# HYDRIDE ABSORPTION REFRIGERATOR SYSTEM FOR TEN KELVIN AND BELOW

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Recent work at JPL has shown that a very long-life, lightweight and efficient hydride absorption refrigerator system can be built to operate at ten Kelvin and below. The system consists of four basic stages of refrigeration. The first stage can be accomplished by means of an active refrigeration system such as a long-life Gifford-McMahon expander or a charcoal-sorption refrigerator, or even by a passive space radiator operating below 120 K. The second stage is operated by a hydride absorption system, wherein a heated hydride powder drives off high pressure hydrogen through a Joule-Thomson/heat exchanger expansion loop such that the hydrogen is partially liquefied (20 K at 1 atmosphere pressure). In the third stage, the vapor pressure over the collected liquid hydrogen is lowered by means of absorbing the hydrogen vapor onto a different low pressure, warm hydride. With a 1.7 torr partial pressure of hydrogen gas in the hydride, liquid hydrogen is solidified and sublimes at 10 K. Long-life adiabatic demagnetization refrigerators, helium desorption, or helium diaphragm compressors may possibly be used to cool to 4 K or below.

Preliminary analysis shows that the hydride concepts provide an extremely efficient means of refrigeration to 10 K, and that an entire sorption refrigeration process can be accomplished solely by using low grade heat energy at about 150°C. Furthermore, an entire sorption refrigerator system has no moving parts, other than long-life valves, which have been life-tested at JPL to an accelerated life of 500 years. Preliminary tests and analyses of the sorption refrigerators indicates an expected lifetime of at least ten years. Present lightweight Gifford-McMahon expanders have lifetimes greater than 2.3 years, although this can likely be increased by means of redesign and/or redundancy.

Key words: absorption; adiabatic demagnetization; adsorption; cryogenic; Gifford-McMahon; hydride; hydrogen; refrigerator; sublimation.

## 1. Introduction

There are a number of mechanical cryogenic refrigerator systems that are capable of producing temperatures as low as ten Kelvin and below. These systems, which include various Stirling cycles, Brayton cycles and Vuilleumier cycles, all require the use of wear-related moving parts and/or very complicated electrical equipment. Although much progress has been made in recent years, none of these systems has, to-date, demonstrated extremely long life (five years or greater), primarily because of mechanical failure problems. It was precisely for this reason that the Jet Propulsion Laboratory decided in 1979 [1] to explore the possibility of using non-mechanical sorption refrigeration systems for sensor cooling systems that would eventually explore the outer planets. These missions require lifetimes of ten years and more. With sorption refrigeration systems, there are no wear-related moving parts, and the controlling electronics can consist entirely of simple, long-life, solid state timers.

Recent work at JPL has demonstrated the feasibility of extremely long life (at least ten years) for hydride absorption coolers in the twenty Kelvin range [2, 3], and analyses has proven

that this temperature range can be lowered to ten Kelvin by means of using an additional, non-mechanical solid hydrogen sublimation stage. This extremely efficient stage then opens the door to even lower temperature, long-life systems when used as a preliminary stage for helium sorption systems or for adiabatic demagnetization refrigerators, both of which will be described later in this paper.

## 2. Principle of Operation

### 2.1 20 K Refrigeration

The basic principle of operation for the 20 K hydride refrigerator is that a continuous flow of high pressure hydrogen gas is generated by means of non-mechanical hydride compressors. The high pressure gas is pumped through a series of heat exchangers and a Joule-Thomson (J-T) valve such that the net cooling effect due to expansion of the gas at the J-T valve lowers the hydrogen temperature to its liquefaction point, 20 K at one atmosphere pressure (Figure 1).

The principle of the compressor operation is based on the fact that the intermetallic hydrides such as LaNi$_5$, absorb about six atoms of hydrogen per unit formula with an equilibrium pressure of a few atmospheres at room temperature. The density of the absorbed hydrogen at room temperature is almost twice the density of free liquid hydrogen at cryogenic temperatures.

