

Large-Scale Production of Densified Hydrogen Using Integrated Refrigeration and Storage

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Nomenclature

dP/dT	=	rate of pressurization/depressurization
IRAS	=	Integrated Refrigeration and Storage
LH ₂	=	liquid hydrogen
LN ₂	=	liquid nitrogen
LOX	=	liquid oxygen
NBP	=	normal boiling point
TP	=	triple point
ZBO	=	zero boiloff

I. Introduction

Low molecular weight of the combustion products and resulting high specific impulse make liquid hydrogen (LH₂) the most efficient fuel for chemical and nuclear rocket applications. But because LH₂ possesses the lowest normal boiling point (NBP) and lowest density of all the propellants, it is more difficult to use and more costly than other fuels. Densified hydrogen, which is a liquid cooled to below the NBP, increases density by up to 8.8%, down to the triple point (TP) temperature, and provides a sensible gain in liquid heat capacity before boiling. One benefit to using densified propellants is reduced tank volumes, which can reduce the gross liftoff weight of a launch vehicle and increase its payload capacity. Another benefit is increased heat gain before boiloff (assuming that the storage tank is maintained at a constant pressure), which translates into longer storage times, increased cooling capacity, and reduced operational cost.

Because of these benefits, the aerospace industry has investigated densified hydrogen for decades. The National Bureau of Standards quantified the thermodynamic properties of densified and slush hydrogen in the 1960s, and Martin Marietta studied densified propellants for a single-stage-to-orbit launch system in the late 1970s [1]. From 1988 to 1994, NASA Glenn Research Center (GRC) worked on slush hydrogen production and transfer for the National Aerospace Plane (NASP), producing slush in 700-gallon batches using evaporative cooling in a freeze/thaw cycle [2]. GRC continued development for another 8 years and made several advances working with the X-33 Program, including two densifier units (first a 2 lb/s unit and then an 8 lb/s unit) using evaporative cooling in a load-and-go process [3], and several transfer and flight tank loading demonstrations with both liquid oxygen (LOX) and LH₂ [4]. At that time, the Space Shuttle Program considered switching to densified propellants as an upgrade to launch more mass to the International Space Station [5], but because of operations concerns and engine test uncertainty, the super-lightweight external tank was modified instead. NASA continued to develop densifier systems with the 2nd Generation Reusable Launch Vehicle Program, funding three separate contractors to build prototype units and investigate refrigeration technologies [6]. Around this same time, small-scale densified hydrogen production using a Gifford McMahon (GM) refrigerator was performed by NASA Kennedy Space Center (KSC) and the Florida Solar Energy Center [7]. Although NASA has yet to fly a rocket using densified propellants, SpaceX is now using densified LOX and RP-1 on the Falcon 9 vehicle.

II. Ground Operations Demonstration Unit for Liquid Hydrogen

Recognizing the need for more capable and efficient cryogenic systems, NASA's Advanced Exploration Systems (AES) Program funded the development of the Ground Operations Demonstration Unit for Liquid Hydrogen (GODU LH₂) in 2012. The GODU LH₂ project is a relevant-scale prototype of an advanced LH₂ system using Integrated

Refrigeration and Storage (IRAS) technology to enable new types of operations not previously possible at the launch pads. IRAS couples state-of-the-art reverse-Brayton-cycle cryogenic refrigerators with liquid storage dewars using a submerged cold heat exchanger to remove energy directly from the fluid. If the refrigeration capacity matches the heat leak, a range of zero boiloff (ZBO) operations are possible. If the refrigerator capacity exceeds the heat leak, the liquid can be conditioned or densified. The GODU LH₂ project had three primary demonstration objectives for using IRAS on a relevant scale: (1) zero-loss storage and transfer of LH₂, (2) densification of LH₂, and (3) liquefaction of gaseous hydrogen (GH₂). This paper focuses on the results of the densification objective, but details on the other tests objectives are now available as well [8].

