chosen a more rational, perhaps unfamiliar, SI-based unit, which is discussed above. For metals it is $mol/m \cdot s \cdot Pa^{1/2}$, and for nonmetals, $mol/m \cdot s \cdot Pa$. Once conversion is accomplished, data may be easily compared and various quantities calculated. Table A-1 in the appendix lists the factors to convert commonly encountered permeation units to $mol/m \cdot s \cdot Pa^{1/2}$. Tables A-2 and A-3 in the appendix give the selected "best" tritium and hydrogen permeabilities for quick reference.

Aluminum

There is a moderate-sized body of literature on the hydrogen/aluminum system. A large part of it, however, deals with diffusion and solubility. As Table 1 shows, of the five sets of permeability data, one (Boeschoeten *et al.*, 1960) is of little use, due to large scatter in the results. The other four divide into two pairs of comparable results. The results of Eichenauer *et al.* (1961) and Cochran (1961) have similar preexponentials, but widely varying activation energies. The results of Eichenauer and Pebler (1957) and Ihle *et al.* (1976) agree quite well, particularly when the former are

divided by the square root of the mass ratios (1.7). Ihle *et al.* combined their higher-temperature data (<400 K) with those of Cochran and compared the combined data with the data of Eichenhauer and Pebler. This combination shows the results to be more similar than are equations fit to each set of data.

At present we take the results of Ihle *et al.* as the best usable temperature-dependent permeability:

$$\Phi(T_2/Al) = 5.8 \times 10^{-5}$$

$$\times \exp{(-14{,}800/T)} \; mol/m \cdot s \cdot Pa^{1/2} \quad .$$

The above equation seems reliable at this point. However, surface conditions of aluminum are likely to strongly affect hydrogen permeability, particularly at lower temperatures, as Boeschoten *et al.* saw. A more thorough understanding of the effect of the surface condition of aluminum on hydrogen permeability is needed.

Beryllium

Jones and Gibson (1967) reported the only known results of tritium permeability, diffusivity, or solubility in beryllium. They loaded an arc-cast beryllium sample with tritium at several temperatures, then outgassed at higher temperatures. The temperature-dependent permeability obtained

Table 1. Aluminum permeability data.

Φ_0 (10^5 mol/m·s·Pa$^{1/2}$)	E_Φ (K)	Temperature range (K)	Pressure (kPa)	Gas	Reference
18	15,250	740-860	80	H_2	Eichenauer and Pebler (1957)
—	30,000	630-670	—	H_2, H_2O	Boeschoten *et al.* (1960)
0.38	18,900	630-870	25-100	H_2	Eichenauer *et al.* (1961)
0.45	12,360	670-870	100	H_2	Cochran (1961)
5.8	14,800	420-520	20	T_2	Ihle *et al.* (1976)

from the product of this measured diffusivity and solubility is the following, in our units:

$$\Phi(T_2/Be) = 5.8 \times 10^{-14}$$
$$\times \exp(-2220/T) \ \mathrm{mol/m \cdot s \cdot Pa^{1/2}} \ .$$

The activation energy is small, indicating very little change in permeability with temperature. We believe this result may be incorrect. Our experience with beryllium indicates that the permeability is lower, although our current samples were formed from sintered beryllium powder, which contained 8 wt% oxygen. We are attempting other experiments with disks of single-crystal beryllium.

More recently, Al'tovskiy et al. (1981) reported hydrogen permeability through sintered tubes and membranes. The presentation of their data is inconsistent and confusing. We have fitted their beryllium "tube" data (labeled in their report as Be1) to the following equation:

$$\Phi(H_2/Be) = 3.5 \times 10^{-7}$$
$$\times \exp(-10,600/T) \ \mathrm{mol/m \cdot s \cdot Pa^{1/2}} \ .$$

This activation energy is similar to the value they mention. Fits to other data in the article produce considerably different results. The activation energies vary from 3000 to 20,000 K, and the preexponentials vary from 10^{-2} to 10^{-11} $\mathrm{mol/m \cdot s \cdot Pa^{1/2}}$. In our analysis, we have assumed that their permeation unit of cm^3 (STP) $mm/cm^2 \cdot h$ included all of the usual variables except pressure, and were, therefore, permeabilities at the given pressure. Al'tovskiy et al. did show a definite proportional dependence of permeability on grain size, which may relate to differences between the work of Jones and Gibson and our own. The only common thread in these results is that all of the permeabilities of Al'tovskiy et al. were considerably higher than those of Jones and Gibson. The numerous inconsistencies make this work, as presented, very unreliable.

Fidelle et al. (1967) reported the permeability of hydrogen in beryllium at room temperature only. It is:

$$\Phi(H_2/Be, 20°C) = 2.5 \times 10^{-16} \ \mathrm{mol/m \cdot s \cdot Pa^{1/2}} \ .$$

The data from these three sources indicate a real need for a more thorough study of hydrogen permeability through beryllium.