The equilibrium pressure of the absorbed hydrogen is a very strong function of temperature, and thus the hydrogen static pressure can be increased from 4 atmospheres at 40°C (313 K) to about 60 atmospheres at 120°C (393 K). Cycling can also be achieved with the use of other metallic hydrides, whose temperature-pressure characterisics are different than LaNi$_5$. By alternately heating and cooling a series of hydride containers, a continuous flow of high pressure gas is thus supplied to the Joule-Thomson (J-T) expansion valve in Figure 1. The expansion of the gas causes partial liquefaction which can then absorb heat at liquid hydrogen temperatures.

The basic advantages of this type of system are that it provides considerably lower temperatures (20 K or lower) than those from passive radiator systems (approximately 80 K), and yet it still has no mechanical moving parts, other than self-operating check valves, which have been life-tested at JPL for an accelerated life of 500 years. It is much simpler, and has a much longer life expectancy than present orbiting mechanical refrigeration systems, and it can be operated using low temperature waste heat or direct solar heat.

In order to provide heating and cooling of the hydride compressors, a simple gas-gap thermal switch can be used (Figure 2). For spacecraft application, heating could be obtained by means of a flat plate solar collector (or possibly a radioactive thermal energy source), and the heat could be transferred to the hydride by injecting a gas into the gold-plated gap between the solar plate collar and the hydride. To cool the compressor, the solar plate thermal switch gap would be evacuated, and gas would be injected into the gap between the compressor and the cooler radiator heat pipe. Low pressure hydrogen, e.g. 1 torr, gas could be used as the thermal switch actuator gas, and this could be stored in a cooled hydride material for very long-life and light-weight storage.

### 2.2 10 K Refrigeration

In order to produce temperatures as low as 10 K, the vapor pressure over collected liquid hydrogen is lowered by means of absorbing the hydrogen vapor onto a very low pressure, warm hydride. With a 1.7 torr partial pressure of hydrogen (corresponding to a hydride temperature of about 0°C), the hydrogen is solidified and sublimas at 10 K (Figure 3). A cryogenic sensor can be continuously maintained at 10 K by means of providing two dewars of hydrogen. While one is being liquified, the other is sublimated. The cryogenic sensor can be attached to both dewars by means of a thermal switch, thus being continuously maintained at 10 K.

Another design for the 10 K stage involves liquefying hydrogen at 20 K by means of a hydride liquifier stage, and then expanding the liquid directly to a low pressure 10 K solid hydrogen "snow". Blockage of the J-T valve could be prevented by means of using a porous copper trap, wherein sensor heat is readily transmitted to the J-T valve itself.

### 3. Overall Cycle Possibilities

#### 3.1 Multi Stage Sorption Refrigeration System

In order to produce an entire spacecraft refrigeration system, a number of possibilities exist in terms of refrigerator staging. Although some spacecraft applications (such as sun-synchronous polar orbits, geosynchronous orbits, and interplanetary missions) adapt themselves well for very low temperature passive radiator stages in the 80 K to 120 K region, most low earth orbit applications cannot readily achieve radiator temperatures below about 150 K. Since this results in relatively high specific power requirements for the hydride system, a number of alternate colder upper stage refrigeration systems have been studied.

For one type of system, solar heat could be collected at 425 K, and used to power four sorption stages (Figure 4), while a small amount of electrical power is used for a thermoelectric cooler (TEC) stage. For the charcoal/methane and charcoal/nitrogen sorption stages, large quantities of gas are absorbed onto charcoal at low pressure and 225 K. The gas is then released at high pressure when the charcoal is heated to 425 K. The process is similar to the hydride system, except that the gases are physically adsorbed onto the surface of the charcoal, while the hydrogen is chemically bonded to the metal hydride powder. The charcoal sorption stages and the TEC are cooled by a 225 K radiator, a temperature which is readily obtainable for radiators facing away from earth, with or without solar light, in low earth orbit. The TEC provides cooling to 175 K, which liquifies methane at high pressure (26.5 atm). When the liquid methane is expanded to 1.8 atm, it cools to 120 K. This 120 K stage is then used to liquefy nitrogen at 24.8 atmospheres. When the nitrogen expands to 1.4 atm, it cools to 80 K, which is then staged to a rare earth, misch metal hydride compressor to reach 18 K for liquid hydrogen. The refrigerator system then cools to 10 K for the solid hydrogen sublimation stage.