The functional diagram of the GODU LH₂ system is given in Figure 1. The GODU LH₂ system uses a 125 m³ vacuum-jacketed (VJ) IRAS tank with multilayer insulation as the primary storage vessel. This tank was previously used on Launch Complex 41 for the Titan-Centaur Program. Pretest analysis estimated the heat leak to be between 300 W and 400 W. The IRAS tank is instrumented to record tank pressure and 20 internal liquid and vapor temperatures. Three temperature rakes with Si-410 silicon diodes accurate to ±0.1 K are installed. The forward and aft rakes include arms to measure temperatures near the wall away from the centerline, whereas the center rake has only centerline diodes for better vertical resolution. The inner tank was modified with stiffening rings to allow for subatmospheric operation [9]. Inside the IRAS tank is a cold heat exchanger (CHX), using gaseous helium which is connected to a cryogenic refrigerator. The CHX consists of 1" tubular stainless-steel supply and return headers located at the 25% and 75% fill levels and connected by 40 parallel ¼" tubes that make up the bulk of the CHX surface area of approximately 900 ft². Two of these tubes are instrumented with diodes to record helium inlet and outlet temperatures. The CHX is supplied with cold gaseous helium (GHe) from a Linde LR1620 refrigerator and RSX compressor at approximately 22 g/s. This system uses a reverse Brayton cycle with piston expansion engine and includes provisions for liquid nitrogen (LN₂) precooling. The refrigerator has a rated capacity of 390 W at 20 K *without* LN₂ precooling and up to 880 W at 20 K *with* LN₂ precooling. The refrigerator has independent command and control and instrumentation, notably for expander speed and cold helium supply and return temperature. The RSX compressor is cooled by a separate 96 kW circulating water chiller at 292 K, and includes systems for oil separation and gas management. The entire refrigeration system is containerized to allow for transportation to other test facilities.

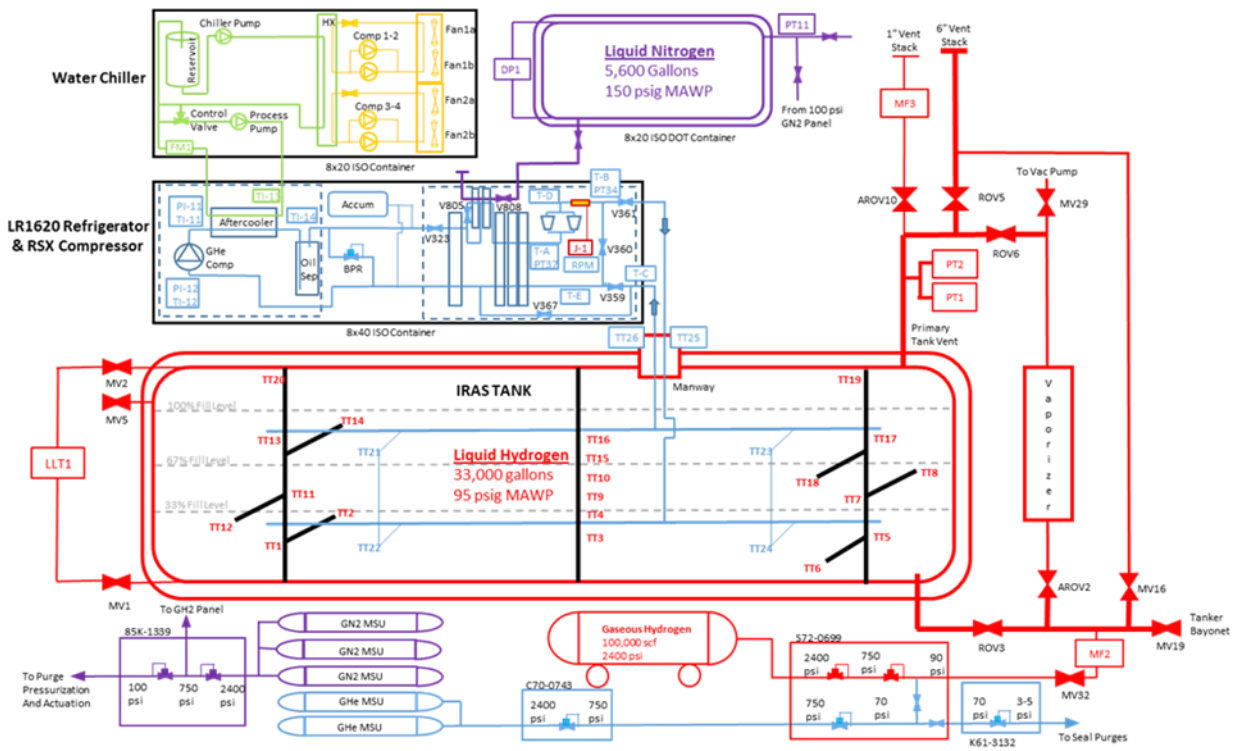


Figure 1. GODU LH₂ System Functional Diagram

In addition to the IRAS tank and refrigeration system, the GODU LH₂ site included the usual support systems, such as pneumatics, command and control, transfer, communications, and instrumentation. The pneumatic system

included fixed gaseous nitrogen and GHe storage bottles and provisions for tube trailers of GH₂. The transfer system includes the VJ transfer lines, vent lines, vaporizer, and associated manual and remote-control valves. The command and control/data acquisition system had to integrate separate systems for the refrigerator and chiller, as well as all the active components at the site for operators in the control room. The test site also has various ancillary subsystems for ground power, vacuum, and safety support.

