Rechargeable Metal Hydrides for Spacecraft Application

J. L. Perry
George C. Marshall Space Flight Center
Marshall Space Flight Center, Alabama
<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>INTRODUCTION</td>
<td>1</td>
</tr>
<tr>
<td>METAL HYDRIDE THEORY</td>
<td>2</td>
</tr>
<tr>
<td>AB Compounds</td>
<td>2</td>
</tr>
<tr>
<td>AB₅ Compounds</td>
<td>2</td>
</tr>
<tr>
<td>Other Compounds</td>
<td>3</td>
</tr>
<tr>
<td>Hydride Phenomena</td>
<td>3</td>
</tr>
<tr>
<td>Temperature Dependence</td>
<td>4</td>
</tr>
<tr>
<td>ENGINEERING PROPERTIES OF METAL HYDRIDES</td>
<td>5</td>
</tr>
<tr>
<td>Plateau Pressure</td>
<td>5</td>
</tr>
<tr>
<td>Hysteresis</td>
<td>5</td>
</tr>
<tr>
<td>Enthalpy of Reaction</td>
<td>5</td>
</tr>
<tr>
<td>Hydrogen Capacity</td>
<td>6</td>
</tr>
<tr>
<td>Chemical Stability</td>
<td>6</td>
</tr>
<tr>
<td>Reaction Kinetics</td>
<td>7</td>
</tr>
<tr>
<td>Surface Poisoning</td>
<td>7</td>
</tr>
<tr>
<td>HYDROGEN STORAGE SAFETY</td>
<td>7</td>
</tr>
<tr>
<td>Conventional Storage</td>
<td>8</td>
</tr>
<tr>
<td>Hydride Storage</td>
<td>8</td>
</tr>
<tr>
<td>SPACE STATION APPLICATIONS</td>
<td>8</td>
</tr>
<tr>
<td>ECLSS Application</td>
<td>8</td>
</tr>
<tr>
<td>U.S. Laboratory Application</td>
<td>8</td>
</tr>
<tr>
<td>A REFERENCE HYDRIDE CONTAINER</td>
<td>10</td>
</tr>
<tr>
<td>Container Features and Geometry</td>
<td>10</td>
</tr>
<tr>
<td>Materials of Construction</td>
<td>11</td>
</tr>
<tr>
<td>Energy Requirements</td>
<td>11</td>
</tr>
<tr>
<td>COMPARISON TO BOTTLED STORAGE</td>
<td>11</td>
</tr>
<tr>
<td>Volume Comparison</td>
<td>12</td>
</tr>
<tr>
<td>Mass Comparison</td>
<td>13</td>
</tr>
<tr>
<td>Pressure Comparison</td>
<td>13</td>
</tr>
<tr>
<td>Storage Technique Safety</td>
<td></td>
</tr>
</tbody>
</table>

PRECEDING PAGE BLANK NOT FILMED
CONCLUSIONS.................................................................................................................. 14
APPENDIX A: Tabulated Absorption and Desorption Data.............................................. 15
APPENDIX B: Engineering Properties............................................................................. 17
APPENDIX C: Hydride Container Sizing Calculations and Bottled Storage Calculations .... 19
APPENDIX D: Energy Requirement Calculations............................................................ 25
REFERENCES...................................................................................................................... 31
# LIST OF ILLUSTRATIONS

<table>
<thead>
<tr>
<th>Figure</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Ideal equilibrium isotherm</td>
<td>3</td>
</tr>
<tr>
<td>2</td>
<td>Van't Hoff relationship for LaNi₅</td>
<td>4</td>
</tr>
<tr>
<td>3</td>
<td>Charging a metal hydride</td>
<td>9</td>
</tr>
<tr>
<td>4</td>
<td>Discharging a metal hydride</td>
<td>9</td>
</tr>
<tr>
<td>5</td>
<td>A rapid-cycling hydride container</td>
<td>10</td>
</tr>
<tr>
<td>6</td>
<td>Storage volume comparison</td>
<td>12</td>
</tr>
<tr>
<td>7</td>
<td>Storage mass comparison</td>
<td>13</td>
</tr>
<tr>
<td>8</td>
<td>Storage pressures at 298 Kelvin</td>
<td>14</td>
</tr>
</tbody>
</table>
RECHARGEABLE METAL HYDRIDES FOR SPACECRAFT APPLICATION

INTRODUCTION

A metal hydride is a chemical compound formed from the reaction of hydrogen with a metal or metal alloy. Many hydrides absorb and release hydrogen like a solid sponge at temperatures and pressures which are useful for storing hydrogen on board the Space Station U.S. Laboratory module. Metal hydrides may also be used to pump, compress, or expand hydrogen gas. These uses may provide safer ways for handling hydrogen onboard spacecraft such as the Space Station.

Metal hydrides undergo a reversible reaction of a solid metal or metal alloy with gaseous hydrogen to form a solid metal hydride according to the reaction,

\[ M + \frac{1}{2}xH_2 \Rightarrow MH_x + \text{heat}. \]

Several metals react directly and reversibly with hydrogen at room temperature and at or near atmospheric pressure. These metals include elements, solid-solution alloys, and intermetallic compounds. Hydrides formed from these materials are known as rechargeable metal hydrides. In these hydrides, the metal behaves like a solid sponge that can be charged and discharged at will.

Metal hydrides possess advantages for storing and transporting hydrogen in spacecraft over other methods such as compressed gas and cryogenic liquid storage. First, metal hydrides have a high volumetric packing density. Table 1 compares the packing density for some typical metal hydrides, compressed gas, and liquid. The volumetric density for hydrides is many times that of the conventional storage methods. Second, storage using metal hydrides requires low pressures. This has significant safety implications. Third, hydride storage is very energy efficient [1].

<table>
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<tr>
<td>LaNi(_5)H(_8).7)</td>
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Although metal hydrides have some significant advantages over high pressure and cryogenic hydrogen storage, they do have some disadvantages. The metals and metal alloys used for rechargeable metal hydrides are extremely rare. This results in high costs for the alloys and a high cost for each kilogram of hydrogen stored. Also, the alloy weight is higher than conventional storage methods. However, this disadvantage has been shown to be insignificant for spacecraft applications when compared to high pressure storage.