A sorption system design to obtain one watt of cooling at 10 K requires a total estimated power of only 600 watts, most of which is supplied directly as solar heat. This is therefore the most efficient 10 K refrigeration system presently possible, primarily because it does not require the use of a very inefficient helium refrigeration cycle. Prior to the hydride sublimation stage design, it simply had not been possible to achieve such low hydrogen pressures for extended periods in a closed-cycle operation. It should also be mentioned that the entire refrigeration system uses absolutely no wear-related moving parts, other than extremely long-life, room-temperature valves. All calculations are based on actual empirical absorption/adsorption data for the respective gases.

A charcoal/helium sorption refrigeration stage, similar to the methane and nitrogen stages, may be possible to obtain temperatures of about 4K, but preliminary calculations have shown that is likely to take many kilowatts of charcoal sorption heating to produce enough helium flow to generate one watt of charcoal/helium J-T cooling. A likely alternative to a charcoal/helium sorption compressor, however, is an oil-free helium diaphragm compressor, although much development work is necessary in order to obtain a contamination-free life of ten years. Another lower stage alternative is to desorb helium from a saturated 10 K charcoal bed. The heat of desorption is relatively high compared to the very low charcoal specific heat at this temperature, and substantial cooling can result. Although temperatures below 4 K have been obtained with this method [4], much development is still needed in this area also.

#### 3.2 Gifford-McMahon Pre-Cooling

Another extremely efficient means to achieve first stage cooling for the hydride system is to use a Gifford-McMahon (G-M) refrigeration cycle [5], which is powered by compressed hydrogen from a hydride system. In the G-M cycle, gas is compressed in a cylinder to a high pressure and the heat of compression is rejected at some temperature, typically room temperature. The gas is then forced through a piston-type cold regenerator that has a low cycle rate and low pressure drop seals. The gas is then allowed to expand to a very low pressure while flowing back through the regenerator and into the low pressure end of the compressor. The isentropic expansion of the gas precools the regenerator for the next cycle. Although not as efficient as some other refrigeration cycles, one of the big advantages of this system is that the primary friction-related part is a piston seal, which has a low cycle rate and a very low pressure drop across it. In fact, present Cryogenic Technology Incorporated (CTI) Gifford-McMahon refrigerators have expansion cooler mean-time-between- failure (MTBF) lifetimes in excess of 20,000 hours [6]. Since present CTI expansion coolers are run at about 72 RPM, if lower speeds or stronger, multiple seals are used, even greater MTBFs can be expected. Also, since a 9 kg expander can provide about 15 watts of cooling at 70 K, multiple expanders could be used and valved off, if the low pressure drop expansion seal begins to show signs of degraded life.

At present, the major problem with G-M upper stage refrigerators is that oil-lubricated compressors are used. Although elaborate oil cleaning systems have been devised, eventually the oil will freeze out and contaminate the cryogenic portions of the system. This requires periodic maintenance every 2000 to 5000 hours. With a hydride compressor, however, there is absolutely no oil present, and the entire system can operate free of contamination, essentially forever.

A sketch of a G-M upper stage expander and a lower stage adiabatic demagnetization refrigerator, discussed in the next section, appears in Figure 5. Pre-cooling of the hydrogen gas at about 70 K can be accomplished with the G-M expander. The overall specific power, i.e. power required per cooling watt, of this system is relatively good, and in-fact, approaches approximately 300 for 20 K cooling, and is about 400 for 10 K when combined with a hydride sublimation cooler. This compares with about 1000 or higher for most other mechanical systems at 10 K.

### 3.3 Adiabatic Demagnetization Lower Stage

An adiabatic demagnetization (ADM) stage, which purportedly achieves 80% of carnot efficiency [7, 8] can be used to reach temperatures of 4 K or lower. For ADM, a wheel of paramagnetic material is rotated partly inside an extremely high superconducting magnetic field, typically about 7 Tesla. The application of the magnetic field causes heat to be generated in the paramagnetic material. This heat can then be removed at some elevated temperature, e.g. 10 K. When the wheel is rotated away from the 10 K heat sink, it rotates out of the high magnetic field, and its temperature is lowered a tremendous amount, typically to 4 K or below. The entire cycle can be designed to use no moving parts, other then a slowly rotating wheel and possibly some low pressure drop helium circulators to enhance heat transfer into and out of the paramagnetic material.

