Increasing the Efficiency of He and H_2 Liquefaction Using Small Coolers

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ABSTRACT

People have been re-condensing helium and hydrogen with coolers for many years. There are a number of companies that sell systems that liquefy helium starting with gas at room temperature. These same coolers can be used to re-liquefy hydrogen at room temperature as well. This author has used coolers to cool-down a superconducting magnet and liquefy helium into the magnet cryogenic vessel. The rate of liquefaction was affected by several factors. This paper will talk about ways of making the liquefaction of helium and hydrogen more efficient. Both helium and hydrogen require much more cooling to cool down the gas from room temperature to the liquefaction temperature at a pressure of 0.1013 MPa boiling point temperature than it does to change the gas to a liquid. This means that the precooling of these gases is important.

INTRODUCTION

The liquefaction of air gases didn't occur until the latter part of the 19th century, but the first air gases were not liquefied by von Linde. The exception was carbon dioxide, which was liquefied earlier in the 19th century by pressurizing the gas at room temperature. The first liquefaction air gases was done by Wroblewski and Olszweski in 1883 [1] by an isenthalpic expansion of high pressure air gasses through an orifice which today we would call a J-T valve. Olszweski also tried to liquefy hydrogen in 1895, but the liquid, if at all, was only transitory. He didn't have a vessel that could hold the liquid. He measured temperatures down to 48 K. The first successful liquefaction of hydrogen in a vessel was carried out James Dewar in 1896 using a process where high pressure hydrogen was cooled by liquid nitrogen at 77 K followed by a J-T valve expansion [2]. Dewar had a vacuum insulated vessel that would hold the liquid hydrogen. Helium was more difficult to liquefy. It wasn't liquefied in1908 by Kamerlingh Onnes using a double cascade process that involved both liquid nitrogen and liquid hydrogen [3]. As a result, superconductivity was discovered by Kamerlingh Onnes in 1911 [4]. Hydrogen and helium refrigeration was typically done using the cascade process with high pressure gases almost until the first Collins cryostat appeared in 1947 [5]. Commercial hydrogen-helium liquefiers became available by 1956 [6]. This opened up low temperature refrigeration to universities and laboratories worldwide.

While MIT was developing refrigeration in the 20 to 35 W range at 4.5 K and helium liquefaction up to 10 L per hour, Syracuse University was developing much smaller cryogenic coolers that would go down into the cryogenic temperature range from 50 to 80 K. The first of these coolers were GM (Gifford McMahon) coolers [7]. Pulse tube coolers came along five years later [8]. The first two stage GM cooler was demonstrated at Syracuse in 1966 [9]. The second stage temperature could be as low as 15 K, which meant that liquid helium could be produced in a separate circuit with flow from a separate compressor,

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regenerative heat exchangers on either side of the two stages, and a final J-T valve to produce liquid helium [10]. GM machines were sold by the thousands for both military and civilian applications. A GM machine could be operated in any orientation, which made it attractive for use on moving platforms. The machines of the 1970s and 1980s did have maintenance issues, but regular maintenance solved the problem. Far fewer machines that could run at 4 K were sold. The purity of the helium in the separate circuit was often a problem. Pulse tube coolers were not widely used in the US. This author saw his first pulse tube cooler in Xian China in 1990. The major thing that pushed the development of coolers was the development of high heat capacity regenerator materials that permitted two-stage cooler cold heads to go below 4 K [11]. Coolers using these materials became available by the late 1990s. Had these coolers been available in 1994, this author may have convinced the paper industry to use low field superconducting NMR magnets for process control of pulping facilities [12].

LOOP COOLING, WHICH IS WHY HE AND H2 LIQUEFACTION MAY BE NEEDED

This author started working with two-stage 4 K GM coolers in the late 1990s on the Advance Slight Source bending magnets that were about 70-meters apart in the electron storage Ring. We felt that coolers were a better choice than a central refrigerator [13]. By this time cooler were being used as re-condensers on MRI magnets. MRI magnets still had large helium tanks and gas cooled shields. This made maintenance easier because the cooler maintenance could be delayed to suitable down time. We did tests of the system with HTS electrical leads that could continuously deliver 300 A to the magnet coil [14]. The cooler second stage cold head was connected directly to the cold mass as shown in Figure1. In addition to the direct connection with the coil mass, a condenser is shown immersed in liquid helium, which does not work if used alone.