The flexibility and advantages of metal hydrides make them very attractive for use on board the Space Station where significant quantities of hydrogen must be handled. These applications include the process fluid supply subsystem of the Process Material Management System on board the U.S. Laboratory Module, the life support system which may produce hydrogen as a by-product, and the propulsion system. However, significant development efforts will have to be made to apply metal hydrides to these areas of the Space Station and other spacecraft and hydriding phenomena must be fully understood.

**METAL HYDRIDE THEORY**

Rechargeable metal hydrides exist in several families of intermetallic compounds which consist of at least one element A that has a high affinity for hydrogen and a second element B that has a low affinity for hydrogen. Among the families of compounds which are practical for hydrogen storage are the AB and AB₅ compounds. These compounds exhibit good hydrogen storage properties at room temperature and atmospheric pressure.

**AB Compounds**

One of the most-used room temperature AB compounds is FeTi. It is relatively inexpensive but has some definite disadvantages. The first is high hysteresis. The alloy has a large hysteresis ratio resulting in a hydriding process which is not completely reversible. Second, FeTi has a low resistance to poisoning by oxygen. Finally, the alloy has a heating requirement for activation. FeTi can be combined with other elements such as Mn, Cr, Co, Ni, and V to produce an alloy which is more useful [1].

**AB₅ Compounds**

The most popular AB₅ compound is LaNi₅. This alloy has very attractive properties for use in storing hydrogen. These properties include convenient plateau pressures, low hysteresis, excellent kinetics, easy activation, and good resistance to poisoning. Because of its overall good performance properties as a rechargeable metal hydride, this alloy was selected as the candidate for use on the Space Station during the Phase B study [1].
Other Compounds

Several other compositions for metal hydrides exist. An example is the A₂B compounds of which Mg₂Ni is a member. This alloy is extremely lightweight and has a small hysteresis. Unfortunately, it requires a temperature of 573°K (300°C) for hydrogen desorption. Also the energy of reaction is high. These problems make it impractical for Space Station application. Other compounds are based on Mg, Zr, and Ti. These compounds show promise but their kinetics are very slow or they are unstable at room temperature and are not cost competitive with other compounds.

Hydride Phenomena

At normal temperatures and pressures, hydrogen cannot be condensed regardless of the amount of pressure applied. However, in the presence of AB or AB₅ compounds, a vapor pressure relationship is established between hydrogen gas and a solid form of hydrogen which is bound in the crystal structure of the metal alloy. The hydride equilibrium conditions are expressed in terms of pressure, temperature, and composition [2]. Figure 1 shows a sample equilibrium isotherm.

At point 1 in Figure 1, the metal phase and hydrogen are separate. A constant temperature is maintained between points 1 and 2 while the hydrogen pressure increases. As the hydrogen pressure increases, a small amount of hydrogen goes into solution in the metal phase. At point 2, the hydriding reaction begins and large quantities of hydrogen are absorbed at constant pressure. This pressure is known as the plateau pressure, Pp. The plateau is characterized by a two phase mixture of metal and metal hydride. At point 3, the alloy has been completely converted to the hydride phase and any increase in pressure results in only a small addition of hydrogen to the

![Figure 1. Ideal equilibrium isotherm.](image)
Hydride phase. This is the ideal behavior of metal hydrides. Ideal behavior is seen occasionally but in most practical hydriding reactions the plateau may be sloped slightly and some pressure hysteresis between absorption and desorption is observed. Hysteresis for LaNi$_5$ is small, making the alloy suitable for many applications.

**Temperature Dependence**

The hydriding reaction is very dependent on temperature. As the temperature increases so does the plateau pressure. The heat of reaction is also a factor since the absorption reaction is exothermic and the desorption reaction is endothermic. The plateau pressure is related to the absolute temperature by the van't Hoff equation

$$\ln\left(\frac{P}{P_0}\right) = \frac{\Delta H}{RT} + C$$

In this equation, $T$ is the absolute temperature, $\Delta H$ is the enthalpy change per mole of $H_2$, $R$ is the universal gas constant, and $C$ is a constant related to the entropy change of the reaction. A family of isotherms can be plotted as $\ln\left(\frac{P}{P_0}\right)$ versus $1/T$ to obtain the van't Hoff plot for a particular material. The slope of the plot gives the value of $\Delta H$. The van't Hoff plot for LaNi$_5$ is given in Figure 2. Data for this plot are tabulated in Appendix A. The enthalpy of reaction for LaNi$_5$ is $-30861.57$ J/mole $H_2$. This is for the absorption reaction. The same amount of energy must be supplied to the hydride during the desorption reaction.

Figure 2. Van’t Hoff relationship for LaNi$_5$. 
Different hydrogen pressures may be provided simply by heating or cooling the hydride to a
temperature corresponding to the pressure desired. By cycling the temperature of a metal hydride,
hydrogen may be absorbed and desorbed under various conditions. This flexibility makes a metal
hydride an excellent medium for handling various hydrogen operations ranging from storage to
compression.

ENGINEERING PROPERTIES OF METAL HYDRIDES

Several engineering properties are important to the application of metal hydrides to storage
and other problems. These include the plateau pressure, hysteresis, enthalpy of reaction, hydrogen
capacity, chemical stability, reaction kinetics, and tolerance to poisoning. These properties aid in
selecting a particular metal hydride for a specific use. Engineering properties for LaNi₅ are
summarized in Appendix B.

Plateau Pressure

The plateau pressure and associated operating pressure of the metal hydride depend on the
application. Plateau pressure is directly related to the metal composition. By altering the composi-
tion, a metal hydride can be tailored specifically to meet the pressure and temperature requirements
of a particular application [1].

Hysteresis

Practical application of metal hydrides to hydrogen storage and other uses requires a small
difference between the absorption and desorption pressures. Hysteresis varies according to the alloy
used in a manner which is not clearly understood. However, empirical knowledge can show which
systems exhibit small hysteresis and thus can be used practically [1]. The degree of hysteresis is
expressed as the ratio between the absorption pressure and the desorption pressure, \( P_a/P_d \) [3].