The 10 K or 14 K hydride stage can be of great advantage for the ADM refrigerator, since ADM requires extremely strong super conducting magnets in order to operate. Since 14 K is just within the superconducting magnet temperature region, the system may be able to "bootstrap" itself from 14 K to lower temperatures by progressively increasing superconductive magnetic field strength with decreasing temperature.

### 4. Experimental Results

### 4.1 Charcoal Sorption Refrigeration

Recent work at JPL has confirmed the use of charcoal sorption to produce a continuous flow of high pressure nitrogen. By alternately heating and cooling from cannisters of charcoal between 250 K and 400 K, a continuous flow of 50 atmospheres (750 psi) nitrogen has been produced. By flowing the gas through a J-T/heat exchanger assembly, a total cooling power of approximately 1/2 watt has been generated.

Although the total average power required for this proof-of-principle test was approximately 200 watts, much lower $LN_2$ specific powers are predicted with methane-staged sorption refrigeration, as previously described and shown in Figure 4.

### 4.2 Liquid Hydrogen Refrigeration

Most of the experimental results regarding liquid hydrogen hydride refrigeration life testing are available in [3, 9], and are summarized for convenience below:

In particular, a complete heated hydride compressor system has been thus far successfully tested at ERGENICS Corporation to 6000 hours of continuous operation, and the JPL cryogenic J-T system has accumulated over 1,000 hours of successful operation between 14 K and 29 K, without any evidence of wear. It should be noted that the ERGENICS compressor that operated for 6000 hours weighed only about 10 kg and yet delivered a hydrogen flow which would have been equivalent to about 3 watts of cooling at 25 K if a J-T/heat exchanger loop were used. With a lower cycle speed, it can be said that it survived at least 18000 hours of 1 watt equivalent cooling and yet still performed close to original specifications. Preliminary third-stage experiments have confirmed the sublimation of hydrogen at 13.8 K, although a reduced pressure drop heat exchanger system is required to reach lower temperatures. Furthermore, the only moving parts in the system, room-temperature check valves, have been life-tested at JPL to an accelerated life of 500 years [10].

Life tests on hydride particle size have confirmed that the hydride particles break down after repeated cycling, but the average particle size reaches a final spherical equivalent diameter of about 1 micron after about 5000 cycles. Although some binary hydride alloys, such as LaNi$_5$, have shown a significant loss of capacity when subjected to many cycles at elevated temperatures [11], small additions of aluminum to rare earth, nickel-type alloys make the alloy greatly resistant to disproportionation. [12]. ERGENICS Corporation has estimated effective hydride disproportionation as approximately 1% per year, based on aluminized misch-metal hydride alloys. For expansion cooling, Joule-Thomson (J-T) valves have long proven their inherent reliability, as evidenced by well over one million hours of accumulated life for the JPL Deep Space Network Maser receivers [13]. Although simple decontamination thaws are recommended every six to twelve months for the JPL maser fixed-orfice, J-T valves, it is likely that no thaws would ever be needed for the new JPL design of non-clogging, spring-loaded, J-T valves [3]. Furthermore, the hydride system has absolutely no oil-contaminated vapors, as are present on the JPL mechanical refrigeration Maser compressors and thus overall contamination can be substantially reduced if proper initial decontamination steps are taken.

### 4.3 Solid Hydrogen Sublimation

There has been considerable work done on solid hydrogen sublimation by Lockheed Corporation [14]. Although hydrogen solidifies at about 14 K and 1 psi pressure, its temperature has been readily lowered to as low as 7 K by means of evacuating the vapor space above the hydrogen, thereby causing it to sublimate directly from the solid phase to the gaseous phase. Since the transition heat from gas to liquid is very high (190 BTU/lbm) and the solid latent heat is relatively low (25 BTU/lbm), the amount of cooling energy required to go from liquid to solid is quite low, although the heat of sublimation, which is approximately 190 + 25 = 215 BTU/lbm, is very high.

That is to say, once liquid hydrogen has been made, it takes very little extra power to produce solid hydrogen. This in fact, has been confirmed by analysis using a specialized JPL sorption computer program [15], using LaNi$_5$ to produce liquid hydrogen and a low pressure hydride to further reduce the vapor pressure, and thus its temperature.