The condenser must be in helium or hydrogen gas, and the object being cooled must be in contact with liquid helium or liquid hydrogen [15]. This would apply to other gases such as neon or nitrogen as well [16]. The condenser and the load being cooled can be separated by meters if a cooling loop is used with the condenser above the object in the liquid [16], which is similar to the system described in [15]. The systems described in [15], [16] have much lower temperature drops than a copper strap between the cold

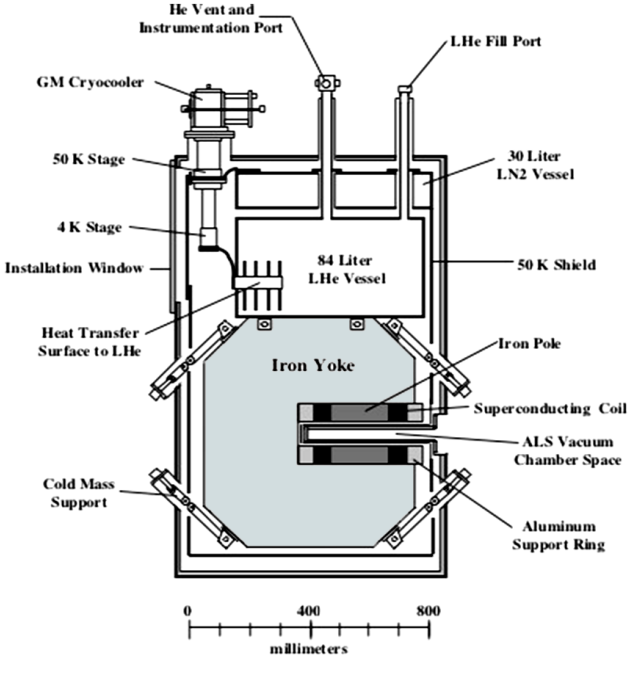

Figure 1. A Cross-section of the LBL light source superconducting dipole magnet

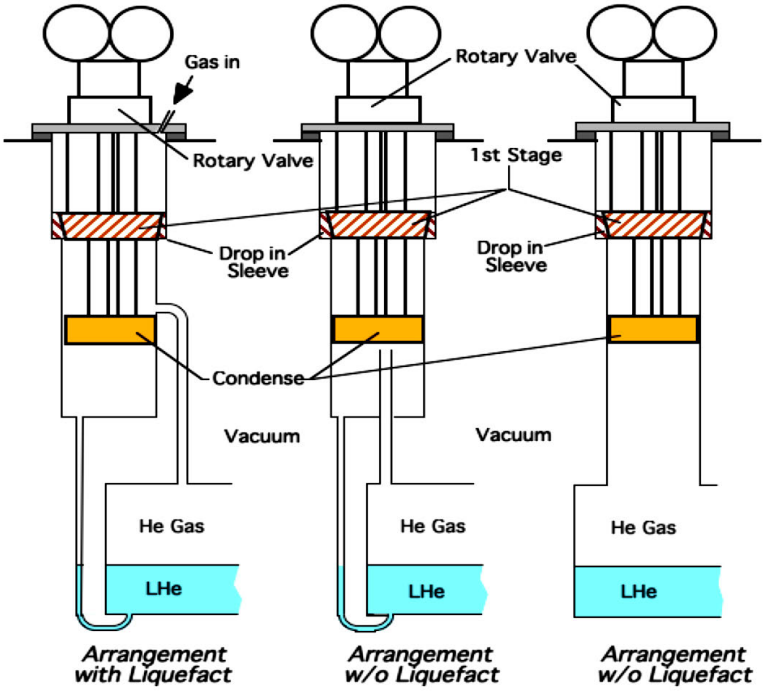

Figure 2. Three configurations of drop-in coolers tested by LBL in 2007 that form cooling loops. The configuration on the left is the only one that permits one to liquefy helium. The two configurations on the tight are only good for re-condensation of helium. The ΔT between the helium and the cold head was less than 0.1 K for helium in a cryostat [18].

head and load. A cooling loop is a good way to get low temperature drops between the cooler cold head and the device being cooled for vertical distances of up to five meters. This allows the cooler to be moved to a lower magnetic field [17] or a lower radiation zone. Figure 2 shows three configurations of drop-in coolers tested by LBL [18]. As long at the heat load into the cryostat is less than the refrigeration on the two cooler stages, it should be possible to liquefy helium or hydrogen, but the cold gas from the first stage must enter above the second stage. Ideally, there should be many small holes in the second stage copper plate, providing a large area for gas condensation. If the cryostat total liquid volume is small, using the coolers to do the liquefaction of the cryogen is a reasonable thing to do. The configuration on the left of Fig. 2 is the best of the three for cooling down a device and the only one that can liquefy warm gas.