III. Test Plan

The GODU LH₂ test matrix included testing of each of the primary objectives at three different fill levels—33%, 67%, and 100%—with one delivered tanker of LH₂ filling roughly 1/3 of the tank. For the zero-loss-storage objective, ZBO tests were performed at each liquid level, using three different control methods: helium supply temperature control, IRAS tank pressure control, and on/off duty cycling. For the zero-loss-transfer objective, all tanker supply operations were performed with no venting losses, saving roughly 10% more of the delivered LH₂ quantity than typical KSC practices. The initial cooldown of the tank was done using the refrigerator to cool the GH₂ in the tank to the NBP.

Densification tests are conducted at each of the three liquid levels. During each densification test, all the tank liquid and vent valves are closed and the tank is locked up. The refrigerator is run at full power mode with LN₂ precooling active. During densification operations, the IRAS tank pressure and liquid temperature decrease as long as the refrigerator capacity is greater than the tank heat leak. No helium gas pressure is applied to the ullage, and the tank pressure does go below atmospheric pressure. When the IRAS tank reaches the NBP, a low-pressure helium gas purge is applied to the stem seals and backside of all isolation valves and around the perimeter of all the flanges to prevent the intrusion of atmospheric air in the event of a leak. Eventually the system reaches an equilibrium temperature where the cryocooler refrigeration power equals the ambient heat load on the tank and refrigeration system. The pretest estimated minimum temperature for the system was 15 K. Data are collected on heat exchanger and refrigerator performance and on the response of the liquid and vapor in the tank.

IV. Densification Test Results

All three of the densification tests successfully met the pretest prediction of 15 K bulk liquid temperatures. The first two tests, at the lower liquid levels, reached the TP, and slush hydrogen was produced. The final test was progressing toward the TP, but testing was terminated due to the end of the project life at the end of fiscal year 2016. Details of each individual test, for the three different liquid levels, are discussed below.

A. 33% Liquid Level

Densification testing at the 33% fill level occurred in late March and early April 2016. The testing started immediately after the 33% tanker offload. The maximum pressure reached during the tanker offload was 47 psia. The refrigerator was turned on and cooling was initiated in the tank at this point. After the refrigerator was turned on, the ullage destratified and the pressure quickly reduced to the saturation pressure at the liquid temperature as the refrigerator cooled the bulk liquid along the saturation line. The dP/dT rate during this time was fairly steady around -0.15 psi/hr. This continued until the IRAS tank pressure reached 14.7 psi at 0830 on March 24, designated as T-0 on the following charts.

The test period shown in Figure 2 covers the entire time that the tank pressure was subatmospheric. The refrigerator was operating until T+330 hours, when the helium compressor failed and needed to be replaced. The response of the tank pressure (PT2) shows that the rapid decrease in the pressure gradually slows down and the depressurization rates decrease. There are two spikes in the PT2 curve that occurred when the the refrigerator was temporarily shut down for valve adjustments. A third event at T+256 hours, shown in the

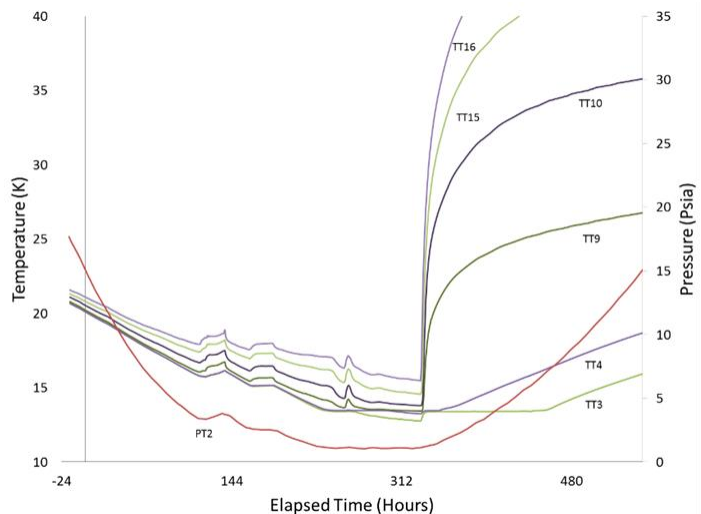


Figure 2. Tank Pressure and Center-Rake Temperatures, 33% Densification Test

hydrogen temperatures, is caused by a lapse of LN₂ precooling while waiting for another LN₂ tanker delivery. The tank pressure stabilizes around 1.1 ±0.05 psia, but never quite reaches the defined LH₂ TP of 1.02 psia. However, because of the behavior of the pressure and temperatures at the end of the test, it is evident that solid phase change was occurring. The slightly higher PT2 reading may be due to instrumentation bias or it may be a slight increase in total pressure in the tank as a result of partial pressure of helium from a leaking fitting on the refrigerator. The pressure started to increase as soon as the refrigerator turned off.