### 5. Summary and Conclusions

Recent work at JPL has shown the possibility of achieving extremely long-life (10 years or greater), efficient, and relatively simple refrigeration systems for obtaining 10 K temperatures and below. The various possible stages are described below:

1.  The first stages of refrigeration can be obtained by either passive radiation (80 K to 120 K for high orbits or interplanetary missions), charcoal/methane sorption (120 K) combined with charcoal/nitrogen sorption (80 K), or by a Gifford-McMahon refrigeration system (40 K minimum) powered by a hydride compressor.

2.  The next stage of refrigeration to 20 K can be accomplished by a standard hydride J/T compressor system, such as the compressor unit recently life tested for 6000 hours [3].

3.  The ten Kelvin temperature stage is obtained by low pressure hydride sublimation evacuation of the vapor space above hydrogen to 1.7 torr pressure. This can be accomplished by batch processing the liquid hydrogen and then reducing the hydrogen pressure by absorption onto the hydride or by direct expansion of liquid hydrogen through a J/T valve to produce solid hydrogen "snow".

4.  Temperatures of 4 K or below may possibly be obtained by adiabatic demagnetization refrigeration, helium desorption, or by an oil-less diaphragm helium compressor.

All of the sorption refrigeration stages have life expectancies of at least ten years, since their only moving parts are room-temperature valves that have been life tested to an accelerated equivalent life of 500 years. Although present CTI Gifford-McMahon expanders have a guaranteed life expectancy of 20,000 hours, their extremely light weight allows for multiple unit redundancy and/or stronger seals and lower cycle frequency to achieve 10 year life (88,000 hours). It is quite likely that longer life seal materials can be made, since the G-M expander seal sees only a very low pressure drop which is at low temperatures. Present G-M refrigerators are life-limited primarily due to oil-contamination from mechanical compressors, a problem which is totally absent with hydride compressor systems.

The overall specific power efficiency to reach 10 K by sorption staging is estimated to be about 600 watts per cooling watt at 10 K, while the specific power with G-M staging is about 400 watts per cooling watt. Most of the power for both systems is in the form of low temperature heat, such as 425 K solar heat or RTG waste heat. This compares with values generally greater than 1000 for every other known 10 K helium cycle refrigeration system.

When compared with all other closed-cycle spacecraft mechanical refrigeration systems, sorption refrigeration, and in particular the 10 K hydrogen sublimation hydride sorption system, appears to be the simplest, most efficient, lightest and least interfering (vibrations and magnetics) and to have the longest potential life of any other existing system.