Cadmium

As with beryllium, Fidelle et al. (1967) give a room-temperature permeability of hydrogen through cadmium of

$$\Phi(H_2/Cd, 20°C) = 4.1 \times 10^{-18} \ \mathrm{mol/m \cdot s \cdot Pa^{1/2}} \ .$$

This is the only result of which we are aware.

Cobalt

The sole measurement of hydrogen permeability through cobalt was reported by Caskey et al. in 1974. They found different permeabilities for the α and ϵ phases of cobalt. Their results with deuterium at 0.5 MPa, between 470 and 820 K are:

$$\Phi(D_2/\epsilon\text{-Co}, <670 \ K) = 6.3 \times 10^{-9}$$
$$\times \exp(-6850/T) \ \mathrm{mol/m \cdot s \cdot Pa^{1/2}}$$

$$\Phi(D_2/\alpha\text{-Co}, >670 \ K) = 3.8 \times 10^{-8}$$
$$\times \exp(-7750/T) \ \mathrm{mol/m \cdot s \cdot Pa^{1/2}} \ .$$

Copper

There have been numerous measurements of hydrogen and deuterium permeability through copper (Table 2). All measurements, except those of Eichenauer and his colleagues, were made using the membrane technique. Eichenauer and colleagues measured diffusivity and solubility separately. Relatively pure copper was used in all experiments. The values of Perkins and Begeal (1972) and Gorman and Nardella (1962) probably represent the best values for hydrogen permeability. These numbers should be good over a wide range of temperature (400–900 K) and pressure (0.1–200 kPa).

Copper Alloys

Begeal (1978) has measured twenty hydrogen and deuterium permeabilities in several copper alloys, including aluminum, silicon, and phosphate bronzes, and copper-beryllium. Several gold-copper brazing alloys were also examined.

Table 2. Copper permeability data.

Φ_0 (10^7 mol/m·s·Pa$^{1/2}$)	E_Φ (K)	Temperature range (K)	Pressure (kPa)	Gas	Reference
0.864	8240	500–730	0.017	H_2	Smithells and Ransley (1935)
8.07	9950	420–600	0.1	H_2	Braaten and Clark (1936)
4.1	8960	530–920	–	H_2	Eichenauer and Pebler (1957)
0.0825	6290	620–770	0.15 and 0.2	H_2	Rudd *et al.* (1961)
5.86	9100	620–970	0.1	H_2	Gorman and Nardella (1962)
52.0	9470	700–900	0.013–0.1	H_2	Eichenauer *et al.* (1965)
32.7	9630	700–900	0.013–0.1	D_2	Eichenauer *et al.* (1965)
8.42	9290	470–710	0.00013–0.13	H_2	Perkins and Begeal (1972)
0.58	8620	375–750	0.1 and 0.5	D_2	Caskey *et al.* (1976)
8.4	9320	470–700	0.00013–0.13	H_2	Begeal (1978)

Outside of the high-gold-content alloys, the preexponentials were in the range 1–9 $\times$ 10^{-7} mol/m·s·Pa$^{1/2}$, and the activation energies are near $-10,000$ K. Temperatures were between 600 and 900 K and pressures in the 10–300 kPa range. See Begeal (1978) for exact values.

Caskey and Derrick (1977) measured the hydrogen permeabilities through several alpha-brasses with up to 35 wt% zinc. There was no consistency in the changes in the preexponential or activation energies with increasing zinc content. A graph of the permeabilities vs temperature indicated that the least squares permeabilities generally decreased with increasing zinc content.

Germanium

The only hydrogen permeability data for germanium were given by Van Wieringen and Warmoltz (1956). Their permeation equation, converted to our units, is

$$\Phi(H_2/Ge) = 1.2 \times 10^{-5}$$

$$\times \exp\left(-24,000/T\right) \text{ mol/m·s·Pa}^{1/2} \quad .$$

Their measurements were taken on a hollow single crystal between 1040 and 1200 K at 1 atm hydrogen pressure, using a modified time-lag method.

Gold

There are two measurements of hydrogen permeation through gold. The results of Caskey and Derrick (1976) and Begeal (1978) are very sim-

ilar. They both used deuterium and the time-lag method, and covered comparable pressure and temperature ranges. Caskey and Derrick's results are given by

$$\Phi(D_2/Au) = 3.1 \times 10^{-6}$$

$$\times \exp\left(-14,800/T\right) \text{ mol/m·s·Pa}^{1/2} \quad ,$$

and Begeal's are given by

$$\Phi(D_2/Au) = 1.14 \times 10^{-6}$$

$$\times \exp\left(-13,800/T\right) \text{ mol/m·s·Pa}^{1/2} \quad .$$

Either equation could be chosen for calculations.

Iron

Measurements of hydrogen permeability in iron are numerous. We found almost 20 references. Thirteen of these were reviewed by Gonzalez (1967). Only the most recent reports, including Gonzalez's review, are enumerated here (Table 3). The results are remarkably similar. The value given by Gonzalez,

$$\Phi(H_2/Fe) = 4.1 \times 10^{-8}$$

$$\times \exp\left(-4200/T\right) \text{ mol/m·s·Pa}^{1/2} \quad ,$$

is likely the most suitable for calculation, because it represents an average of many measurements. Small impurities and grain size seemingly have no significant effect on the permeability of hydrogen through iron.