Enthalpy of Reaction

The enthalpy of reaction is very important from a container design viewpoint. Energy is
generated during the exothermic absorption reaction while energy must be provided to sustain the
endothermic desorption reaction. As a result of the energy requirements to support the hydride
cycling, the hydride container must be an effective energy exchanger [1]. Most applications require
rapid cycling to be effective and the more efficiently the container can exchange energy during
each cycle, the more use can be obtained from the hydride.
Hydrogen Capacity

The hydrogen capacity is a function of the crystal structure and base composition of the hydride-forming alloy. Metallurgical factors which result from alloy preparation may also be involved. The hydrogen capacity may also drive the size and cost of a particular hydride system since it varies according to the desired pressure range required for the application [3]. Large volume changes are also associated with the absorption and desorption reactions. For example, LaNi₅ may expand by as much as 25 percent during absorption and contract by a similar amount during desorption. This expansion and contraction must be considered in container design [1].

Chemical Stability

The chemical stability of the compound must be considered for high temperature applications. Under certain conditions some undesirable reactions can occur resulting in reduced performance of the compound. An example is disproportionation. CaNi₅ undergoes the reversible reaction,

\[ \text{CaNi}_5 + 3\text{H}_2 \rightleftharpoons \text{CaNi}_5\text{H}_6 \]

near room temperature. However, the disproportionation reaction is thermodynamically preferred.

\[ \text{CaNi}_5 + \text{H}_2 \rightleftharpoons \text{CaH}_2 + 5\text{Ni} \]

At high temperatures, diffusion of the metal atoms can be significant and result in the disproportionation reaction being favored over the reversible reaction. The result is a loss of reversible capacity. Most compounds used for reversible metal hydrides are unstable with respect to disproportionation and their tendency to disproportionate varies from compound to compound. Most compounds which are useful at room temperature exhibit more resistance to disproportionation than those used in high temperature applications [1].

A second form of chemical instability lies in degradation resulting from temperature induced cycling. Most applications for reversible metal hydrides involve repeated absorption-desorption cycles. Studies of this phenomenon have concluded that LaNi₅ at 2.17 MPa (315 psia) cycled between room temperature and 573°K (300°C) retains only 26 percent of its original hydrogen-absorbing capacity after 1550 cycles at a rate of three cycles per hour. An extrapolation of the data of these studies indicates that the alloy would lose 50 percent of its absorbing capacity at room temperature after $4 \times 10^5$ cycles at a rate of one cycle every ten minutes for eight years. The degradation rate also depends on the pressure and rates of absorption and desorption [4]. A reasonable limit for replacing a metal hydride to maintain its efficiency would be after 50 percent of its original absorbing capacity had been lost. This maintenance would occur after a relatively long service life and shows that metal hydrides would require little or no maintenance during its useful life.
Reaction Kinetics

The reaction kinetics of metal hydrides are very important to the selection of operating temperatures, pressures, and cycle times for hydrogen storage and other applications. Kinetics are a function of the alloy composition and its crystal structure. Investigations of the reaction kinetics of LaNi₅ at room temperature show that the absorption and desorption reaction rates are extremely fast and are limited by energy transfer in practical applications. A properly designed container with good energy transfer characteristics would provide an excellent means for rapidly storing and providing hydrogen [5,8].

Surface Poisoning

Impurities in the hydrogen used in the absorption or desorption reaction results in a loss of hydrogen absorption or desorption kinetics. Gaseous impurities such as O₂, H₂O, and CO may form a surface film or structure which poisons the surface catalytic properties of the alloy. The poisoning may result from surface film formation, surface physisorption, or surface chemisorption. Little information is available about the actual mechanisms of hydride surface poisoning. Cyclic poisoning tests of several alloys show that these impurities do reduce the ability of hydride alloys to absorb and desorb hydrogen. LaNi₅ shows the most resistance to surface poisoning when compared to alloys of FeTi and Mn-substituted FeTi. Hydrogen containing 300 ppm of oxygen and water cause LaNi₅ to lose some absorption-desorption ability after approximately four 0.5-hr cycles. However, the alloy recovers and returns to an absorption-desorption ability which is very close to its original ability. Hydrogen containing 300 ppm of CO causes the alloy to lose its hydriding ability after approximately 16 of these cycles. Fortunately, it can be reactivated by using suitable procedures such as continued cycling or heating [6].

HYDROGEN STORAGE SAFETY

Conventional Storage

The main safety threats encountered when handling hydrogen are fire and explosion. These threats are present any time hydrogen is transported since the container can be damaged or ruptured releasing its contents into the laboratory module. For gaseous and liquid hydrogen storage, such a rupture would release all the contents of the container. Several minutes would be required for the spill to dissipate. On Earth this type of spill would be diluted by air and present a fire risk for several minutes. The time depends, of course, on the size of the spill. On board the Space Station and other spacecraft, the diluting atmosphere has a much smaller volume, causing an increased risk for explosion or fire. This risk causes the design of hydrogen storage containers, which cannot leak or rupture, to be extremely important. The high pressures and cryogenic temperatures involved in conventional storage techniques contribute to the risks of handling hydrogen. High pressures cause any leak or rupture to release hydrogen rapidly and cause the rupture itself to be dangerous since the storage container could be launched like a rocket. The cryogenic temperatures cause a risk of injury to personnel who may experience direct contact to their skin causing burns. Liquid hydrogen could also adhere to surfaces which are not easily accessed, causing a spill to be even more difficult to contain.
Hydride Storage

Storage using rechargeable metal hydrides requires low temperatures and pressures. This results in very little free hydrogen present at any time. If the container is damaged or ruptured the hydride would only liberate free hydrogen at a rate dependent on the hydride bed temperature, the ambient temperature, and the rate of energy transfer to and from the hydride bed [7]. Hydrogen may be released at a significant rate if the hydride reactor is operating at a high temperature, however, the rate can be reduced quickly by cooling the hydride bed as quickly as possible. Overall, the rechargeable metal hydride is safer since an explosive rupture is highly unlikely and any flame would be for only a brief time. An example is a test which involved firing incendiary shells into a hydride tank. This test resulted in only a brief flame [7].

SPACE STATION APPLICATIONS

Application of rechargeable metal hydrides to hydrogen handling problems on board the Space Station may be possible in several areas. Among these are as an accumulator between the electrolyzer and the CO₂ reduction process, an accumulator for excess hydrogen generated by the ECLSS, and hydrogen supply for U.S. Laboratory experiments and operations.

ECLSS Application

A hydrogen accumulator can be useful to reduce transient flow problems between the electrolyzer and the CO₂ reduction process. This application can also prove useful for collecting any residual hydrogen generated by the ECLSS. A container having a 10.2 kg mass and 1.25 percent useable hydrogen by weight may be used. The reference hydride is an AB₅ type. Assuming the electrolyzer produces 1.13 kg/day of hydrogen, the hydride accumulator would require a 200 W load to be removed. This load may be removed by water flowing at 0.6 liters/min with a 5°K temperature differential between the inlet and exit temperatures. The 293°K (20°C) equipment loop may be used for the shell side fluid [2].