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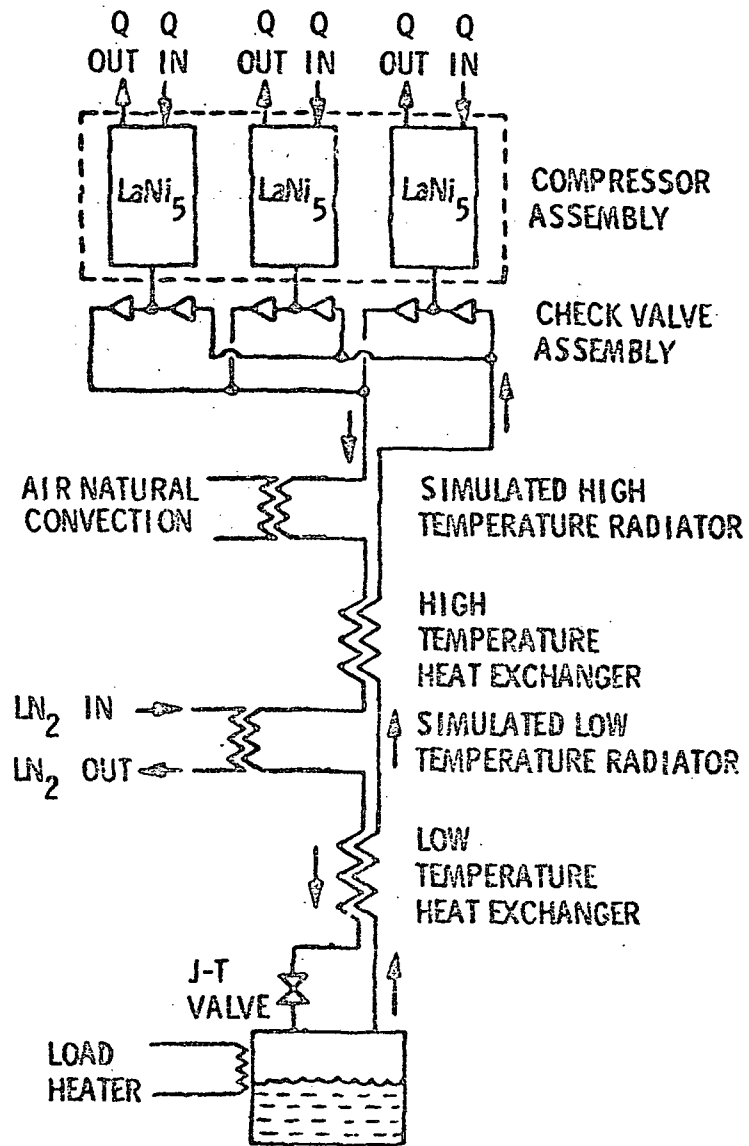
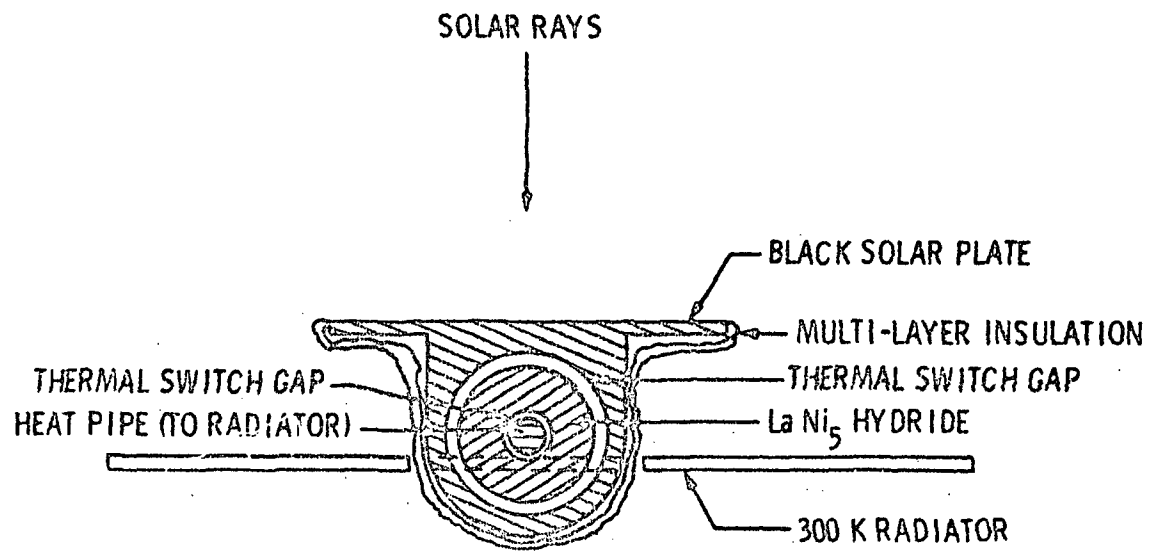Figure 1    Hydride Refrigerator Operational Schematic

SOLAR RAYS

BLACK SOLAR PLATE

MULTI-LAYER INSULATION

THERMAL SWITCH GAP

THERMAL SWITCH GAP

HEAT PIPE (TO RADIATOR)