Table 3. Iron permeability data.

Φ_0 $(10^8 \text{ mol/m} \cdot \text{s} \cdot \text{Pa}^{1/2})$	E_Φ (K)	Temperature range (K)	Pressure (kPa)	Gas	Reference
4.1	4200	>375	up to 10 MPa	H_2	Gonzalez (1967)
3.58	4100	330-770	10-1000	H_2	Nelson and Stein (1973)
19.5	4700	260-700	20-500	H_2	Louthan et al. (1975)
8.13	4700	260-700	20-500	T_2	Louthan et al. (1975)
8.63	4300	480-875	10	H_2	Wagner and Sizmann (1975)
6.78	4300	275-335	1-100	H_2	Kumnick and Johnson (1975)
4.98	4217	282-346	1-100	H_2	Kumnick and Johnson (1977)
6.60	4290	322-783	0.4-60	H_2	Quick and Johnson (1978)

Lead

We could only find one measurement of hydrogen permeation through lead: the early work of Deming and Hendricks (1923). They give three data points, at 530, 540, and 555 K. Our fit to this equation is

$$\Phi(H_2/Pb) = 0.121$$
$$\times \exp\left(-11,900/T\right) \text{ mol/m} \cdot \text{s} \cdot \text{Pa}^{1/2} .$$

At 550 K this gives a permeability of 4×10^{-11} mol/m·s·Pa$^{1/2}$. We feel this is too high compared with other low-permeability materials, and therefore we would not use the above equation.

Molybdenum

Many authors have measured the permeability of hydrogen through molybdenum. No tritium data exist. Table 4 lists ten references. In addition to the amount of data available, the similarity of the results is remarkable. A review by Chandler and Walter (1968) gives an average value of permeability, from several works, of:

$$\Phi(H_2/Mo) = 2.3 \times 10^{-7}$$
$$\times \exp\left(-9710/T\right) \text{ mol/m} \cdot \text{s} \cdot \text{Pa}^{1/2} .$$

The three later reports do not contradict this "best fit" to any significant degree. The various

Table 4. Molybdenum permeability data.

Φ_0 $(10^7 \text{ mol/m} \cdot \text{s} \cdot \text{Pa}^{1/2})$	E_Φ (K)	Temperature range (K)	Pressure (kPa)	Gas	Reference
3.59	10,100	820-1670	0.1-10	H_2	Smithells and Ransley (1935)
22.7	12,280	850-1250	100	H_2	Hill (1960)
1.65	9,150	900-1500	100	H_2	Huffine and Williams (1960)
4.0	11,000	900-1200	100	H_2	Steigerwald (1963)
2.4	9,660	520 and 620	85	H_2	Jones et al. (1966)
3.62	10,800	1100-2000	0.3 and 30	H_2	Frauenfelder (1968)
2.33	9,710	500-1700	−	H_2	Chandler and Walter (1968)
1.4[a]	9,300	679-1270	0.1-30	H_2	Zakharov et al. (1973)
3.6[b]	9,700				
5.3	8,760	540-910	0.1-300	H_2	Gutherie et al. (1974)
3.22	9,980	523-730	100	H_2	Caskey et al. (1975)

[a] Single-crystal molybdenum.

[b] Polycrystalline molybdenum.

results span wide ranges of temperature (500–1700 K) and pressure (0.1–300 kPa). With one exception, the samples were polycrystalline, and the "time-lag" method was used with tubes or membranes.

Nickel

As with iron, there has been considerable study of hydrogen permeation through nickel (Table 5). The many results are comparable, except for those of Eichenauer et al. (1965) and Tanabe et al. (1977). The permeabilities given by either Ebisuzaki et al. (1967), Robertson (1973), or Louthan et al. (1975, 1976) could be used in calculations.

Niobium

Clean niobium reacts readily and exothermically with hydrogen. Having such a negative heat of solution, it is different from the endothermic resistant metals, whose hydrogen solubility increases with temperature. Diffusivity and solubility are oppositely influenced by temperature, as is the case for palladium and the other two Group VB metals, vanadium and tantalum. There are no direct measurements of hydrogen permeation through niobium, so we multiply reliable diffusivity and solubility measurements together. Völkl and Alefeld (1975) summarize six measurements of diffusivity over the temperature range −50 to 600°C. The data are self-consistent, with an average value of:

$$D = 5.0 \times 10^{-8} \exp(-1230/T) \; m^2/s \quad .$$

Converting the solubility data of Veleckis and Edwards (1969) to our units, we have the α-phase hydrogen solubility in niobium:

$$S(H_2/Nb) = 0.126 \exp(4240/T) \; mol/m^3 \cdot Pa^{1/2} \quad .$$

Steward (1975) reports both hydrogen and deuterium solubilities in niobium. His solubility for hydrogen, again translated into the same units, is similar to that of Veleckis and Edwards and is