The temperature profile on the center rake also demonstrates that the liquid reaches the TP. The temperatures decrease steadily, although there is noticeably more stratification as the liquid becomes densified. This trend is evident even before the refrigerator problems, but becomes more pronounced afterward. At T+240 hours, the liquid temperatures reach 13.5 K and hold steady there while the ullage temperatures continue to decrease. This is colder than the TP. When the pressure and temperature are held constant and heat is being removed from the tank, it is evident that a liquid/solid phase change is occurring. After leveling out at 13.5 K for a period of time, some liquid temperatures decrease again beyond 13.5 K. Figure 3 shows a closer look at all the temperatures in the tank around this time. TT1 through TT6 are all in the liquid region and all show the same behavior, leveling out between 13.3 K and 13.5 K around T+240 hours. The ullage temperatures continue to drop, decreasing the stratification. Then, starting at T+260 hours with TT5, the liquid temperatures start to decrease below 13.5 K. TT1 and TT3 are next to follow the trend, finally accompanied by TT6, TT4, and then TT2. The order in which the temperatures decrease follows the relative proximity of the diode to the CHX inlet inside the tank. The spread of this supercooling shows the variable nature of the CHX performance during transient periods. The lowest temperature recorded is 12.55 K on TT5, right before the compressor shuts down, with the diodes still decreasing in temperature. This value is a full 1.25 K below the TP temperature.

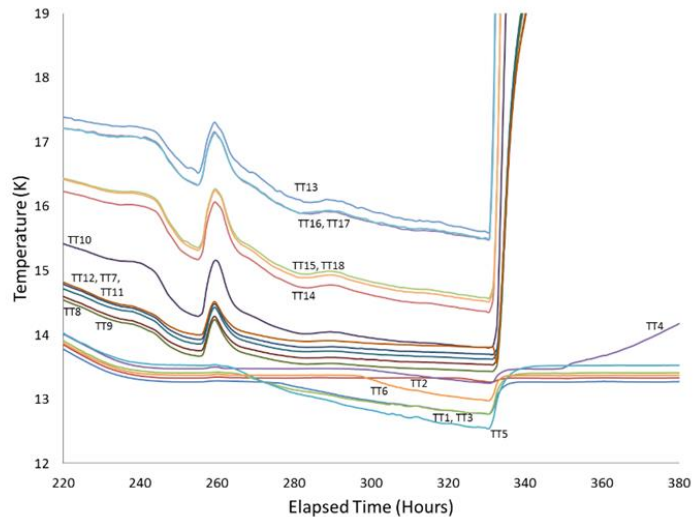


Figure 3. Detailed Tank Temperatures, 33% Densification Test

Further effort is needed to explain how such cold temperatures were achieved. The first thought was instrumentation error. But the Si-410 diodes are accurate to ±0.1 K between 2 K and 25 K. During boiloff testing, comparisons between recorded liquid temperatures (TT1 through TT6) and the calculated saturated liquid temperatures based on tank pressure show a general bias downward of between -0.14 psi and -0.25 psi. And the fact that all the diodes in the liquid responded similarly seems to indicate that this phenomenon is real. From an energy balance on the system (see refrigerator performance below), we can see that while solid hydrogen was being created, there was not enough time or cooling to solidify all the LH₂. Because the diodes are located some distance from the CHX surface, it is unlikely that they are encapsulated in solid hydrogen ice. Based on this unlikelihood, combined with the spread of cooling through the tank, which appears to be convective, it is concluded that the diodes are reading actual liquid temperatures that are below the TP. There are two possible explanations for this. The ultrapure LH₂ may not be freezing and may be in an unstable supercooled liquid. Or the helium gas may be being mixed into the ullage, and the system may no longer be a pure fluid and may need to be treated as a mixture. In either case, as soon as the refrigerator is turned off, these supercooled temperatures increase back to the near TP temperature and the liquid temperature is steady for a period of time while the slush fully melts.