U.S. Laboratory Application

Rechargeable metal hydrides can be used on board the U.S. Laboratory Module for storing hydrogen for experiment use or for collecting waste hydrogen from experiments. A suggested system would collect hydrogen from the ECLSS which is not used by the CO₂ reduction system. This application is summarized in Figures 3 and 4. Hydrogen is generated by the electrolyzer at 298°K (25°C) and 1.79 kPa (26 psia). When the Bosch process is used for CO₂ reduction, approximately 0.206 kg H₂/day is left over. A survey of experiment requirements shows that 0.006 kg H₂/day is required to maintain a 14 experiment set. This leaves 0.200 kg H₂/day which must be stored or used as a fuel source. Once the hydride has been charged, it is replaced by another hydride container. The charged container is then taken to the laboratory module where it is discharged to support experiment operations.
Figure 3. Charging a metal hydride.

Figure 4. Discharging a metal hydride.
A REFERENCE HYDRIDE CONTAINER

Container Features and Geometry

The design of a container for rechargeable metal hydrides must perform several functions. It has to be a pressure vessel, a heat exchanger, a gas distribution system, and a filter, in addition to containing the hydride powder. The container cannot provide these functions at optimal levels since some conflict exists between them; therefore, the overall design is a compromise among the tasks it performs.

The container provides a closed volume for the powdered alloy which absorbs and desorbs hydrogen. The powdered alloy is contained in tubes like those found in a shell and tube heat exchanger. Each tube also contains a filter and a copper brush which enhances heat transfer. Seven tubes are included in each container. The shell is large enough to reduce pumping power requirements of fluids flowing axially along the tubes. The tubes have an outside diameter of 1.270 cm and an inside diameter of 1.2192 cm. Similarly, the shell has an outside and inside diameter of 4.445 cm and 4.343 cm, respectively. The tubes are capable of holding 5 g H$_2$/meter of tube. The maximum tube length is 40 cm and the overall container length is 44.44 cm. This design has a 0.000690 m$^3$ volume and a total dry mass of 1.45 kg. Sizing calculations are summarized in Appendix C. Fourteen grams of hydrogen can be stored in a single container. This is a 2.5 day supply of hydrogen for a 14 experiment payload complement. Figure 5 shows a metal hydride container [2].

Figure 5. A rapid-cycling hydride container.
Materials of Construction

For Space Station use, the container used must also be lightweight. This requirement can be met by selecting a material with low density and high yield stress. Many of the low density metals which may be used structurally, such as magnesium and titanium, are attacked by hydrogen and may become brittle. However, aluminum shows excellent resistance to hydrogen in addition to its good structural properties. It should be noted that high temperature applications would require using stainless steels and other more corrosion resistant alloys.

Energy Requirements

Energy requirements for a typical hydride container are based on the geometry of the container and the hydrogen absorption and desorption rates required. In order to provide the 5.54 g H₂/day required by the 14 experiment set, energy must be removed during the absorption reaction and supplied during the desorption reaction. The reference container has 1117.2 cm² of total surface area available for energy transfer. An energy balance on each tube yields an energy flux of 0.00667 W/cm² or 52.2 W during the entire hydride charging process. This equals a total flux of 0.0467 W/cm² for the entire exchanger. During the desorption reaction, energy must be supplied at the same rate it is removed during the absorption reaction. Coolant, assumed to be water at 277°K (4°C) inlet temperature and 282°K (9°C) outlet temperature, must be supplied at 0.0025 kg/s to provide the absorption rate necessary to support the experiments. The energy input for desorption requires a heater which will provide 102.9 W. This heater will be able to raise the hydride temperature from room temperature, 298°K, to its upper operating limit, 353°K, in five minutes. This will provide a plateau pressure of 1.17 MPa (169.3 psia). The energy required to sustain the desorption reaction is small compared to the heating requirement. It is 0.981 W. The same amount of energy, 102.9 W, must be removed by the coolant when the hydride is being cooled to room temperature from its maximum operating temperature. This requires 0.00294 kg/s of water at 277°K inlet temperature and a 282°K outlet temperature. For an entire cycle which includes absorption, hydride heating to its maximum operating temperature, sustained desorption, and cooling to the original temperature, the total net energy requirement is 51.22 W removed. Appendix D summarizes the energy requirement calculations.

COMPARISON TO BOTTLED STORAGE

Rechargeable metal hydrides offer an alternative to bottled storage, but to actually compare the two storage techniques requires comparison of weight and volume for both the hydride container and bottled storage. This comparison has been made for several hydrogen storage amounts to show the mass and volume characteristics for varying requirements.
Volume Comparison

The total volume for a reference hydride container 4.445 cm in diameter and 44.44 cm long is 0.000690 m$^3$. Each container can hold 14.0 grams H$_2$. This is equivalent to a 2.5 day supply of hydrogen for the U.S. Laboratory. Bottled storage for laboratory use would be approximately 0.00230 m$^3$ and the same amount of hydrogen would be stored in this container at 7.85 MPa (1138 psia). An external storage concept proposed during the Phase B study provides 0.0116 m$^3$ of storage from two storage tanks. As the amount of hydrogen storage is increased, the volume for the two internal storage techniques also increases. Two days of storage requires a hydride container which occupies 0.000560 m$^3$ while a single 0.00230 m$^3$ bottle will store the hydrogen at 5.92 MPa (858.2 psia). Up to 15 days of storage has been investigated. This storage amount requires six hydride containers with a total volume of 0.00414 m$^3$ while bottled storage requires six bottles with a total volume of 0.0138 m$^3$. Figure 6 illustrates this comparison.

Mass Comparison

The total mass of the storage techniques is also important to spacecraft application. The reference hydride container has a mass of 1.45 kg while the bottle’s mass is 7.0 kg. The Phase B external storage option has a total mass of 68.8 kg. For up to 15 days of storage, the hydride container mass climbs to 8.70 kg while the bottled storage mass reaches 42.0 kg. This comparison is shown by Figure 7.