$$S(H_2/Nb) = 0.127 \exp(5550/T) \; mol/m^3 \cdot Pa^{1/2} \quad .$$

Calculation of permeation from combining these two results with Völkl and Alefeld's gives

$$\Phi(H_2/Nb) = 6.3 \times 10^{-9}$$
$$\times \exp(3010/T) \; mol/m \cdot s \cdot Pa^{1/2}$$

for the results of Veleckis and Edwards, and

$$\Phi(H_2/Nb) = 6.3 \times 10^{-9}$$
$$\times \exp(4270/T) \; mol/m \cdot s \cdot Pa^{1/2}$$

for the results of Steward. The results are similar, and either can be used for calculations. Caution should be exercised in using these numbers at high hydrogen pressures or low temperatures, where hydride formation or nonideality may occur. Pressures below 1×10^4 Pa and temperatures above 400°C should be observed.

Table 5. Nickel permeability data.

Φ_0 (10^7 mol/m·s·Pa$^{1/2}$)	E_Φ (K)	Temperature range (K)	Pressure (kPa)	Gas	Reference
30.0	6670	650–873	100	H_2	Ham (1933)
4.65	6640	675–1125	100	H_2	Gorman and Nardella (1962)
6.73	4770	650–920	10–100	H_2	Eichenauer et al. (1965)
4.76	4600	650–920	10–100	D_2	Eichenauer et al. (1965)
4.06	6623	480–690	100	H_2	Ebisuzaki et al. (1967)
3.22	6590	300–775	1–100	H_2	Robertson (1973)
3.9	6660	300–550	40–400	H_2	Louthan et al. (1975)
1.54	6500	300–650	0.1–30	T_2	Louthan and Derrick (1976)
10.4	8300	600–900	5–10	H_2	Louthan and Derrick (1976)
3.33	7350	600–900	5–10	D_2	Tanabe et al. (1977)
2.38	6290	600–800	<12 MPa	H_2	Gala et al. (1981)

Palladium

The palladium-hydrogen system is probably the most studied of any metal-hydrogen system. At least 25 measurements of the diffusivity are available. The more reliable results are quite consistent. The palladium-hydrogen phase diagram and thermodynamic properties have also been heavily studied, as evidenced by Lewis's (1967) review book. Despite this wealth of data, we know of only two permeability results. The first report was by Koffler *et al.* (1969). In our units, they gave the permeability as

$$\Phi(H_2/Pd) = 2.20 \times 10^{-7}$$

$$\times \exp\left(-1885/T\right) \text{ mol/m} \cdot \text{s} \cdot \text{Pa}^{1/2}$$

from 0.04 to 7 Pa and from 300 to 709 K.

Balovnev (1974) erroneously compared his later results to Koffler *et al.*'s, but they are nonetheless similar. His value of permeability is:

$$\Phi(H_2/Pd) = 8.11 \times 10^{-7}$$

$$\times \exp\left(-1860/T\right) \text{ mol/m} \cdot \text{s} \cdot \text{Pa}^{1/2} \quad .$$

His results covered the ranges 3×10^{-5} to 7×10^{-2} Pa, and 370 to 900 K. We have some preference for the work of Koffler *et al.* because they seemed to do a more reliable data analysis and comparison.

Platinum

There are surprisingly few data for hydrogen permeability through platinum. We could find only four references (Table 6). The results of Ebisuzaki *et al.* (1968) and Katsuta and McLellan (1979) seem the most reliable in that they are similar and the samples were of very high purity.

The reported permeabilities of Katsuta and McLellan are half the value of the product of the diffusivity and solubility fitted to their data. This product, including both Φ_0 and E_Φ, corresponds quite well to the results of Ebisuzaki *et al.* The results of Ham (1933) approximate the others and may differ because of the limited vacuums and materials purity available 50 years ago. The results of Ebisuzaki *et al.* would be a good choice for calculations. Combined with Katsuta and McLellan's results, Ebisuzaki *et al.*'s permeability is useful over a wide range of temperature (500–1200 K) and pressure (10^{-8}–100 kPa).

Solubility data in a super-pure platinum single crystal were measured by Yei and McLellan (1979) but were greater by about one order of magnitude over a similar temperature range than previous work from the same laboratory (Katsuta and McLellan).

Silicon

Van Wieringen and Warmoltz (1956) provide the only hydrogen permeation data through pure silicon and germanium, as mentioned earlier. The measurements were with single-crystal silicon between 1240 and 1485 K at 1 atm hydrogen. They also measured the helium permeabilities in these semiconductors. A least-squares fit of their permeability data gives the following equation, in our units and allowed accuracy:

$$\Phi(H_2/Si) = 1.4 \times 10^{-5}$$

$$\times \exp\left(-27,000/T\right) \text{ mol/m} \cdot \text{s} \cdot \text{Pa}^{1/2} \quad .$$

Ichimiya and Furuichi (1968) measured tritium diffusivity between 670 and 770 K and

Table 6. Platinum permeability data.

