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D. K. Benson, T. F. Potter, C. E. Tracy
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David K. Benson
Thomas F. Potter
C. Edwin Tracy

National Renewable Energy Laboratory
Golden, CO 80401

ABSTRACT

This paper describes one approach to the design of a variable-conductance vacuum insulation. In this design, the vacuum insulation consists of a permanently sealed, thin sheet steel, evacuated envelope of whatever geometry is required for the application. The steel envelope is supported internally against the atmospheric pressure loads by an array of discrete, low-conductance, ceramic supports, and radiative heat transfer is blocked by layers of thin metal radiation shields. Thermal conductance through this insulation is controlled electronically by changing the temperature of a small metal hydride connected to the vacuum envelope. The hydride reversibly absorbs/desorbs hydrogen to produce a hydrogen pressure typically within the range from less than 10^{-6} to as much as 1 torr. Design calculations are compared with results from laboratory tests of bench scale samples, and some possible automotive applications for this variable-conductance vacuum insulation are suggested.

INTRODUCTION

VACUUM INSULATION — Vacuum insulation is commonly used in spherical or cylindrical containers ranging in size from lunch bottles to over-the-road liquified gas transport tankers. In such geometries, the container has sufficient strength to support the atmospheric pressure loads, and it is unnecessary to provide strong internal supports between the inner and outer walls of the evacuated envelope. The evacuated envelope may be completely empty, as in the lunch bottle, or filled with a compacted, multi-layer wrap of thin aluminum foil separated by polymer or glass fiber spacers, as in the liquified gas tank. The vacuum is permanent and the insulation value is fixed.

But what if the insulated container surrounds a system which must be insulated part of the time and must reject or absorb heat from the outside at other times? Usually, such an application would require a heat exchange loop to pass through the container. Such a loop would add heat loss paths through the container, com-

promising its insulating performance and complicating the system design.

VARIABLE-CONDUCTANCE VACUUM INSULATION — Variable-conductance vacuum insulation is a form of evacuated thermal insulation which can be made conductive by introducing low-pressure hydrogen gas. The pressure of hydrogen is electrically controlled by heating a small container of metal hydride such as zirconium hydride. Such hydrides produce thermodynamically determined, equilibrium hydrogen pressures that are a predictable function of the hydride temperature.

Hydrogen gas has a very high thermal conductivity, even at pressures of less than 10^{-5} atm, so that a very small amount of hydrogen is sufficient to fill a large vacuum-insulation enclosure and make it thermally conductive. When the hydride is allowed to cool, it will quickly reabsorb the hydrogen, reestablishing the vacuum and restoring the thermal resistance of the insulation. Control of the thermal conductance is continuously adjustable by controlling the hydride temperature.

This paper describes some of the design considerations for a variable-conductance insulation (VCI) and suggests some possible applications in the automotive industry. Topics covered include the thermal conductivity of low pressure hydrogen, the dependence of vacuum insulation thermal conductance upon the pressure of internal hydrogen gas, the quantity of gas required, characteristics of selected metal hydrides and their required quantity, and remaining problems of manufacturing VCI. The design calculations are compared with test results from a bench-scale VCI panel.

DESIGN ANALYSIS

THERMAL CONDUCTIVITY OF LOW-PRESSURE HYDROGEN GAS - Hydrogen is a low molecular weight, diatomic gas which has a very high thermal conductivity. The elementary, kinetic theory of gas dynamics is adequate to accurately calculate the thermal conductivity of low-pressure hydrogen at temperatures above about 100 K [1]. At atmospheric pressure, the conductivity of

hydrogen is 0.173 (W/mK), compared to air at 0.024 and helium at 0.142 [2].