$La\ Ni_5$ HYDRIDE

300 K RADIATOR

Figure 2    Hydride Compressor Design With Thermal Switches

LOW PRESSURE
HYDROGEN
ABSORBENT

HYDROGEN
GAS FLOW

2 TORR PRESSURE
(.0026 ATM)

SUBLIMATION

SOLID
HYDROGEN
(10 KEVLIN)
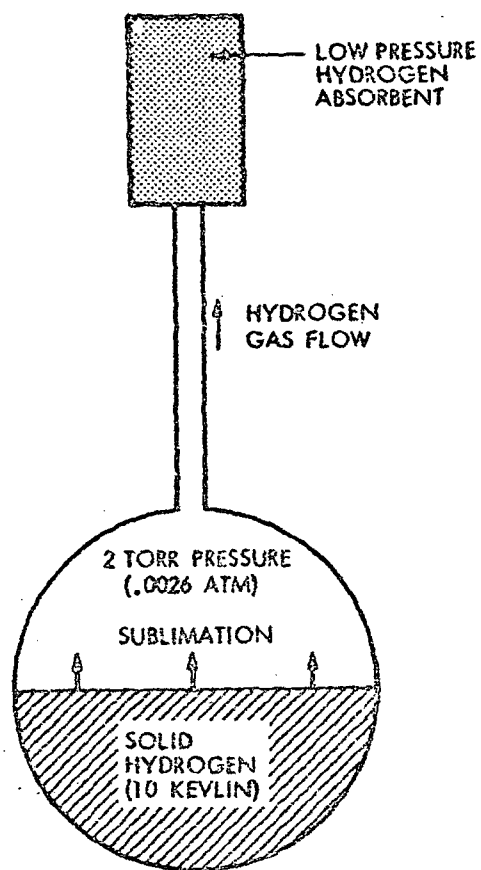