Φ_0 (10^7 mol/m·s·Pa$^{1/2}$)	E_Φ (K)	Temperature range (K)	Pressure (kPa)	Gas	Reference
8.1	9400	705–827	100	H_2	Ham (1933)
1.2	8500	540–900	70	H_2	Ebisuzaki *et al.* (1968)
0.58 ± 0.15	7700	830–1210	10^{-6}–10^{-8}	H_2	Katsuta and McLellan (1979)
1	8300	830–1210	10^{-6}–10^{-8}	H_2	Katsuta and McLellan (1979)
0.165	6910	725–875	2×10^5	H_2	Harvie *et al.* (1980)

solubility between 1200 and 1500 K in boron-doped (p-type) silicon, using the absorption outgassing method. The calculated permeability from their combined diffusivity and solubility is

$$\Phi(H_2/Si[B]) = 1.8 \times 10^{-9}$$

$$\times \exp(-24{,}000/T) \ \text{mol/m·s·Pa}^{1/2} \ .$$

The two results differ by about 1000 in the temperature range measured. The low results of Ichimiya and Furuichi may be due to the boron doping or oxides. Extrapolating either result to low temperatures is a mistake, even though the two results agree well at room temperature.

Silver

Zvedin and Belyakov (1967) reported the only direct permeability data for hydrogen through silver. Unfortunately, the data are presented only graphically. A calculation of the slope and intercept of their data yields the following results, in our standard units:

$$\Phi(H_2/Ag) = 3 \times 10^{-8}$$

$$\times \exp(-7350/T) \ \text{mol/m·s·Pa}^{1/2} \ .$$

This equation does not correspond to a permeation constant at 930 K (660°C) given in a table in the same paper, thus lending suspicion to their results.

Katsuta and McLellan (1979) measured the diffusivity of hydrogen in silver with a membrane permeability apparatus. They did not report, though, the permeability results they obtained. By combining these diffusivity results with solubility results reported earlier on different samples by McLellan (1973), who obtained them by another technique, we have perhaps a more reliable permeability value:

$$\Phi(H_2/Ag) = 2.21 \times 10^{-7}$$

$$\times \exp(-10{,}440/T) \ \text{mol/m·s·Pa}^{1/2} \ .$$

We have some faith in this value because it agrees within a factor of 5 with our own measurement of tritium permeation through silver at 100°C.

Tantalum

As with the other VB metals, there are no reported measurements of hydrogen permeation through tantalum. As we did for those metals, we combine the solubility data of Veleckis and Edwards (1969) with the selected data of Völkl and Alefeld (1975) to obtain the permeability:

$$S(H_2/Ta) = 0.132 \exp(4050/T) \ \text{mol/m}^3\text{·Pa}^{1/2} \ ,$$

$$D(H_2/Ta) = 4.4 \times 10^{-8} \exp(-1620/T) \ \text{m}^2/\text{s} \ ,$$

$$\Phi(H_2/Ta) = 5.8 \times 10^{-9}$$

$$\times \exp(2430/T) \ \text{mol/m·s·Pa}^{1/2} \ .$$

As with the other VB metals, caution is advisable in using this formula. Pressure and temperature ranges below 10 kPa and above 350°C are appropriate to avoid nonlinearity and the two-phase region.

Tungsten

Three of the five tungsten permeation experiments we have collected agree reasonably well, both in the preexponential and the activation energy (Table 7). The earliest (Steigerwald, 1963) and latest results (Zakharov et al., 1973) disagree the most. We favor Frauenfelder's work (1968, 1969) and that of Aitken et al. (1967) because they cover the widest temperature and pressure ranges and the results are similar. We choose Aitken

Table 7. Tungsten permeability data.

Φ_0 (10^7 mol/m·s·Pa$^{1/2}$)	E_Φ (K)	Temperature range (K)	Pressure (kPa)	Gas	Reference
0.0084	11,000	970–1150	100	H$_2$	Steigerwald (1963)
7.77	17,000	1600–2900	10–100	H$_2$	Aitken et al. (1967)
7.6	15,900	1100–2400	10^{-7}–13	H$_2$	Frauenfelder (1968)
6.0	16,600	1100–2400	Degas. expt	H$_2$	Frauenfelder (1969)
0.73	12,800	1000–1330	0.1–30	H$_2$	Zakharov et al. (1973)

et al.'s result as the "best" value below, because it falls between the two results of Frauenfelder:

$$\Phi(H_2/W) = 7.8 \times 10^{-7}$$
$$\times \exp(-17,000/T) \text{ mol/m} \cdot s \cdot Pa^{1/2} \quad .$$

Vanadium

As with niobium and tantalum, no direct measurements of hydrogen permeation through vanadium are available. We combine the solubility data of Veleckis and Edwards (1969),

$$S(H_2/V) = 0.138 \exp(3490/T) \text{ mol/m}^3 \cdot Pa^{1/2} \quad ,$$

with the average diffusivity values given by Völkl and Alefeld (1975),

$$D(H_2/V) = 2.9 \times 10^{-8} (-500/T) \text{ m}^2/s \quad ,$$

and obtain a permeability:

$$\Phi(H_2/V) = 4.0 \times 10^{-9}$$
$$\times \exp(2990/T) \text{ mol/m} \cdot s \cdot Pa^{1/2} \quad .$$

This data is for the α-phase V/H_2 system and should be used at pressures less than 1×10^4 Pa and greater than 300°C.