At low pressures, the gas molecules can move uninhibited from a hot surface where they pick up heat energy to a cooler surface where they deposit this energy. Adding more molecules increases the pressure and the amount of thermal power which is conducted between the warm and cool surfaces. Consequently, the thermal power conducted by a gas across an area, A, at low pressure is proportional to the pressure:

$$\text{Power}_{LP} = P * \alpha * \kappa * (T_{hot} - T_{cold}) * \frac{A}{Z} \quad (1)$$

where Z is the thickness of the gas-filled gap and

$$\alpha = (\alpha_1 \alpha_2) / (\alpha_1 + \alpha_2 - \alpha_1 \alpha_2) \quad (2)$$

is a measure of the efficiency with which the gas molecules exchange energy with the solid surfaces. The α_1 and α_2 are called accommodation coefficients for the gas on the surfaces number 1 and number 2, where

$$\alpha_i = (T_r - T_o) / (T_i - T_o) \quad (3)$$

Their values can range from 0 to 1 but most commonly are between 0.25 and 0.33 for hydrogen on clean surfaces [3]. κ is the mean free-molecular heat conductivity of the gas:

$$\kappa = \frac{1}{8} [(\gamma + 1) / (\gamma - 1)] * \frac{v}{T_{hot}} \quad (4)$$

where γ is the ratio of specific heats C_p/C_v and v is the molecular velocity:

$$v = (8 kT / \pi m)^{1/2} \quad (5)$$

At higher pressures, the movement of the gas molecules is impeded by collisions with one another, and the addition of more molecules per unit volume (increased pressure) no longer increases the conductivity, which becomes limited by the viscosity of the gas:

$$\text{Power}_{HP} = 1/4 (9\gamma - 5) * \eta * C_v * (T_{hot} - T_{cold}) * \frac{A}{Z} \quad (6)$$

where η is the gas viscosity

$$\eta = (0.499 (4 m k T)^{1/2}) / (\pi^{3/2} d_o^2) \quad (7)$$

C_v is the specific heat of the gas at constant volume, and γ is the ratio of specific heats $C_p/C_v = 1.4$ for simple diatomic gases such as hydrogen. The effective diameter of the molecule, d_o , determines the extent to which molecules collide and naturally influences the gas viscosity (above) and the mean-free distance between collisions (the mean-free path length) described below. where n_c is the gas density in number of molecules per unit volume and is related to the gas pressure through

$$\text{mfp} = (\pi d_o^2 n_c \sqrt{2})^{-1} \quad (8)$$

the ideal gas law:

$$n_c = N_A P_c / RT \quad (9)$$

where R is the universal gas constant. The pressure at which the transition occurs between pressure-dependent and pressure independent thermal conductivity is the pressure at which the mean-free path becomes comparable to the distance between the hot and cold surfaces. In this transition pressure regime, the total gas-phase conductivity is a combination of the two processes and may be calculated by adding the two kinds of gas-phase thermal resistances processes in parallel [4]:

$$\text{Power} = (1/\text{Power}_{LP} + 1/\text{Power}_{HP})^{-1} \quad (10)$$

The conductance through a thickness of gas is equal to the gas thermal conductivity divided by the thickness. Figure 1 shows the calculated thermal conductance of hydrogen gas across a 20 mm gap as a function of pressure. Notice that the pressure-dependent change in thermal conductance occurs over a limited range of pressures. The transition between pressure-dependent and pressure independent conductance begins at about 10^{-2} torr, where the mfp is about 10 mm.