The MICE project used many small coolers for the magnets, for the hydrogen absorbers, and for the detector modules inside of the detector solenoids. The British wanted the groups that provided the magnet modules to provide the cooling for them. The British had to provide cooling from a central refrigerator to a pion decay solenoid. With some extra cryogenic plumbing, a standard large refrigerator could have cooled all of the magnets in the experimental channel. Coolers would have been needed only for hydrogen liquefaction in the focusing modules and in the two detectors in the detector solenoids. This author thinks that if there is a lot of cooling needed within a short distance, it is better to use a central refrigerator. However, the problems that occurred on MICE were valuable lessons to the people who had to make the coolers work cooling magnets, including the author. The MICE problems led to success on other projects.

LIQUIFACTION WITH COOLERS AND THE FACTORS THAT MAKE IT WORK

The liquefaction of He or H2 is limited by the removal of the sensible heat that must be removed from the gas before the vapor can be turned to liquid. A typical two stage cooler that produces about 1.5 W of cooling at 4.2 K will produce ~20 W at 20 K. The heat of vaporization of helium at 4.2 K is 20.7 J g^{-1}. The heat of vaporization of para H2 at 20.5 K is about 445 J g^{-1}. If the sensible heat didn't have to be removed

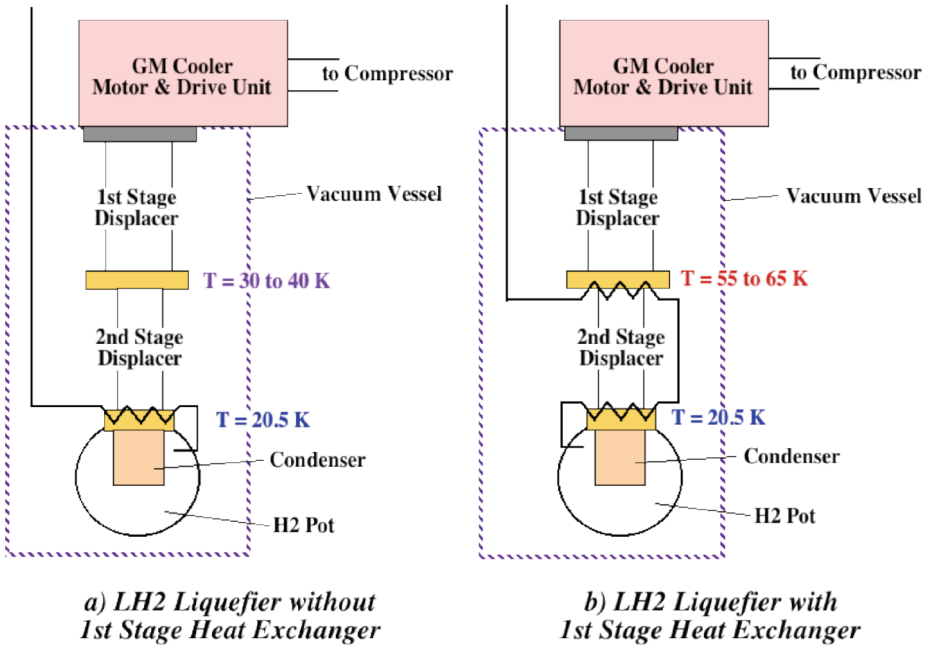

Figure 3. The first two cases of a GM cooler being used as a hydrogen liquefier. The same diagram would apply for a helium liquefier except the second stage temperature would be in the 4 to4.5 K range [18].

Table 1. The ideal liquefaction for Helium and Hydrogen for the three cases describe below.

CASE Number	He Liquefaction (g s^{-1})	H_2 Liquefaction (g s^{-1})
1	~0.001	~0.0047
2	~0.008	~0.027
3	~0.015	~0.035

first, a 1.5 W cooler could liquefy just over 2 L hr^{-1} of each gas. From 4.2 K to 300 K, the specific heat of helium is ~5.2 J g^{-1} K^{-1} and for hydrogen the specific heat is ~14.5 J g^{-1} K^{-1}. From 300 K to 80 K, one can remove the sensible heat from He and H2 and converting ortho H2 to para H2 (~300 J g^{-1}) at 300 K by cooling with liquid N2.