The overall timeline of the test and knowing the tank heat leak and refrigerator performance allow the amount of solid hydrogen that is formed to be estimated. Looking at the representative data from TT3, we see that the diode reaches its steady phase change temperature of 13.4 K at T+237 hours. The temperature remains at or below that point until the compressor shuts off at T+330 hours, for an overall time of 93 hours. The refrigerator performance during that time shows that the average lift was approximately 721 W, and with a measured heat leak of 290 W, the tank was seeing a net removal of heat equal to 431 W. But much of this refrigeration is being removed from the top of the CHX in the ullage, and finding a true balance of energy between the liquid and vapor requires more information. After the refrigerator was shut down, the supercooling quickly disappeared and TT3 was reading steady at 13.4 K between T+334 hours and T+438 hours. During this 104-hour period, the heat leak into the liquid portion of the tank was

causing the hydrogen slush to melt at constant temperature. During the boiloff testing, the total heat leak of 290 W was estimated to be split between the liquid region (170 W) and the vapor region (120 W). So if it requires 170 W of heat leak to melt hydrogen for 104 hours, an estimated 1052 kg of solid hydrogen is produced. With the estimated mass in the tank of 4080 kg, this is a solid mass fraction of roughly 26%.

Figure 4 shows the refrigerator performance during the 33% densification test. Despite the LH₂ temperatures that were achieved, the performance could have been improved. The engine speed was steady at 227 RPM until T+48 hours. Then the speed started decreasing until T+120 hours, when a series of valve adjustments were performed, which helped the net lift. The net lift started around 950 W with a return temperature of 21.7 K, but had decreased to around 700 W at 14.5 K by the end of the test. The helium temperatures were dropping for the majority of the test but eventually settled down at a supply temperature of 9.8 K and a return temperature of 14.6 K while slush was being produced. At T+330 hours, the helium compressor failed and the liquid was allowed to warm back up to the NBP. The stored refrigeration energy in the densified liquid allowed the tank to be locked up for 18 days while the compressor was replaced, and no hydrogen was lost during that time.

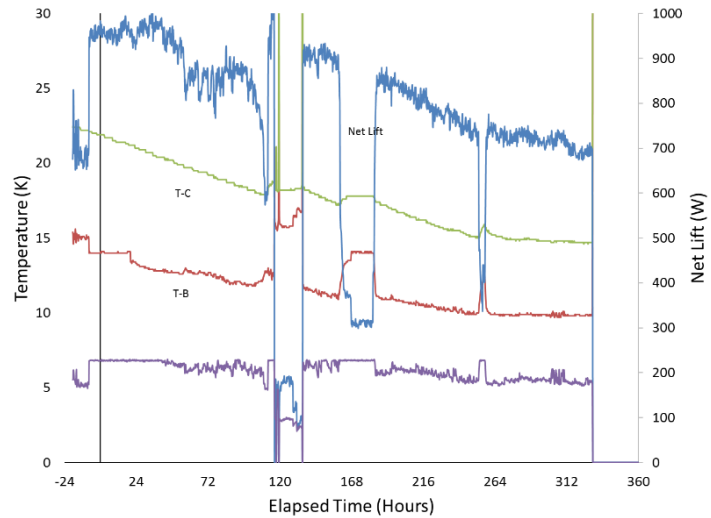


Figure 4 Refrigerator Performance, 33% Densification Test

B. 67% Liquid Level

The 67% densification test was conducted between June 30 and July 25, 2016. The system was in a ZBO test at the time, with a fixed helium supply temperature of 18 K. The tank pressure was slowly trending up around 17.1 psia, and the center-rake temperatures were close to isothermal between 20.6 K and 21.9 K. The refrigerator was placed in full power mode with LN₂ precooling at 1015 on June 29. The tank depressurized at a fairly constant rate of -0.10 psi/hr, becoming subatmospheric at 1045 on June 30.

The period of time the tank was subatmospheric for the 67% fill densification test is shown in Figure 5. The refrigerator was operating on full power with LN₂ precooling for the beginning of the test, and LN₂ precooling was turned off at T+394 hours. The response of the tank pressure (PT2) shows that the rapid decrease in the pressure gradually slows down to become isobaric near the TP. The tank pressure stabilizes at 1.12 ± 0.03 psia, stays there for approximately 60 hours, but again never reaches the defined LH₂ TP pressure of 1.02 psia, possibly due to reasons discussed above. The system was run for another 100+ hours without LN₂ precooling to try to determine if the TP conditions could be maintained without the use of LN₂, but the heat leak at those temperatures overcame the refrigerator capacity and the tank gradually started to warm up. At T+500 hours, a GH₂ supply was introduced into the tank. This allowed the tank to increase back to atmospheric pressure more quickly so follow-on testing could continue.