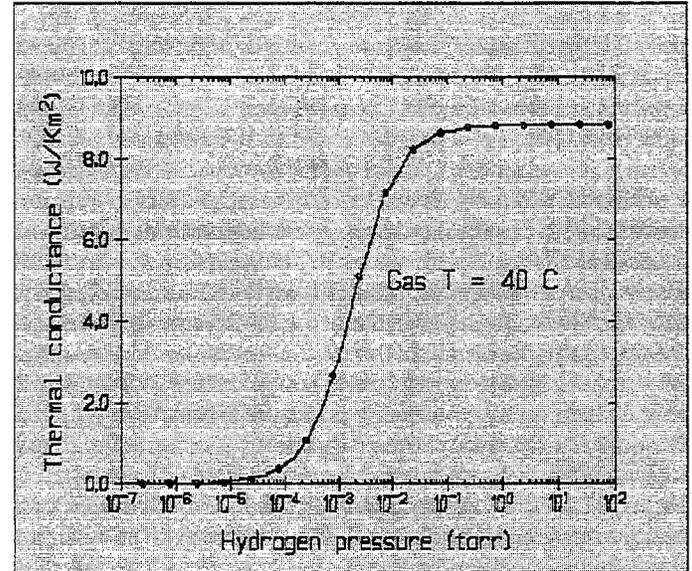


Figure 1. Calculated thermal conductance through the 20 mm thickness of a VCI panel as a function of internal hydrogen pressure.

QUANTITY OF HYDROGEN REQUIRED - Combining EQs (8) and (9) gives the critical hydrogen pressure, above which little change in thermal conductance occurs:

$$P_c \approx RT / (10 Z \pi d_o^2 N_A \sqrt{2}) \quad (11)$$

The total amount of hydrogen required to reach this pressure (per unit area of VCI) is

$$M_{H_2} = MW_{H_2} P_c Z/RT = \frac{MW_{H_2}}{10 N_A d_o^2 \sqrt{2} \pi} \quad (12)$$

METAL HYDRIDES — A large number of metal hydrides could be considered for use in VCI. Table 1 lists some of these and their characteristics. Zirconium alloys which are used in the manufacture of non-evaporable getters form reversible hydrides with suitable properties and have been used commercially for this purpose. A commercially available alloy used as a hydrogen source is Zr-V-Fe (.7-246-.054) [6]. This material is used as a fine powder contained within a porous stainless-steel cylinder containing a ceramic insulated, tungsten heater coil. The equilibrium pressure of hydrogen over this powder is given by a Sieverts' equation [7].

$$P_{H_2} = P_o x^2 \exp\left(\frac{-E_a}{BT_a}\right) \quad (13)$$

where P is the equilibrium hydrogen pressure, E_a is the activation energy, and T_a is the hydride operating temperature in Kelvin. The parameters in the Sieverts' equation for this commercial alloy are $P_o = 1.08 \cdot 10^9$ Pa and $E_a = 1.21$ eV/atom. Figure 2 shows the equilibrium pressure over similar alloys as a function of hydrogen concentration in the alloy at different temperatures. The hydrogen concentration is expressed as x, the atomic fraction of H in ZrH_x , since the behavior of these alloys follows the Zr-H phase diagram and the other alloy components do not play a prominent role in the hydrogen absorption.

Table 1.
Hydrogen Densities in Hydrogen Sources [5]

Hydrogen Source	H (10^{28} atoms/ m^3)	Disassociation Temperature (C)
TiH ₂	9.5	650
UH ₃	8.2	430
ZrNiH ₃	7.4	295
LaNiH _{6.7}	7.6	25

QUANTITY OF HYDRIDE — The geometry of the VCI enclosure dictates the critical pressure, P_c (EQ(11)), of hydrogen needed to saturate the gas-phase conductance and thereby inactivate the vacuum insulation. For a VCI with an interior spacing of 3 mm between the layers of radiation shields, this critical hydrogen pressure is about 10^{-2} torr (Fig. 1).

The critical hydrogen pressure can be reached by heating a hydride alloy to a temperature at which it is in thermodynamic equilibrium with hydrogen at that critical pressure (Fig. 2). The larger the initial hydrogen content of the alloy, the lower the temperature required to reach the critical pressure. For a value of $x = 0.01$ in the ZrH_x