![Figure 6. Storage volume comparison.](image-url)
A third area of comparison for the storage techniques is pressure. At room temperature, the hydride container stores hydrogen at 2.00 kPa (29 psia) while bottled storage requires 7.85 MPa (1138 psia). The external storage pressure depends on the amount of hydrogen stored since the volume does not vary in this comparison. For 2.5 days of storage, the pressure is 1.49 kPa (21.60 psia) while 15 days of storage results in a pressure of 8.97 kPa (130.06 psia). Figure 8 shows the pressure differences between the three storage techniques as the mass of hydrogen stored increases.

**Storage Technique Safety**

The overall safety of a storage technique is a combination of several factors. Among these are the storage pressure and volume. Bottled storage requires higher pressures for storing an equal amount of gas as a rechargeable metal hydride at the same temperature. In the event of a storage container failure such as a rupture or valve failure, a large amount of hydrogen may be released into the laboratory if the container is a pressurized bottle. The bottle contains free hydrogen and the higher the pressure, the more rapid the leak. This implies that any bottled storage should be low pressure, small volume. Unfortunately, this approach results in a large number of bottles to meet the hydrogen supply requirements. Metal hydride storage is a low pressure technique and very little hydrogen is present in its free form. Therefore, a container failure would not release all the hydrogen into the laboratory. The major safety risk with metal hydrides is the high temperatures which may be necessary to provide higher pressure hydrogen. These temperatures may not be necessary if a compressor is used but this introduces a mechanical system which may leak or be more prone to failure. These factors indicate rechargeable metal hydrides may be safer with respect to leaks and container failure.
Figure 8. Storage pressures at 298°K.

CONCLUSIONS

Analysis of the techniques available for storing hydrogen on board the Space Station indicates that rechargeable metal hydrides may be competitive with conventional storage techniques such as bottled and liquid storage. However, a substantial amount of development work and further analysis of specific applications is required to determine the actual usefulness of rechargeable metal hydrides in spacecraft. Preliminary analysis shows the volume required by the hydride container is less than storage bottles. The hydride container mass is also less. In addition, lower pressures are needed by metal hydrides to store hydrogen at room temperature. The lower pressure and small amount of free hydrogen present in a hydride container may increase the safety of the storage technique over bottled storage. Energy exchange is required for cycling the hydride between absorption and desorption but the net energy requirement is relatively small. The apparent advantages rechargeable metal hydrides have over conventional storage techniques in volume, mass, and safety make them very attractive for spacecraft use.
APPENDIX A

TABULATED ABSORPTION AND DESORPTION DATA
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<td>0.00</td>
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</tr>
</tbody>
</table>

Absorption and Desorption Data for Lanth.
APPENDIX B

ENGINEERING PROPERTIES
Metal Hydride Calculations and Notes

Reaction: \( \text{LaNi}_5 + 3\text{H}_2 \rightleftharpoons \text{LaNi}_5\text{H}_6 \)

Plateau Pressure-Temperature Relationship:

\[ M + \frac{3}{2}\text{H}_2 \rightleftharpoons \text{MH}_3 \]

van't Hoff relation: \( \ln(P/P_o) = \frac{\Delta H}{RT} + C = A/T + C \)

\( A = -3712 \text{ K} \) and \( C = 12.96 \)

Absorption-desorption Enthalpy:

\[ \Delta H/R = -3712 \text{ K} \Rightarrow (-3712 \text{ K})(8.314 \text{ J/mole-K}) = \Delta H \]

\[ \Delta H = -30861.57 \text{ J/mole} \]

Solution for Plateau Pressure:

Desorption: \( \ln(P_d/P_o) = -3712/T + 12.96 \)

\[ P_d = P_o \exp\left[(-3712/T) + 12.96\right] \]

where \( P_o = 1.01 \text{ kPa} \) (14.696 psia)

Absorption: \( P_a/P_d = 1.21 \Rightarrow P_a = 1.21P_d \)

or \( P_a = 1.21P_o \exp\left[(-3712/T) + 12.96\right] \)

Solution for Plateau Temperature for Absorption and Desorption:

\[ \ln(P/P_o) = -3712/T + 12.96 \]

\[ T\ln(P/P_o) = -3712 + 12.96T \]

\[ T = -3712/[\ln(P/P_o) - 12.96] \]

Engineering Properties of \( \text{LaNi}_5 \):

\[ \Delta H_a = -30861.57 \text{ J/mole H}_2 \]

\( \text{LaNi}_5: \rho = 8.3 \text{ g/cm}^3 \) (ref 1) \( C_p = 0.364 \text{ J/g-K} \) (ref 2)

\( \text{LaNi}_5\text{H}_6: \rho = 6.59 \text{ g/cm}^3 \) (ref 1) \( C_p = 0.435 \text{ J/g-K} \) (ref 2)

Hydrogen mass percent = 1.38; 80%-90% exchanged in a cycle

Hysteresis pressure ratio = 1.21 = \( P_a/P_d \) (ref 3)

Powder thermal conductivity = 1.32 W/m-K (ref 2)

Hydrogen requirements:

Excess hydrogen: 18.57 kg/90 days or 0.206 kg/day

Hydrogen required by experiments: 0.44 kg/90 days + 10%

\[ = 0.499 \text{ kg/90 days} \]

\[ \Rightarrow 0.00554 \text{ kg/day} \]
APPENDIX C

HYDRIDE CONTAINER SIZING CALCULATIONS AND BOTTLED STORAGE CALCULATIONS
Laboratory Hydride Exchanger Sizing (for maximum exchanger size):

Maximum tube length = 40 cm
Reference exchanger holds 5 g H₂/meter of tube
Seven tubes are in a single exchanger
Total tube length = (40 cm)(7) = 280 cm = 2.80 m

Amount of hydrogen stored in a single exchanger:

\[(5 \text{ g H}_2/\text{m})(2.80 \text{ m}) = 14.0 \text{ g H}_2\]

Provides \(~ (14.0 \text{ g})/(5.54 \text{ g/day}) = 2.5 \text{ days of hydrogen storage}\)

Exchanger dimensions:

Shell I.D. = 4.343 cm
Shell O.D. = 4.445 cm
Tube I.D. = 1.2192 cm
Tube O.D. = 1.270 cm

Exchanger length:

\[L = \text{tube length} + 4.44 \text{ cm} = 40 \text{ cm} + 4.44 \text{ cm} = 44.44 \text{ cm}\]