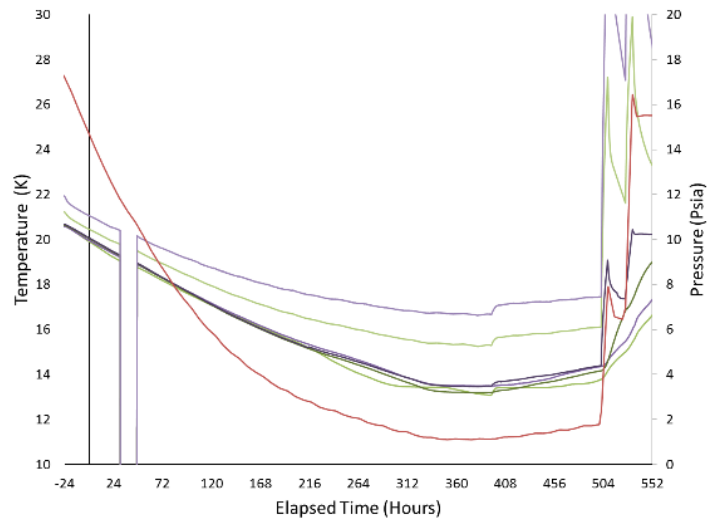


Figure 5 Tank Pressure and Center-Rake Temperatures, 67% Densification Test

Looking in more detail at the entire set of tank temperatures in Figure 6, we see evidence that slush hydrogen was produced during this test, as well as the previous one. The lowest set of diodes all reach steady temperatures of between 13.3 K and 13.6 K at around T+310 hours. It takes another 36 hours for the next set of diodes to reach these steady temperatures. These diodes, clustered in a group between 13.2 K and 13.5 K, are all located in the liquid region, but above the inlet header of the CHX. The liquid above the bottom of the CHX will eventually reach the same temperatures as the liquid below the CHX, demonstrating the mixing ability of the liquid and the distributed cooling performance of the CHX geometry. After the entire set of liquid temperatures reach the TP, there is a similar temperature decrease for certain diodes below the TP that was found during the 33% fill test. Starting with TT3 at T+350 hours, TT5 (T+357 hours) and TT1 (T+367 hours) all exhibit temperature decreases below the TP. Interestingly, this was a different sequence of supercooling initiation compared to the 33% test, when the TT5 sensor showed this effect first. All these sensors are along the centerline of the tank, near the inlet header. During this test, there was no further cooling beyond the TP for sensors located either radially outward on the tank (TT2, TT6) or above the CHX header (TT4) as was found in the earlier test. Once the LN₂ precooling was turned off, the supercooling effect could not be maintained and these diodes quickly reached their previous TP temperature. The liquid above the CHX inlet header gradually began to warm back up as a group, but the lower level of liquid maintained nearly constant TP temperatures for another 48 hours before starting to increase. At T+500 hours, with the tank pressure at 1.8 psia, GH₂ flow was initiated into the tank to warm up the contents. Two batches of gas were introduced over the next 36 hours, and the tank reached atmospheric pressure around 1400 on July 22, 2016.

The refrigerator performance for the 67% densification test is shown in Figure 7. The performance was above pretest predictions during the initial part of the test, with cooling of 960 W at a return temperature of 20 K. This performance level gradually decreased as the temperatures fell, with a final performance of roughly 760 W at 15.5 K when the LN₂ cooling was stopped. The engine speed was 227 RPM during the entire test, including after the LN₂ supply was turned off. The refrigerator continued to produce between 280 W and 300 W of cooling, without LN₂ precooling, as the return temperature increased from 15.9 K to 16.7 K.

C. 100% Liquid Level

The 100% liquid level densification test occurred between September 12 and October 5, 2016. The system was in a ZBO temperature control mode test before the refrigerator was turned on to full power at 0830 on September 12. The IRAS tank was already at the saturation state and quickly started depressurizing at a rate of -0.05 psi/hr. The