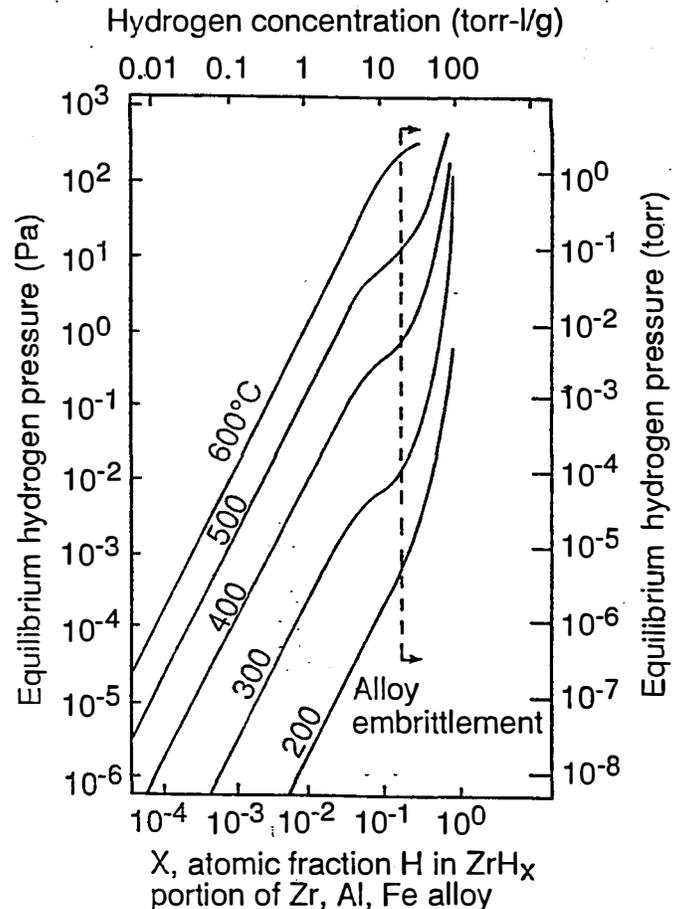


Figure 2. Equilibrium hydrogen pressure over various zirconium alloy hydrides(5).

hydride, the hydride dissociation temperature required is nearly 600°C, whereas for $x = 0.1$, the required dissociation temperature is only about 450°C.

As the hydrogen is evolved from the hydride, the value of x decreases. Consequently, so does the equilibrium hydrogen pressure which can be sustained by the hydride at the dissociation temperature. Therefore, the value of x must be selected somewhat higher than the required final value at the hydride operating temperature.

Enough hydride must be incorporated into the VCI to provide the required amount of hydrogen and still leave the fraction of hydrogen, x, in the hydride large enough to remain above the critical value associated with the critical hydrogen pressure at the hydride operating temperature. The amount of hydrogen needed is given by EQ(12) in terms of the critical hydrogen pressure and VCI separation between hot and cold surfaces. The amount of hydrogen may also be expressed in terms of the change in x in the ZrH_x portion of the hydride alloy:

$$M_{H_2} = \Delta x M_{H_V} F_{Zr} [MW_{H_2}/2MW_{Zr}] \quad (14)$$

where F_{Zr} is the weight fraction of zirconium in the (un-hydrided) alloy and $(MW_{H_2}/2MW_{Zr})$ is the ratio of a hydrogen atom weight to a zirconium atom weight expressed in terms of standard molecular weights. Combining this equation with EQ(12) yields an expression for Δx :

$$\Delta x = (2P_c z MW_{Zr}) / (RTM_{HY} F_{Zr}) \quad (15)$$

The final value of x must be large enough to assure a critical hydrogen pressure, P_c , at the hydride operating temperature, T_a . This value of x_{final} is obtained from EQ(13):

$$x_{final} = [P_c / (P_o \exp(-E_a/BT_a))]^{1/2} \quad (16)$$

Finally, the initial value of x is dependent upon the amount of hydride alloy to be used as well as the other details of the design, such as the VCI geometry and the hydride operating temperature:

$$\begin{aligned} x_{initial} &= x_{final} + \Delta x \\ &= [P_c / (P_o \exp(-E_a/BT_a))]^{1/2} \\ &\quad + (2P_c z MW_{Zr}) / (RTM_{HY} F_{Zr}) \end{aligned} \quad (17)$$