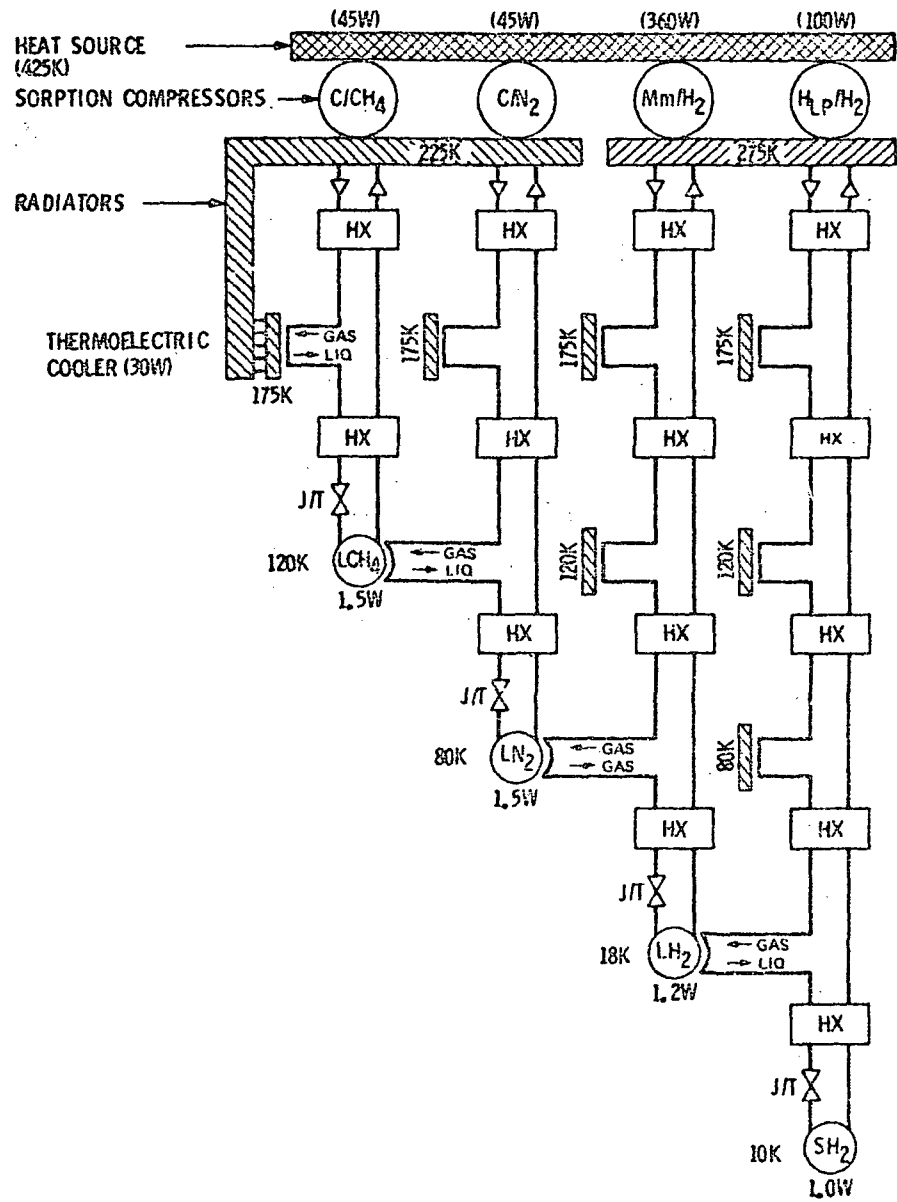Figure 3    Ten Kelvin Hydrogen Sublimation Refrigerator
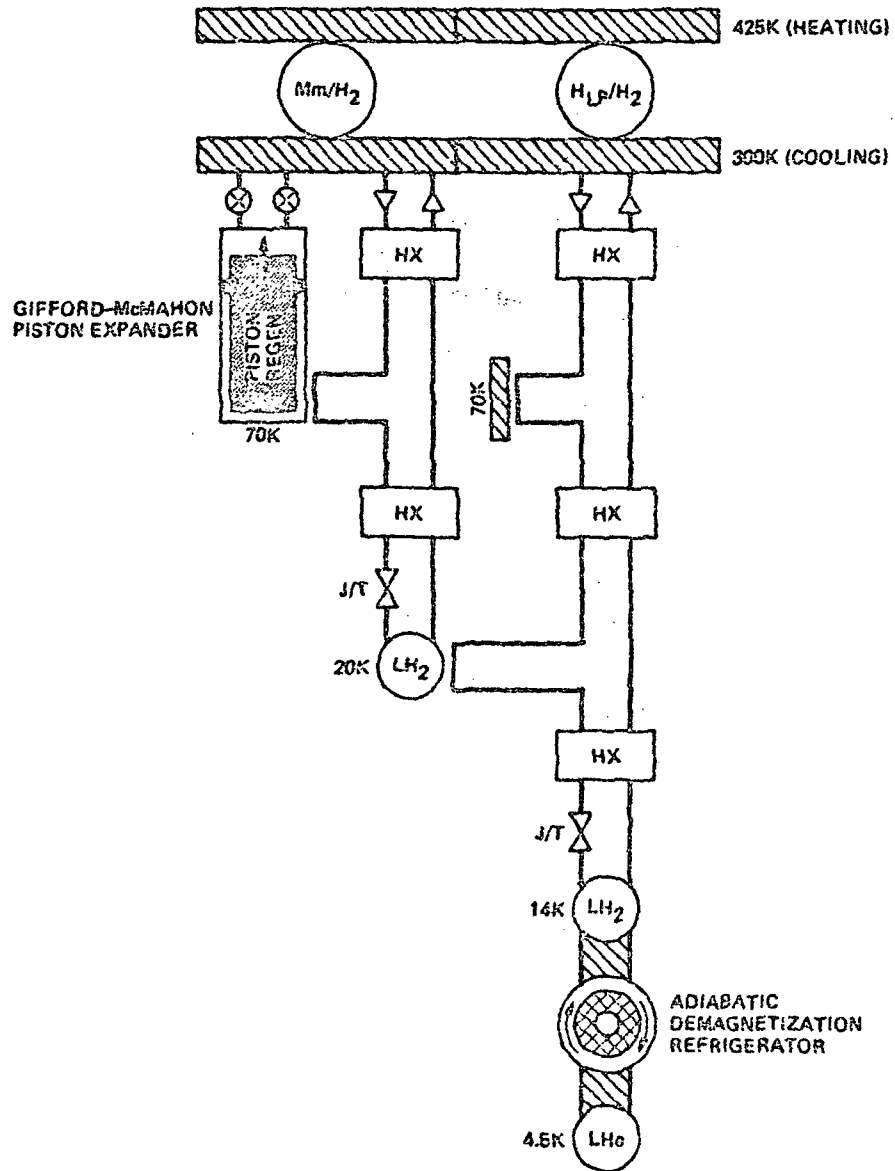
Figure 4    Four Stage Sorption Refrigerator System

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Figure 5    Hydride Refrigerator With G-M Upper Stage and ADM Lower Stage