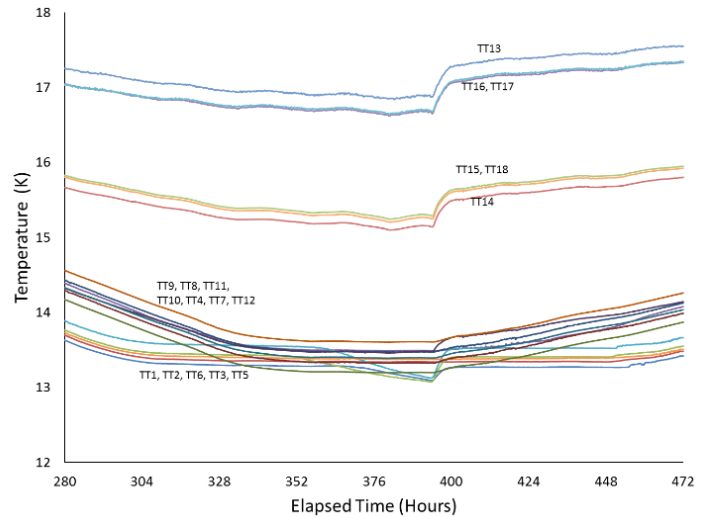


Figure 6 Detailed Tank Temperature, 67% Densification Test

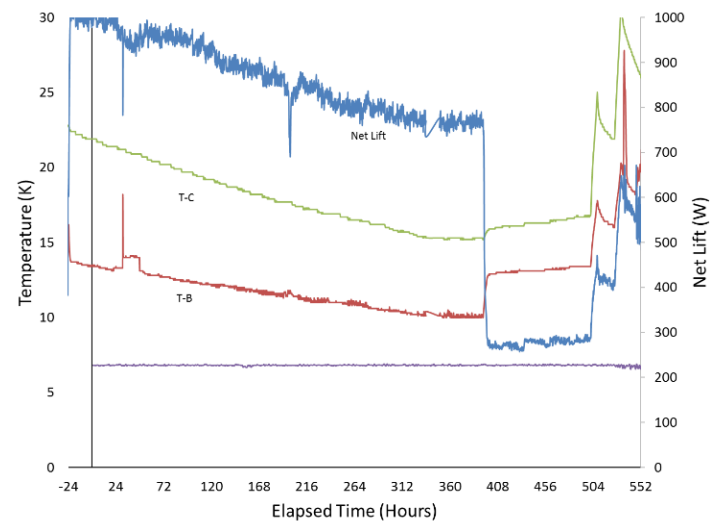


Figure 7 Refrigerator Performance, 67% Densification Test

system reached the NBP (T-0) at 0730 on September 14, and continued to operate with few problems for the next 15 days. But this was at the end of the project life, and the refrigerator was turned off and active testing was stopped on September 29.

The tank temperatures and pressure during the final densification test are shown in Figure 8. The system was running fairly nominally until LN₂ ran out at T+270 hours. At that point, the tank was at 3.6 psia and the liquid below the inlet header was under 15 K, the original pretest prediction. The refrigerator could not sustain that condition without the LN₂, however, and the test continued for two more days while the pressure and temperature slowly increased. The refrigerator was turned off at 2030 on September 29, and the pressure and temperature increased until reaching the NBP on October 5. At that point, the tank was stratified, and while the pressure was above atmospheric, the bulk liquid temperature remained densified for some time. Other items of note on the charts are a temperature data dropout for 14 hours due to power spikes on the data acquisition system, and unexplained data jumps in TT16 (shown in Fig 8), TT13, and TT17.

Figure 9 shows the refrigerator performance during the 100% full test. The performance was solid exception for a 2-day degradation due to valve problems. After the valve stems were adjusted, the capacity returned to normal. The capacity was above 900 W with a 20 K return temperature, and was still producing over 800 W at a return temperature of 17.7 K when the LN₂ ran dry. Prior boiloff testing estimated the tank heat leak at this liquid level to be 315 W.

D. Summary of Results

The densification timelines for the different liquid levels are compared in Figure 10. As expected, the time required to change the storage state is directly proportional to the mass in the tank. The 33% fill test, corresponding to roughly 4080 kg of LH₂, needed just 107 hours to densify to a saturated pressure of 3.5 psia, whereas the 67% fill test took 181 days to densify to the same conditions, and the 100% fill test, with 8680 kg of LH₂, took 298 days. At the moment in testing when the tank reaches the NBP, the 33% full tank is depressurizing at 0.15 psi/hr, compared to 0.10 psi/hr for the 67% full tank and just 0.05 psi/hr for the 100% full tank. At the 33% and 67% fill levels, the data suggest that significant amounts of slush hydrogen were produced as LH₂ temperatures were observed to stabilize and then drop below the TP temperature of 13.8 K.