The amount of hydride alloy (in terms of grams per square meter of VCI projected surface area) can be expressed in terms of the critical hydrogen pressure and the selected $x_{initial}$ as:

$$m_{hy} = 2P_c z MW_{Zr} / [RTF_{Zr} (x_i - P_c / P_{H_2})^{1/2}] \quad (18)$$

where P_c and P_{H_2} are obtained from EQs (11) and (13), respectively. It is interesting that the amount of hydride is not dependent upon the thickness of the vacuum gap in the VCI. This independence results from the cancellation of two effects: the critical pressure is inversely proportional to the gap thickness, whereas the volume of hydrogen at the critical pressure is directly proportional to the gap thickness.

The VCI designer can choose to use more or less hydride alloy depending on how high an operating temperature he chooses for the hydride and how high a value of $x_{initial}$ is to be used. However, operating at higher temperatures may cause other design problems such as excessive heat loss from the hydride, and the use of too high a value of $x_{initial}$ may risk hydride degradation because the hydride particles may break up. Therefore, the design of the hydride component must be made judiciously. If the hydride is likely to be significantly contaminated by gases evolved from the internal components of the VCI during its operation, then a larger quantity of hydride will be required because hydrogen absorption capacity will be lost due to this contamination.

FABRICATION OF A BENCH-SCALE VCI TEST PANEL

A flat panel of VCI was fabricated from stainless-steel sheet and ceramic internal supports. The overall dimensions were 0.45 m x 0.45 m x 0.02 m. The

perimeter was permanently sealed by laser welding a band of 0.025 mm thick 304 stainless steel foil around the panel to seal the top and bottom sheets. As a part of this sealing process, the thin foil was also laser welded to itself. The internal zirconia ceramic supports were held in place in interference fit holes milled into 0.6-mm-thick aluminum sheets. The internal supports were separated from one another by a distance of 80 mm on a hexagonal pattern, and two layers of supports were used to decrease the thermal conductance through them. A 0.6-mm-thick stainless-steel sheet separated the layers of ceramic supports. The steel and aluminum sheets serve as effective thermal radiation barriers; but two additional 0.025-mm-thick copper foils were added to further decrease the radiative heat transfer.

Two small, standard, copper-gasketed vacuum flanges were installed on the top surface of the panel. One of the flanges was used to attach the metal hydride and the other was used as the evacuation port. The metal hydride was a commercial unit—a cylindrical, porous steel capsule containing about 100 mg of hydride and with an integral electrical heater [7]. The hydride was connected to an electrical feedthrough which included thermocouple leads as well as power-leads. A 0.075-mm-thick thermocouple was welded to the hydride to monitor its temperature during operation. Once the panel was evacuated and baked out at 200°C to remove adsorbed gasses, it was permanently sealed by pinching off the copper tube used for evacuation.

The thermal conductance through the thickness of the test VCI panel was measured using semiconductor heat flux gages that were calibrated against National Institute of Standards and Testing standard reference material 1449 [8]. The panel was placed on a uniformly heated surface and was insulated around the perimeter to prevent convective heat loss from the sides. The two 2-cm-diameter flux gages were placed near the center of the top surface along with a surface thermocouple. The thermal conductance was calculated as the measured heat flux (W/m^2) divided by the temperature difference between the top and bottom surfaces.

Figure 3 shows one cycle of operation of the VCI panel. While the temperature of the hydride changes very rapidly, the thermal conductance through the panel changes much more slowly because of the thermal inertia of the panel. The apparent thermal conductance of the panel during the transient portion of the recording is not likely to be accurate, however the nearly steady-state values of 9.0 and 1.0 are reproducible and accurate within the calibration uncertainty of the flux gages (about $\pm 5\%$) for a given range of hydride temperatures.