Zinc

We could find no satisfactory work on zinc. Hendricks and colleagues (Hendricks and Ralston, 1929; Deming and Hendricks, 1923) used the membrane method between 600 and 700 K. The temperature dependence is not clear. A permeation value at 650 K is:

$$\Phi(H_2/Zn, 650 \text{ K}) = 5 \times 10^{-12} \text{ mol/m} \cdot s \cdot Pa^{1/2} \quad .$$

We believe that this number is not reliable, though. Moon (1971) measured diffusion coefficients between 325 and 525 K and 5 and 100 kPa hydrogen using gas-volumetric measurements. His results were:

$$D(H_2, 99.99\% \text{ Zn}) = 5.8 \times 10^{-7}$$
$$\times \exp[(-7000 \pm 100)T] \text{ m}^2/s \quad ,$$

and

$$D(H_2, 99.9\% \text{ Zn}) = 4.2 \times 10^{-7}$$
$$\times \exp[(-11,100 \pm 100)T] \text{ m}^2/s \quad .$$

Other Pure Metals

We did not find reports of hydrogen permeabilities for any of the following pure metals: antimony, arsenic, bismuth, boron, chromium, indium, iridium, manganese, osmium, rhenium, rhodium, ruthenium, tin, and thallium. Other metals were considered strong hydride formers.

Hydrogen solubilities in rhodium, ruthenium, and iridium were measured by McLellan and Oates (1973). Sieverts and Moritz measured the hydrogen solubility in manganese (1937).

Iron and Nickel Alloys

Because iron and nickel alloys are such an important class of materials, we reviewed them. The results are divided between stainless steels, other iron alloys such as Kovar and Ceramvar, and nickel-rich alloys (Tables 8 through 10).

Considering the wide variety of composition and microstructure, there is not a large difference in preexponentials or activation energies for the stainless alloys. The preexponential is in the 10^{-7} mol/m $\cdot$ s $\cdot$ Pa$^{1/2}$ range, and the activation energies are near -8000 K. The carbon steels as a group have a lower preexponential by about a factor of 10 and half the activation energy.

Ryabov, Salii, and Gel'd have measured permeabilities of several iron alloys with up to 10 wt% germanium (Ryabov *et al.*, 1976) and silicon (Salii *et al.*, 1973). The results were similar to those for iron. The silicon-containing alloys had decreasing preexponentials and increasing activation energies with increasing silicon content. With the germanium-containing alloys the preexponential increased; this was an unexpected result, since silicon and germanium have similar hydrogen permeabilities.

Glasses

Souers *et al.* (1978) have reviewed the literature for hydrogen isotope permeability through

Table 8. Stainless steel permeability data.

Φ_0 $(10^7 \text{ mol/m} \cdot \text{s} \cdot \text{Pa}^{1/2})$	E_Φ (K)	Temperature range (K)	Pressure (kPa)	Gas	Alloy	Reference
3.5	8200	775-1175	100	H_2	303	Gorman and Nardella (1962)
4.9	8600	825-1175	100	H_2	304	Gorman and Nardella (1962)
19.5	8660	375-575	100	H_2	304	Chaney and Powell (1970), Powell *et al.* (1970)
10.4	7700	350-900	10-3000	H_2	304	Nelson and Stein (1973)
1.2	7100	425-875	1-40	H_2	309S	Kass and Andrzejewski (1972)
1[a]	8000	500-700	100	D_2	309S	Swansiger *et al.* (1974)
2.36	7650	350-1050	10^{-3}-10^{-4}	H_2	316	Van Deventer and Maroni (1980)
3.1	7800	575-1075	10-1000	H_2	321	Phillips and Dodge (1968)
0.853	7150	350-1050	10^{-3}-10^{-4}	H_2	321	Van Deventer and Maroni (1980)
1.38	7932	370-700	1-10	T_2	21-6-9	Swansiger (1976)

[a] Average of several results.

Table 9. Permeability data of other iron alloys.

Φ_0 $(10^7 \text{ mol/m} \cdot \text{s} \cdot \text{Pa}^{1/2})$	E_Φ (K)	Temperature range (K)	Pressure (kPa)	Gas	Alloy	Reference
3.15	8050	775-1225	100	H_2	Kovar	Gorman and Nardella (1962)
0.5	4300	600-900	100	H_2	Cold-drawn steel	Gorman and Nardella (1962)
1.11	7000	450-775	10-100	H_2	Ceramvar	Perkins and Begeal (1972)
0.291	4800	350-900	10-3,000	H_2	4130 normal	Nelson and Stein (1973)
0.364	4340	350-900	10-3,000	H_2	4130 quenched/tempered	Nelson and Stein (1973)
3 to 1 $\times 10^{-8}$	4250-5500	600-1000	–	H_2	Fe, 0-10 wt% Si	Salii *et al.* (1973)
3 to 4 $\times 10^{-8}$	4250-5000	600-1000	–	H_2	Fe, 0-10 wt% Ge	Ryabov *et al.* (1976)

Table 10. Nickel alloy permeability data.