Figure 3(a) shows a GM cooler cooling of the gas only on the second stage cold head, whereas Fig. 3(b) has the pre-cooling of the gas occuring on both the first stage and the second stage cold head. A third case, not shown in Fig. 3, is when the gas is cooled to 80 K using liquid nitrogen before the gas is piped to the first and second stages for further pre-cooling before liquefaction on the second stage cold head. Table 1 shows an estimate of the best-case liquefaction for the three cases for both helium and hydrogen. To convert liquefaction to L hr^{-1} for helium multiply the value above by 28.8, and for hydrogen multiply the value above by 51.2. Much of the ortho to para transition can occur at 80 to100 K with the proper catalyst in a tube. It is clear that liquefying hydrogen gas with a cooler is easier than liquefying helium gas.

Figure 4 shows an experiment that was developed over ten years ago at Cryomech using a PT405 0.5 W pulse tube cooler. The purpose of the experiment was to see if additional refrigeration is available from the cooler at a point between the first and second stages. The result was 1.25 W of refrigeration at 8 K with a reduction at 4.2 K of 0.12 W. This suggests that one could extract heat to the cooler between the two stages to cool-down gas to be liquefied at the second-stage cold head. Figure 5 shows a Cryomech PT410 pulse tube cooler that was modified to do this.

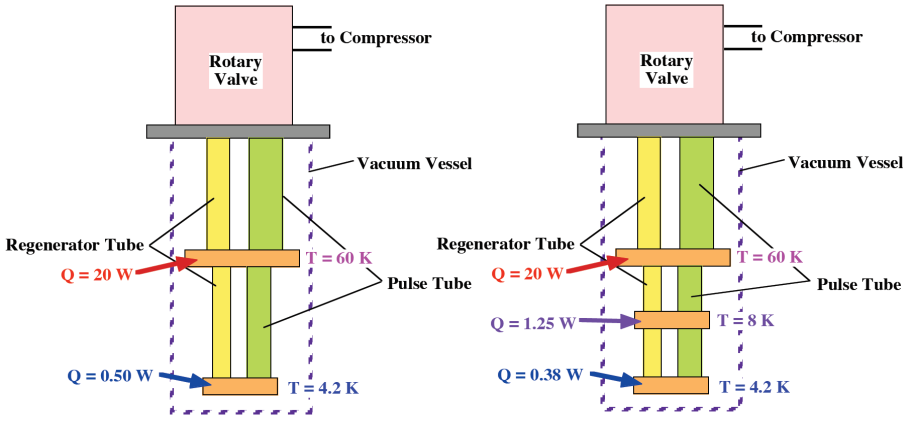

Figure 4. A demonstration that additional heat can be pulled from a pulse tube cooler

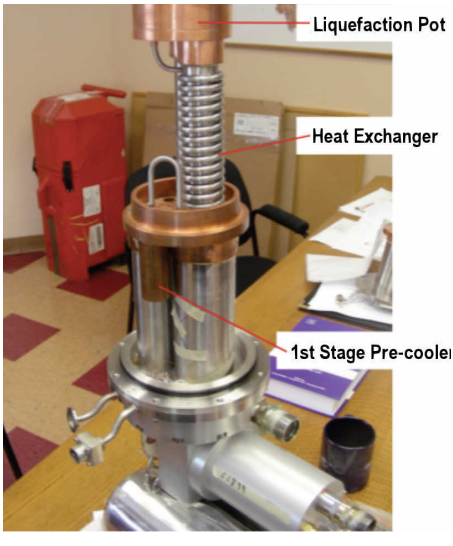

Figure 5. Modification of a PT410 cooler to make helium liquefaction more efficient. A helium condensing pot is connected to the second stage cold head. There is a tube carrying helium around the second stage tubes from the first stage to the second stage. There is no heat exchanger around the tubes above the first stage. The helium gas enters the system from the top where it enters the 1st stage pre-cooler connected to the first stage. It is not clear that there is any heat exchange between the first stage tube and the gas, but there might be. The helium liquefied on the condenser drips out of the bottom of the liquefaction pot.

The pulse tube cooler shown in Fig. 5 liquefied 0.022 g s^{-1} using a PT410 cooler which produces 1.0 W at 4.2 K on the second-stage cold head. The system refrigeration to liquefaction coefficient is ~45 Jg^{-1} [22], which is better than many larger helium refrigerator-liquefiers including ones with liquid nitrogen pre-cooling. A 2800 Cti refrigerator that produces 250 W with liquid nitrogen pre-cooling of cooling has a liquefaction coefficient of 85 J g^{-1}.