The cyclic reproducibility of the hydride hydrogen source was tested by repeatedly cycling the direct current through the hydride heater at fixed peak amperage (about 4.5 A at 2.5 V). This produces a hydride temperature range from about 75°C to about 500°C over a six-minute cycle more than 1200 times before the integral hydride heater failed. Periodic repeat measurements of the change in thermal conductance of the panel (on a much slower cycle) showed that its behavior was reproducible

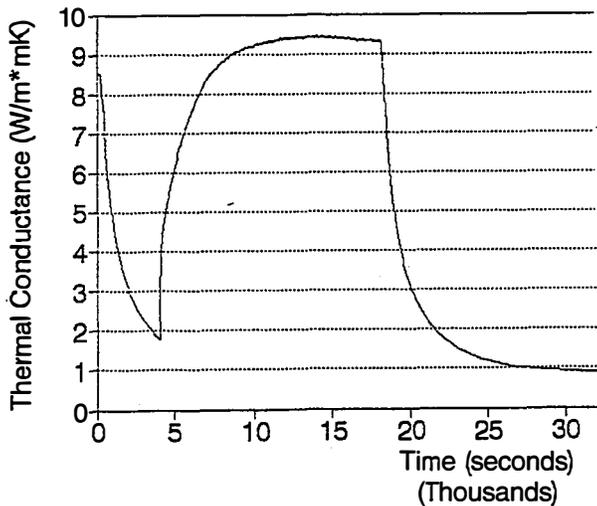


Figure 3. Measured thermal conductance through VCI panel during a cycle of operation.

within a range of about $\pm 10\%$, a variation which could be attributed to the variation in the range of hydride temperatures.

COMPARISON OF MEASURED AND CALCULATED VCI THERMAL CONDUCTANCES

Without a significant pressure of hydrogen, the lowest value of the panel conductance is made up of solid-phase conductance through the array of internal supports, the conductance through the perimeter foil seal, plus the radiative heat transfer conductance. The perimeter foil seal contributes to the minimum panel conductance no more than the following:

$$C_{\text{foil}} = 4 \cdot l \cdot t \cdot k_{\text{SS}} / (w \cdot l^2) = 0.18 \text{ W/m}^2\text{K}$$

where l is the panel edge length, t is the foil thickness, k_{SS} is the thermal conductivity of the foil (16.2 W/mK), and w is the thickness of the panel spanned by the foil perimeter seal. The radiative heat transfer is effectively limited by the multiple layers of radiation shields. The majority of the minimum panel conductance is due to the internal array of supports.

The measured range of VCI conductances is consistent with the expected range in hydrogen gas conductances (Fig. 1). Adding the minimum panel conductance of 1.0 W/m²K (fully evacuated with a cool hydride) to the calculated hydrogen gas conductance of about 8.4 W/m²K at pressures above 1 torr gives 9.4 W/m²K compared with the measured maximum panel conductance of 9.6 W/m²K. The agreement is well within the uncertainty of the measurements.

SOME POSSIBLE APPLICATIONS FOR VCI

A variable-conductance vacuum insulation may be applicable wherever the requirements for insulation vary over time and heat transfer should be controlled electron-

ically. Some possible applications in automobiles may include protecting lead-acid batteries against excessive underhood temperatures and thermally managing "hot" batteries.

VCI MANUFACTURING ISSUES

Several issues remain to be resolved before practical VCI enclosures can be fabricated. These include the choice construction materials, the durability of the hydrogen source, and the welding of the low-thermal-conductance edge seals required in most VCI designs.

The materials used in the VCI bench-scale demonstration were much too heavy to be practical. However, much lighter-weight vacuum insulation structures have been fabricated using laser-welded 0.2-mm-thick stainless steel envelopes and lighter-weight internal supports [9] demonstrating that such structures should be possible for VCI as well.