Exchanger volume:

\[V = \frac{1}{4} \pi D^2 L = \frac{1}{4} \pi (4.445 \text{ cm})^2 (44.44 \text{ cm})\]
\[V = 689.6 \text{ cm}^3\]

Hydride mass:

\[M_{\text{H}} = \left(\frac{\pi}{4}\right) \rho_{\text{H}} D^2 L\]
\[\rho_{\text{H}} = 3.95 \text{ g/cm}^3 \text{ (Ref. 2, p. B-2)}\]
\[D = 1.2192 \text{ cm}\]
\[L = 280 \text{ cm}\]
\[M_{\text{H}} = 1291 \text{ g} = 1.291 \text{ kg}\]

Tube mass:

\[M_t = \left(\frac{\pi}{4}\right) \rho_t (D_1^2 - D_2^2) L\]
\[\rho_t = 2.70 \text{ g/cm}^3\]
\[M_t = 75.1 \text{ g} = 0.0751 \text{ kg}\]
Shell mass:

\[ M_s = \frac{\pi}{4} \rho_s (D_s^2 - D_e^2) L \]
\[ \rho_s = 2.70 \text{ g/cm}^3 \]
\[ L = 44.44 \text{ cm} \]
\[ M_s = 84.5 \text{ g} = 0.0845 \text{ kg} \]

Total dry mass:

\[ M_{\text{TOT}} = M_H + M_t + M_s \]
\[ M_{\text{TOT}} = 1.291 \text{ kg} + 0.0751 \text{ kg} + 0.0845 \text{ kg} = 1.451 \text{ kg} \]

Exchanger characteristics summary:

Length: 44.44 cm
Volume: 689.6 cm³
Hydride mass: 1.291 kg
Tube mass: 0.0751 kg
Shell mass: 0.0845 kg
Hydrogen mass stored: 0.0140 kg
Days of storage supply: 2.5

Shell I.D.: 4.343 cm
Shell O.D.: 4.445 cm
Tube I.D.: 1.2192 cm
Tube O.D.: 1.270 cm

Exchanger material: 6016 Al, TIG welded joints, post assembly heat treat to T-6 in vacuum

Note: Austenitic stainless steel or inconel is required for high temperature applications.
Bottled Hydrogen Comparison:

Bottle volume for lab use: 2300 cm³

14.0 g hydrogen stored = 6.94 moles hydrogen

Redlich-Kwong Equation of State (Properties of Gases and Liquids, Reid, et al.)

\[ P = \frac{RT(V-b)}{a/[T^{0.5}V(V+b)]} \]

\( T = 298 \text{ K} \quad T_c = 33.2 \text{ K} \)

\( R = 82.06 \text{ cm}^3 \text{ atm/mol K} \quad P_c = 12.8 \text{ atm} \)

\[ a = \left[9(2^{0.333} - 1)\right]^{-1}RT_c^{0.5}P_c = 1428283.133 \text{ cm}^3 \text{ atm/mol}^2 \text{ K}^{1.5} \]

\[ b = \left[(2^{0.333} - 1)/3\right]RT_c/P_c = 18.44 \text{ cm}^3/\text{mol} \]

Solve for the bottled pressure:

\[ P = \frac{(82.06)(298)/\{(2300)/(6.94)\} - 18.44\} - 1428283.133/(\{(298)^{0.5}\{(2300)/(6.94)\} + 18.44\}) \]

\( P = 77.4 \text{ atm} = 7.84 \text{ MPa} \)

Boeing End Item Data Book: Hydrogen Storage

2 storage tanks: 34.4 kg, 0.48 m diameter each
Total mass: 68.8 kg

\[ V = \frac{2}{3}\pi r^3 = \frac{2}{3}\pi (0.48/2)^3 = 0.0579 \text{ m}^3 \]

\[ P = \frac{(82.06)(298)/\{(115812/n) - 18.44\} - 1428283.133/(\{(298)^{0.5}\{(115812/n)\{(115812/n + 18.44)\}) \}
\]

where \( n \) = moles of hydrogen stored
External Storage Summary:

<table>
<thead>
<tr>
<th>Mass of Hydrogen Stored (grams)</th>
<th>Storage Pressure (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.54</td>
<td>0.0588</td>
</tr>
<tr>
<td>11.08</td>
<td>0.118</td>
</tr>
<tr>
<td>14.00</td>
<td>0.149</td>
</tr>
<tr>
<td>28.00</td>
<td>0.298</td>
</tr>
<tr>
<td>42.00</td>
<td>0.447</td>
</tr>
<tr>
<td>56.00</td>
<td>0.597</td>
</tr>
<tr>
<td>70.00</td>
<td>0.746</td>
</tr>
<tr>
<td>84.00</td>
<td>0.897</td>
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</tbody>
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Hydride and Bottled Storage Comparison

<table>
<thead>
<tr>
<th>Mass of Hydrogen Stored (g)</th>
<th>Exchanger Mass (kg)</th>
<th>Days of Storage</th>
<th>Exchanger Volume (cm³)</th>
<th>Number of Exchangers</th>
<th>Number of Bottles</th>
<th>Bottle Mass (kg)</th>
<th>Bottle Pressure (MPa)</th>
<th>Bottle Volume (cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.54</td>
<td>0.579</td>
<td>1.0</td>
<td>314.6</td>
<td>1</td>
<td>3</td>
<td>6</td>
<td>15.47</td>
<td>1320</td>
</tr>
<tr>
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<td>2.0</td>
<td>560.2</td>
<td>1</td>
<td>1</td>
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<td>5.92</td>
<td>2300</td>
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<td>689.7</td>
<td>1</td>
<td>1</td>
<td>7</td>
<td>7.84</td>
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<td>7.84</td>
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<td>7.84</td>
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</tr>
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<td>6</td>
<td>6</td>
<td>42</td>
<td>7.84</td>
<td>13800</td>
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### Bottled Gas Storage Pressures at 298 K