The liquefier in Fig. 5 has been used at the South Pole to re-condense helium in a 4000 L storage dewar that previously had a boil-off of 14 L per day. The liquefier uses laminar flow heat exchangers with small hydraulic diameters. The down side of this system is that it is easily clogged with impurities. As a re-liquefier, the cooler could put 3 L per day of helium into the tank. While the liquefier shown in Fig. 5 looks like a simple thing to do, it was difficult to fabricate. The key is increasing the U factor in laminar flow in a way that is similar to the technique used in gas-cooled electrical leads for helium-cooled superconducting magnets [23]. With modern 3D printer fabrication, one might be able to fabricate efficient heat exchangers that have a higher U factor and more area.

LIQUIFACTION LESSONS LEARNED FROM THE MSU GAS STOPPER MAGNET

The Michigan State Cyclotron Gas-stopper Magnet consists of two superconducting coils in separate cryostats that operate at 4.5 K. The total mass of the magnets, their cryostats, and the room temperature iron are about 172 tons [24]. Each of the two coil cryostats has a cold mass of 1240 kg. The magnet is built in two parts with a coil cryostat around each iron pole. The two poles and return yokes associated with them come apart for maintenance of the cyclotron. They guide and slow-down the beam of exotic heavy ions from energies of ~50 MeV to energies of a few eV so they can be accelerated to higher energies by the FRIB. The reduced energy beam emittance is much lower than the beam that entered the gas-stopper cyclotron magnet from the target that produced the ions. The magnet is an iron dominated sector cyclotron magnet with a peak induction of about 2.7 T.

The magnet forces are supported by the iron pole to which the coil cryostat is mounted. The magnetic forces will push the coil cryostat into the iron [25]. Each magnet coil includes 1767 turns of a wire composed of niobium titanium in a copper matrix that fits into a coil package that is 80 by 80 mm within the stainless-steel cryostat. The assembled magnet self-inductance is 178 H. With 200 A in each coil, the stored energy of the magnet is 3.56 MJ. The peak coil current density is 54.9 A mm^{-2}, so the magnet is essentially self-protecting [26, 27].

Each coil cold mass is cooled-down using a free convection cooling loop where the cold helium enters the magnet cryostat at the bottom [28]. The helium in each cryostat is cooled using three Cryomech PT-415-RM pulse tube coolers that can provide 4.5 W of cooling at 4.2 K. The coolers are located at the top of the five-meter high coil system (including the iron). The cooling loop is driven by the difference in densities of the gas coming from the cooler cold head condenser and the gas being warmed by heat from the coil and cryostat. Warm gas from the top of the coil cryostat enters the tops of the condensers to be cooled back down by the cooler cold heads before going down to the bottom of the magnet cryostat.

Figure 6 shows a simple cooling loop that runs like the cyclotron gas stopper coil cooling loops. Cooling loops can be either open or closed. Closed loops must be designed for higher operating pressures,