Although metal-hydride hydrogen sources are commercially available, they have not been designed to sustain a large number of repeated uses and further development of more durable sources is required. Nor is it clear that these hydrogen sources will not fail prematurely as a result of contamination from gases evolved from the surfaces and from within the volume of the materials of the VCI construction. More work is required to ensure the materials compatibility. It may even be necessary to use additional, separate reactive metal getter material in addition to the metal hydride hydrogen source to selectively trap the potentially contaminating gasses.

Perhaps the most difficult manufacturability problem is the design of the thin-foil, low-thermal-conductance edge seals which will be required for most VCI enclosure designs. We have shown that a very thin stainless-steel foil can be laser welded to form a vacuum-tight perimeter seal. This welding required extreme care in the fixturing and will require further development before it is practical for a mass-production process. A vacuum furnace brazing operation has also been used successfully with somewhat thicker (0.075 mm) stainless-steel foil and may be a more forgiving process. Further work is required.

SUMMARY AND CONCLUSIONS

A design for a variable-conductance vacuum insulation has been described and demonstrated to function as predicted in a bench-scale prototype panel of VCI. The use of hydrogen gas to provide controlled thermal conductance through such an insulation whenever and to the extent needed appears to be feasible from these calculations and experiments.

The upper limit to the thermal conductivity of the VCI is provided by the hydrogen gas and is limited to about 0.17 W/mK; the lower limit to the effective thermal conductivity of the vacuum insulation (with hydrogen reabsorbed into the metal hydride dispenser) is determined by the details of the insulation design, such as the thickness of the insulation, the perimeter-to-area

conductivity of the vacuum insulation (with hydrogen reabsorbed into the metal hydride dispenser) is determined by the details of the insulation design, such as the thickness of the insulation, the perimeter-to-area ratio, thickness of the perimeter seal, and the number and geometry of the internal supports and radiation shields. Designs of VCI with a dynamic range (the ratio of maximum conductance to minimum conductance) that is much higher than reported here, appears to be possible.

The VCI appears to have potential applicability wherever the management of heat flow into and out of an insulated enclosure is required. Some applications in automobiles were suggested. The application of VCI to the thermal management of automobile "hot" batteries is presently under development at NREL.

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NOMENCLATURE

A	area (m)
NA	Avagadro's number (number of molecules per mole) = 6.022×10^{23}
C_p	specific heat at constant pressure = 14309(J/mole K) for H ₂
C_v	specific heat at constant volume = 10201(J/mole K) for H ₂
d₀	molecular diameter = $.274 \times 10^{-9}$ (m) for hydrogen
F_{Zr}	weight fraction of hydride which is zirconium (dimensionless)
k	Boltzmann's constant = 1.380×10^{-23} J/K or 8.616×10^{-5} eV/atom K
k_{ss}	thermal conductivity of the edge foil seal (16.2 W/mK)
l	VCI panel edge length (m)
m	molecular mass = 0.335×10^{-23} g for H ₂
mfp	mean-free path length of gas molecule (m)
MW_{H2}	molecular weight of hydrogen (2.016 g/mole)
MW_{Zr}	molecular weight of zirconium (91.22 g/mole)
n	number density of gas molecules (m ⁻³)
n_c	critical concentration of hydrogen gas molecules (m ⁻³)
P₀	parameter in Sievert's equation (13) in units of Pa
P_c	critical pressure of hydrogen (Pa)
R	molar gas constant = 8.3145 (J/mole K)
t	edge foil thickness (m)

T	gas temperature (K)
T_a	hydride alloy operating temperature (K)
T_i	initial gas molecule temperature (K)
T_r	gas molecule temperature after rebounding from a surface (K)
T_s	surface temperature (K)
w	thickness of VCI panel (m)
z	thickness of gas-filled gap between warm and cold surfaces (m)
α_n	accommodation coefficient of a gas/surface _n
γ	the ratio of specific heats, C _p /C _v = 1.4 for H ₂
κ	free-molecular conductivity of a gas
η	gas viscosity (Pa-s)

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