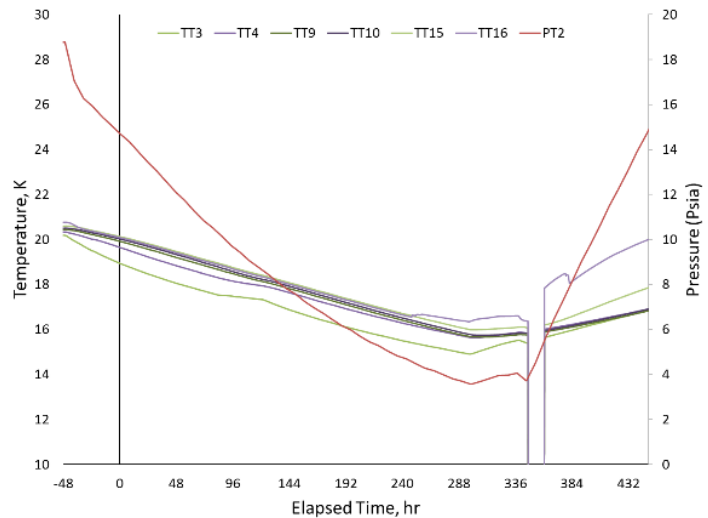


Figure 8 Tank Pressure and Center Rake Temperatures, 100% Densification Test

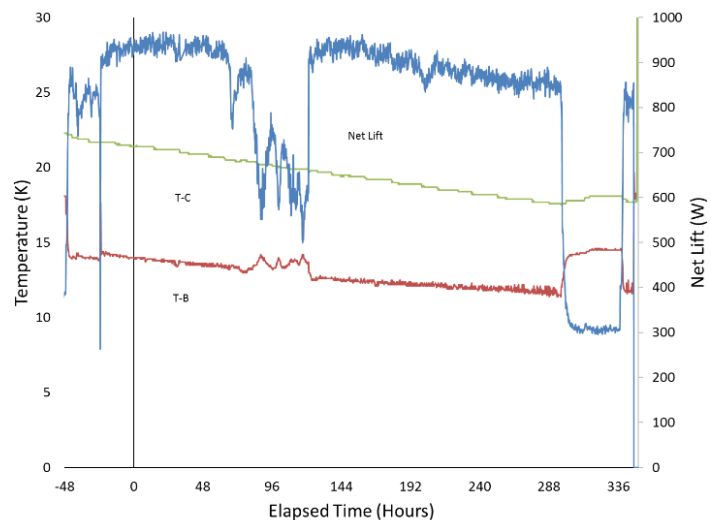


Figure 9 Refrigerator Performance, 100% Densification Test

V. Conclusion

Using the Integrated Refrigeration and Storage (IRAS) technology, NASA cryogenic engineers have demonstrated the capability to produce large batches of densified hydrogen. A Linde LR1480 Brayton cycle refrigerator, nominally capable of producing 880 W of cooling at 20 K while circulating 22 g/s of cold GHe, was integrated into a 125 m³ LH₂ dewar using an SS304 tube heat exchanger distributed in the inner liquid region. The tank was locked up and the refrigerator was run at full power to densify the LH₂ in the tank. Testing was completed at three different liquid levels corresponding to the 33%, 67%, and 100% full levels. All the tests were successful, with the first two liquid levels achieving a hydrogen slush mixture. The 100% full level test was displaying similar behavior toward the triple point, but testing was terminated at the end of the project life.

This new capability is now available to further develop densified-propellant technology. Because the system was designed to be transportable, this new capability can be used at suitable engine test stands to provide densified hydrogen down to slush conditions for qualification testing. NASA is currently looking for interested partners to work in helping develop this technology.

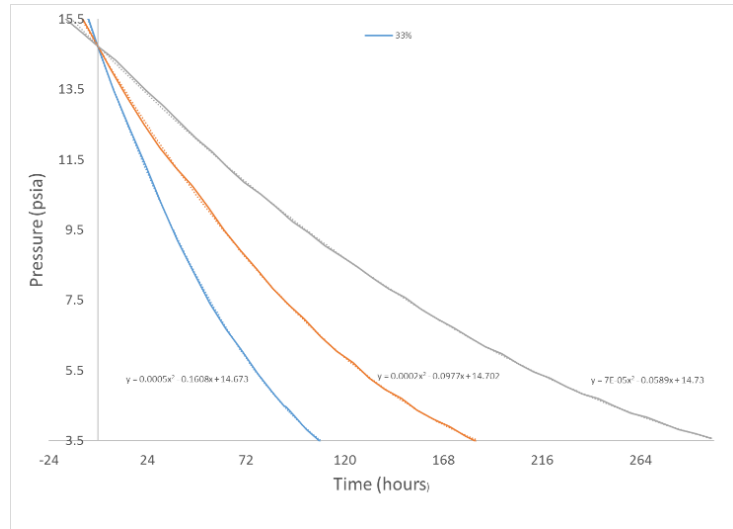


Figure 10 Comparison of Densification Timelines

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