<table>
<thead>
<tr>
<th>Volume (cm³)</th>
<th>Mass of Hydrogen Stored (g)</th>
<th>Storage Pressure MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>2300 (7 kg 10 cm x 36 cm)</td>
<td>5.54 11.08 14.00 28.00 42.00 56.00 70.00 84.00</td>
<td>3.03 6.19 7.92 16.84 26.94 38.50 51.84 67.40</td>
</tr>
<tr>
<td>6900 (14 kg 15 cm x 23 cm)</td>
<td>5.54 11.08 14.00 28.00 42.00 56.00 70.00 84.00</td>
<td>0.993 2.01 2.54 5.18 7.92 10.77 13.74 16.84</td>
</tr>
<tr>
<td>126300 (86 kg 38 cm x 137 cm)</td>
<td>5.54 11.08 14.00 28.00 42.00 56.00 70.00 84.00</td>
<td>0.0539 0.107 0.137 0.272 0.410 0.547 0.685 0.823</td>
</tr>
<tr>
<td>438900 (590 kg 57 cm x 305 cm)</td>
<td>5.54 11.08 14.00 28.00 42.00 56.00 70.00 84.00</td>
<td>0.0155 0.0310 0.0392 0.0784 0.118 0.157 0.197 0.236</td>
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</tbody>
</table>
APPENDIX D

ENERGY REQUIREMENT CALCULATIONS
Energy Analysis of a LaNi₅ Alloy:

Energy Balance in Cylindrical Coordinates

\[
\rho C_p \left( \frac{\partial T}{\partial t} + V_r \frac{\partial T}{\partial r} + V_\theta \frac{\partial T}{\partial \theta} + V_z \frac{\partial T}{\partial z} \right) = k\left[ \left( \frac{1}{r} \right) \frac{\partial}{\partial r} \left( r \frac{\partial T}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 T}{\partial \theta^2} + \frac{\partial^2 T}{\partial z^2} \right] \\
+ 2\mu \left[ \left( \frac{\partial V_r}{\partial r} \right)^2 + \left( \frac{\partial V_\theta}{\partial \theta} + \frac{\partial V_r}{\partial r} \right)^2 + \left( \frac{\partial V_z}{\partial z} \right)^2 \right] \\
+ \mu \left[ \left( \frac{\partial V_\theta}{\partial \theta} \right)^2 + \left( \frac{\partial V_r}{\partial r} + \frac{\partial V_\theta}{\partial \theta} \right)^2 + \left( \frac{\partial V_z}{\partial r} + r \frac{\partial}{\partial r} \right)^2 \right] \\
+ S
\]

where \( S = N\Delta H \)

S > 0 for an endothermic reaction
S < 0 for an exothermic reaction

Simplification gives:

\[
\rho C_p \left( \frac{\partial T}{\partial t} \right) = k \left( \frac{1}{r} \right) \left( \frac{\partial}{\partial r} \right) \left( r \frac{\partial T}{\partial r} \right) + S
\]

Boundary condition 1: at \( r=0, \) \( q_r = \text{finite} \)

Boundary condition 2: at \( r=R, \) \( T=T_o \)

Initial condition: at \( t=0, \) \( T=T_o \) for all \( r \)

Integrate the right side of the energy balance equation:

\[
\rho C_p \left( \frac{\partial T}{\partial t} \right) \left( \frac{r^2}{2} \right) - \frac{Sr^2}{2} + C_i = k \left( r \frac{\partial T}{\partial r} \right) \quad \text{at B.C. 1, } q_r = \text{finite} \quad \therefore C_i = 0
\]

But \( q_r = -k \left( \frac{\partial T}{\partial r} \right) \)

Substitute and solve for \( q_r \)

\[
q_r = \frac{Sr^2}{2} - \rho C_p \left( \frac{\partial T}{\partial t} \right) \left( \frac{r^2}{2} \right) \quad \text{at B.C. 2, } \frac{T}{t} = 0
\]

Energy flux at the hydride container tube surface:

\[
q_r = \frac{Sr^2}{2}
\]
Energy Requirements for Hydride Cycling:

Calculate energy flux from absorption tube, heater requirements, and coolant flow rates:

1. Energy flux from a single hydride tube:

\[ q_r = \frac{Sr}{2} \quad \text{at} \ r=R, \ q_R = \frac{SR}{2} \]

\[ S = \text{volumetric energy source term} = \frac{n\Delta H}{V} \]

\[ \Delta H = \text{heat of absorption or desorption} \]
\[ n = \text{molar flow rate of hydrogen} \]
\[ V = \text{hydride volume} \]

Heat of absorption = $-30861.57 \text{ J/mole hydrogen absorbed}$

\[ n = 5.54 \text{ g H}_2/\text{day} \times \text{ mol H}_2/2.016 \text{ g H}_2 \times \text{ day/24 hr} \times \text{ hr/3600 s} \]
\[ n = 0.0000318 \text{ mol H}_2/\text{s} \]

\[ V = \frac{M_n}{\rho_n} = \frac{\pi D^2 \rho_n L}{\pi D^2} = \frac{(\pi/4)(1.219 \text{ cm})^2(40 \text{ cm/tube})}{\pi} \]
\[ V = 46.68 \text{ cm}^3 \text{ for a single tube of the reference exchanger} \]

\[ S = (0.0000318 \text{ mol/s})(-30861.57 \text{ J/mol})/(46.68 \text{ cm}^3) \]
\[ S = -0.0210 \text{ J/cm}^3\text{s} \]

\[ q_r = \left[(-0.0210 \text{ J/cm}^3\text{s})(1.270 \text{ cm}^2)/2 \right] \]
\[ q_r = -0.00667 \text{ watt/cm}^2 \text{ for each tube of the reference exchanger} \]

For the entire exchanger,

\[ q_{r, \text{TOT}} = \sum_{i=1}^{7} q_{R,i} = 7(-0.00667 \text{ watt/cm}^2) = -0.0467 \text{ watt/cm}^2 \]

Total surface area available for heat exchange:

\[ A = 2\pi r L = \pi DL = \pi(1.270 \text{ cm})(280 \text{ cm}) = 1117.2 \text{ cm}^2 \]
Energy release for absorption rate:

\[ Q = q_{\text{r, tot}}A = (-0.0467 \text{ watt/cm}^2)(1117.2 \text{ cm}^2) = -52.2 \text{ watts} \]

Water mass flow rate to sustain the absorption rate:

\[ -Q = mC_p \Delta T \]
\[ m = -Q/C_p \Delta T = -(-52.2 \text{ watts})(0.23901 \text{ cal/s\cdot watt})/(1.0 \text{ cal/g\cdot K})(5 \text{ K}) \]
\[ m = 2.5 \text{ g/s} = 0.0025 \text{ kg/s} \]