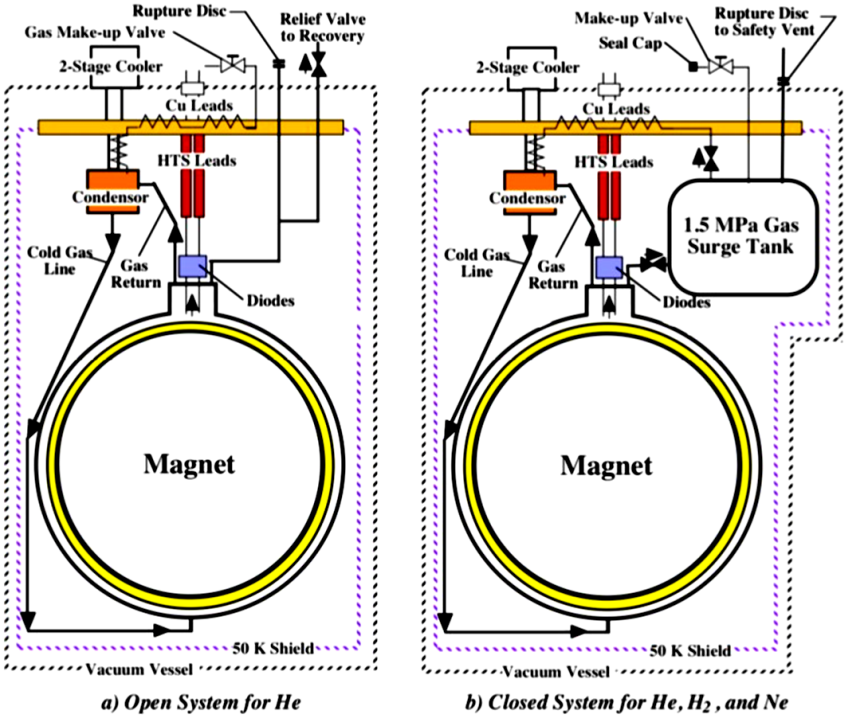

Figure 6. Open and closed two-phase free-convection cooling loops that can be used for cooling down and liquefying the gas into the cryogenic vessel. The closed two-phase loop is best for H2 and Ne for safety and cost reasons. The closed loop must be designed for higher pressures than the open loop system.

which is desirable for reducing the magnet cooldown time. This means that all components (including bellows) in the system must withstand the highest pressure (perhaps high enough to involve the pressure vessel code). No liquid nitrogen system is shown in Fig. 6. The MSU cyclotron gas stopper magnet has a liquid nitrogen shield. The shield on the gas-stopper magnet had to be liquid nitrogen cooled because the temperature difference in the magnet shield would have been too large. Orientation of a pulse tube cooler must be with the cold head down, so it is best that it be located at the highest point on the magnet.

We found that small things had an effect on the time to fill the tank with helium. With three coolers, one had to have the same flow resistance in the circuits that connected the coolers to the manifold. The Cooler with larger flow resistance didn't provide as much cooling as the other two coolers. This extended the cooldown time and the liquefaction times. Small thing such as where the external helium entered the circuit made a big difference in the cryostat fill time. If one has more than one cooler, it is best if all coolers are involved in the liquefaction process. For a given circuit pressure hydrogen is the best fluid for cooling down the cold mass using free convection cooling [29]. Helium is slightly worse (~7 percent longer) than hydrogen in terms of cooldown times. The cooldown times with neon are 55 percent longer than with helium.

Figure 6 shows no pre-cooling at liquid nitrogen temperature. Since the MSU magnet had a liquid nitrogen cooled shield, that shield was used to pre-cool the helium being liquefied. It was also used as an intercept for heat down the low RRR copper conduction cooled leads that were between 300 K and the tops of the HTS leads and the cooler first stages. Thus, the cooler first stage temperature was lower. After the MSU magnet cryostat vessel was filled with helium, one of the coolers was turned off. The magnet coils were kept at 4.5 K by two coolers. The heat leak from the cooler not operating was taken up by the first stages of the other two coolers.

CONCLUDING COMMENTS

GM coolers and pulse tube coolers were introduced in 1959 and 1964, respectively. The first two-stage GM cooler that could reach 15 to 20 K made helium and hydrogen liquefaction achievable in 1966. The first cooler that produced helium refrigeration was run in 1969. Between 1970 and the 1990s there was limited production of machines based on a two-stage GM cooler with an added compressor, regenerative heat exchangers and a J-T valve.

The key to low temperature cooler development was the discovery of rare earth high heat capacity regenerator materials. This allowed companies to produce reliable two-stage 4 K coolers of both the GM and the pulse tube variety. The high heat capacity regenerators made it possible for GM and pulse-tube coolers to be used as a helium liquefier. Liquefiers made with both types of coolers are being sold commercially. The key to doing this efficiently is the technique used to remove the sensible heat from the gases being liquefied. Liquefaction with small coolers is a laminar flow process. This is key to increasing both the U factor and the heat exchanger area A. In laminar flow, one must reduce the hydraulic diameter of the flow channel in order to get a high heat exchanger U factor. In order to do this economically one must explore different ways of manufacturing the heat exchangers and connecting to the appropriate cooler parts.

Observed failures are often from the little things that one does when integrating the liquefier with the object being cooled. In cases where cool-downs and filling the cryostat with cryogens with a cooler liquefier are done infrequently, one should consider cooling down the gas entering the cryostat with liquid nitrogen. Purity of the gas going into the system is very important. All gas should be run through a purifier before it enters the system where liquefaction occurs. Simple things like using the wrong type tube to transfer the gas to whatever is being cooled-down can cause a lot of problems. If one is cooling-down a device that is cooled down with a free-convection cooling loop, one must make sure that all parts that are exposed to high pressure must be able to safely handle that pressure. The applicable pressure vessel codes should be followed especially with hydrogen, which comes under the flammable gas pressure vessel code.

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