Φ_0 $(10^7 \text{ mol/m} \cdot \text{s} \cdot \text{Pa}^{1/2})$	E_Φ (K)	Temperature range (K)	Pressure (kPa)	Gas	Alloy	Reference
9.87	8400	775-1175	100	H_2	Inconel 600	Gorman and Nardella (1962)
2.03	6440	725-1225	100	H_2	Monel 400	Gorman and Nardella (1962)
2.12	7570	500-1000	0.01-10	H_2	Haynes 188	Van Deventer (1977)
2.56	7240	400-1000	0.1-10	H_2	Inconel 625	Van Deventer and Maroni (1980)
0.959	6780	400-1000	0.1-10	H_2	Inconel 718	Van Deventer and Maroni (1980)

glasses. Following the lead of Altemose (1961), they included the mole percent M of non-network-forming or modifier oxides, e.g., Na_2O, CaO, and MgO, as variables affecting hydrogen permeability over a limited range in both the preexponential and activation energy. Doremus (1961) and Shelby (1972a, b) concluded that the permeability varies linearly as well as exponentially with temperature, giving the modified temperature-dependent equation:

$$\Phi = \Phi_0 T \exp\left(-E_\Phi/T\right) \ . \tag{10}$$

The units of permeability through glasses and ceramics are $\text{mol/m} \cdot \text{s} \cdot \text{Pa}$, since several results have shown first-power dependence of permeability on pressure, unlike the metals. This is readily explained by the lack of hydrogen dissociation in the nonmetals, as compared with metals. The preexponential term Φ_0 has in this form the unusual units $\text{mol/m} \cdot \text{s} \cdot \text{Pa} \cdot \text{K}$.

We repeat the summarized results of Souers *et al.* in Table 11 since they already represent a compilation of a wide body of data. Their results cover data that were taken over large ranges of temperature (300–1500 K) and pressure (100 Pa–1 GPa). Because of the scatter in the available results, their best-fit equations are as accurate as can be expected.

Table 11.   Glass permeability data.

Φ_0 $(10^{17} \text{ mol/m} \cdot \text{s} \cdot \text{Pa} \cdot \text{K})$	E_Φ (K)	Gas	References
Silicate glasses			
$\left[3.4 + (8 \times 10^{-4}) \, M^3 \right]^a$	$3600 + 165 \, M$	H_2, D_2	Altemose (1961), Doremus (1961), Shelby (1972a, b)
Soda-lime glasses			
9	6700	H_2, D_2	Altemose (1961), Doremus (1961), Shelby (1972a, b)
20	5600	H_2	Souers *et al.* (1978)
4	5600	D_2, DT	Souers *et al.* (1978)

[a] M is mole percent of non-network-forming or modifier oxides.

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Appendix

Table A-1 lists the factors to convert commonly encountered permeation units to $mol/m \cdot s \cdot Pa^{1/2}$. Tables A-2 and A-3 give the selected "best" tritium and hydrogen permeabilities for quick reference.

Table A-1. Permeation rate conversion factors.

To convert to $mol/m \cdot s \cdot Pa^{1/2}$	
Multiply these units	**by these factors:**
$\dfrac{cm^3 \ (STP)}{cm \cdot s \cdot atm^{1/2}}$	1.4015×10^{-5}
$\dfrac{cm^3 \ (STP) \cdot mm}{cm^2 \cdot s \cdot atm^{1/2}}$	1.4015×10^{-6}
$\dfrac{cm^3 \ (STP) \cdot mm}{cm^2 \cdot min \cdot atm^{1/2}}$	2.3358×10^{-8}
$\dfrac{cm^3 \ (STP) \cdot mm}{cm^2 \cdot h \cdot atm^{1/2}}$	3.8932×10^{-10}
$\dfrac{cm^3 \ (STP)}{cm \cdot s \cdot (torr)^{1/2}}$	3.8637×10^{-4}
$\dfrac{cm^3 \ (STP) \cdot mm}{cm^2 \cdot s \ (torr)^{1/2}}$	3.8637×10^{-5}
$\dfrac{torr \cdot liter}{cm \cdot s \cdot (torr)^{1/2}}$	5.096×10^{-4}
$\dfrac{\mu m(Hg) \cdot liter}{cm \cdot s \cdot atm^{1/2}}$	1.8441×10^{-8}

To convert from $mol/m \cdot s \cdot Pa^{1/2}$ to the units above, divide by the accompanying factor.

Table A-2. "Best" tritium permeability values for metals.