Energy required to heat the hydride from \( T_1 \) to \( T_2 \):

\[ Q = \rho_n V_n C_{p,n} \Delta T \]
\[ Q = (6.59 \text{ g/cm}^3)(7)(46.68 \text{ cm}^3)(0.435 \text{ J/g\cdot K})(T_2 - T_1) \]

Let \( T_1 = 298 \text{ K} \)

\[ T_2 = f(P_d) = -3712/\ln(P_d/P) -12.96 \]

Tabulated results of the energy calculation:

<table>
<thead>
<tr>
<th>Temperature, ( T_2 ) (K)</th>
<th>Plateau Pressure (MPa)</th>
<th>Energy Required (J)</th>
</tr>
</thead>
<tbody>
<tr>
<td>305</td>
<td>0.223</td>
<td>6556.9</td>
</tr>
<tr>
<td>310</td>
<td>0.272</td>
<td>11240.5</td>
</tr>
<tr>
<td>315</td>
<td>0.328</td>
<td>15924.0</td>
</tr>
<tr>
<td>320</td>
<td>0.395</td>
<td>20607.5</td>
</tr>
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<td>325</td>
<td>0.472</td>
<td>25291.1</td>
</tr>
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<td>330</td>
<td>0.561</td>
<td>29974.6</td>
</tr>
<tr>
<td>335</td>
<td>0.663</td>
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</tr>
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<td>0.915</td>
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</tr>
<tr>
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<td>48708.7</td>
</tr>
<tr>
<td>353</td>
<td>1.167</td>
<td>51518.9</td>
</tr>
</tbody>
</table>
Time for heat up and heater requirement for each exchanger:

\[ q_m = m_h C_p.m (dT/dt) \]
\[ q_{ht} = m_h C_p.m (T_2 - T_1) \]
\[ t = m_h C_p.m (T_2 - T_1)/q_m \]

Let \( T_2 = 353 \) K, \( T_1 = 298 \) K, and \( t = 300 \) s for the worst case:

\[ q_m = (1290 \text{ g})(0.435 \text{ J/g.K})(353 \text{ K} - 298 \text{ K})/300 \text{ s} \]
\[ q_m = 102.9 \text{ watts for each exchanger} \]

\[ t = (5.45 \text{ s/K})(T_2 - T_1) \]

Results of heating time calculation:

<table>
<thead>
<tr>
<th>Temperature, ( T_2 ) (K)</th>
<th>Plateau Pressure (MPa)</th>
<th>Heating Time (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>305</td>
<td>0.223</td>
<td>38.15</td>
</tr>
<tr>
<td>310</td>
<td>0.272</td>
<td>65.40</td>
</tr>
<tr>
<td>315</td>
<td>0.328</td>
<td>92.65</td>
</tr>
<tr>
<td>320</td>
<td>0.395</td>
<td>119.90</td>
</tr>
<tr>
<td>325</td>
<td>0.472</td>
<td>147.15</td>
</tr>
<tr>
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<td>0.561</td>
<td>174.40</td>
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<td>335</td>
<td>0.663</td>
<td>201.65</td>
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<td>340</td>
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<td>228.90</td>
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</tr>
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</tr>
<tr>
<td>353</td>
<td>1.167</td>
<td>299.75</td>
</tr>
</tbody>
</table>
Energy required to sustain the desorption reaction:
(The desorption reaction is endothermic)

\[ q_d = n \Delta H = (0.0000318 \text{ mol H}_2/s)(30859.55 \text{ J/mol H}_2) = 0.981 \text{ watts} \]

Energy required to cool the hydride from \( T_2 \) to the initial temperature, 298 K:

\[ q_c = m C_p (T_2 - T_1)/t \]
\[ q_c = (1290 \text{ g})(0.435 \text{ J/g.K})(298 \text{ K} - 353 \text{ K})/300 \text{ s} \]
\[ q_c = -102.9 \text{ watts} \]

Coolant flow rate to provide energy removal:
From the energy equation:

\[ q_r = S r/2 + \rho C_p (T/\text{t})(r/2) \]
\[ q_r = \rho C_p (\text{dT/dt})(r/2) \]
\[ \int q_r \text{dt} = -\rho C_p (r/2) \int \text{dT} \]
\[ q_r = \rho C_p (r/2)(T_2 - T_1) \]
\[ q_r = \rho C_p (r/2t)(T_2 - T_1) \]

\[ q_r = \{(3.95 \text{ g/cm}^3)(0.3995 \text{ J/g.K})(1.270 \text{ cm}/2)/2(300 \text{ s})\}(298 \text{ K} - 353 \text{ K}) \]
\[ q_r = -0.0919 \text{ watt/cm}^2 \]

\[ Q_c = q_r \Delta T = m C_p \Delta T \]
\[ Q_c = -(0.0919 \text{ watt/cm}^2)(1117.2 \text{ cm}^2) = -102.6 \text{ watts removed} \]

\[ m = Q_c / C_p \Delta T \]
\[ m = 102.6 \text{ watts}/(4.184 \text{ J/g.K})(282 \text{ K} - 277 \text{ K}) \]
\[ m = 4.18 \text{ g/s} = 0.00418 \text{ kg/s} \]

Net energy requirement for an entire cycle:

\[ Q_{\text{TOT}} = Q_d + Q_{\text{cool}} + Q_c + Q_r \]

Cycle from \( T = 298 \text{ K} \) to \( T = 353 \text{ K} \) and back to \( T = 298 \text{ K} \)

\[ Q_{\text{TOT}} = -52.2 \text{ watts} + 103 \text{ watts} + 0.981 \text{ watts} - 103 \text{ watts} = -51.22 \text{ watts} \]

51.22 watts must be removed
REFERENCES


Abstract

Storing hydrogen on board the Space Station presents both safety and logistics problems. Conventional storage using pressurized bottles requires large masses, pressures, and volumes to handle the hydrogen which will be used by experiments in the U.S. Laboratory Module and residual hydrogen generated by the ECLSS. Rechargeable metal hydrides may be competitive with conventional storage techniques. The basic theory of hydride behavior is presented and the engineering properties of LaNi₅ are discussed to gain a clear understanding of the potential of metal hydrides for handling spacecraft hydrogen resources. Applications to the Space Station and the safety of metal hydrides are presented and compared to conventional pressurized storage. This comparison indicates metal hydrides may be safer and require a lower pressure, less volume, and less mass to store an equivalent mass of hydrogen.

Key Words

Hydrogen
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Metal Hydride

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