Metal	$\Phi_0{}^a$ $(mol/m \cdot s \cdot Pa^{1/2})$	E_Φ (K)	Temperature range (K)	Pressure	Ref. gas	References
Aluminum	5.8×10^{-5}	14,800	420-520	20 kPa	T_2	Ihle *et al.* (1976)
Beryllium	5.8×10^{-14}	2,220	670-1170	Outgassing	T_2	Jones and Gibson (1967)
Cobalt	5.1×10^{-9} (ϵ-Co)	6,850	<670	0.5 MPa	D_2	Caskey *et al.* (1974)
	3.1×10^{-8} (α-Co)	7,750	>670	0.5 MPa	D_2	Caskey *et al.* (1974)
Copper	4.9×10^{-7}	9,320	470-700	0.13-130 kPa	H_2	Begeal (1978)
Germanium	7×10^{-6}	24,000	1040-1200	100 kPa	H_2	Van Wieringen and Warmoltz (1956)
Gold	2.5×10^{-6}	14,800	500-900	100-500 kPa	D_2	Caskey and Derrick (1976)
Iron	2.4×10^{-8}	4,200	>375	<10 MPa	H_2	Gonzalez (1967)[b]
Lead	–	–	–	–	–	Deming and Hendricks (1923)[c]
Molybdenum	1.3×10^{-7}	9,710	500-1700	–	H_2	Chandler and Walter (1968)
Nickel	1.54×10^{-7}	6,500	300-650	0.1-30 MPa	T_2	Louthan and Derrick (1976)
Niobium	3.6×10^{-9}	−4,270	>700	<10 kPa	H_2	Völkl and Alefeld (1975), Steward (1975)
Palladium	1.3×10^{-7}	1,885	300-709	0.04-7 Pa	H_2	Koffler *et al.* (1969)
Platinum	6.9×10^{-8}	8,500	540-900	70 kPa	H_2	Ebisuzaki *et al.* (1968)
Silicon	8×10^{-6}	27,000	1240-1485	100 kPa	H_2	Van Wieringen and Warmoltz (1956)
Tantalum	3.3×10^{-9}	−2,430	>650	<10 kPa	H_2	Völkl and Alefeld (1975), Veleckis and Edwards (1969)
Tungsten	4.5×10^{-6}	17,000	1100-2400	10-100 kPa	H_2	Aitken *et al.* (1967)
Vanadium	2.3×10^{-9}	−2,990	>600	<10 kPa	H_2	Völkl and Alefeld (1975), Veleckis and Edwards (1969)

[a] $\Phi_0(T_2)$ adjusted with $\sqrt{m_{H_2} m_{D_2}}/\sqrt{m_{T_2}}$ mass ratios.

[b] The value for T_2 of Louthan *et al.* (1975) seems too high.

[c] Data unreliable.

Table A-3. "Best" hydrogen permeability values for metals.

Metal	$\Phi_0{}^a$ (mol/m·s·Pa$^{1/2}$)	E_Φ (K)	Temperature range (K)	Pressure	Ref. gas	References
Aluminum	5.8×10^{-5}	14,800	420-520	20 kPa	T_2	Ihle *et al.* (1976)
Beryllium	5.8×10^{-14}	2,220	670-1170	Outgassing	T_2	Jones and Gibson (1967)
Cobalt	6.3×10^{-9} (ϵ-Co)	6,850	<670	0.5 MPa	D_2	Caskey *et al.* (1974)
	3.8×10^{-8} (α-Co)	7,750	>670	0.5 MPa	D_2	Caskey *et al.* (1974)
Copper	8.4×10^{-7}	9,320	470-700	0.13-130 kPa	H_2	Begeal (1978)
Germanium	1.2×10^{-5}	24,000	1040-1200	100 kPa	H_2	Van Wieringen and Warmoltz (1956)
Gold	3.1×10^{-6}	14,800	500-900	100-500 kPa	D_2	Caskey and Derrick (1976)
Iron	4.1×10^{-8}	4,200	>375	<10 MPa	H_2	Gonzalez (1967)
Lead	–	–	–	–	–	Deming and Hendricks (1923)[a]
Molybdenum	2.3×10^{-7}	9,710	500-1700	–	H_2	Chandler and Walter (1968)
Nickel	4×10^{-7}	6,600	300-775	0.1-400 kPa	H_2	Ebisuzaki *et al.* (1967), Robertson (1973), Louthan *et al.* (1975).
Niobium	6.3×10^{-9}	-4,270	>700	<10 kPa	H_2	Völkl and Alefeld (1975), Steward (1975)
Palladium	2.20×10^{-7}	1,885	300-709	0.04-7 Pa	H_2	Koffler *et al.* (1969)
Platinum	1.2×10^{-7}	8,500	540-900	70 kPa	H_2	Ebisuzaki *et al.* (1968)
Silicon	1.4×10^{-5}	27,000	1240-1485	100 kPa	H_2	Van Wieringen and Warmoltz (1956)
Tantalum	5.8×10^{-9}	-2,430	>650	<10 kPa	H_2	Völkl and Alefeld (1975), Veleckis and Edwards (1969)
Tungsten	7.8×10^{-7}	17,000	1100-2400	10-100 kPa	H_2	Aitken *et al.* (1967)
Vanadium	4.0×10^{-9}	-2,990	>600	<10 kPa	H_2	Völkl and Alefeld (1975), Veleckis and Edwards (1969)

[a] Data unreliable.

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