RE-CONDENSATION AND LIQUEFACTION OF HELIUM AND HYDROGEN USING COOLERS

M. A. Green

Lawrence Berkeley National Laboratory
Berkeley CA 94720, USA

ABSTRACT

Coolers are used to cool cryogen free devices at temperatures from 5 to 30 K. Cryogen free cooling involves a temperature drop within the device being cooled and between the device and the cooler cold heads. Liquid cooling with a liquid cryogen distributed over the surface of a device combined with re-condensation can result in a much lower temperature drop between the cooler and the device being cooled. The next logical step beyond simple re-condensation is using a cooler to liquefy the liquid cryogen in the device. A number of tests of helium liquefaction and re-condensation of helium have been run using a pulse tube cooler in the drop-in mode. This report discusses the parameter space over which re-condensation and liquefaction for helium and hydrogen can occur.

KEYWORDS: Coolers, Re-condensation, and Liquefaction

INTRODUCTION

The magnets for the Muon Ionization Cooling Experiment (MICE) [1] cooling channel are in seven superconducting solenoid modules that will be kept cold using sixteen Cryomech PT415 pulse tube coolers that generate 1.5 W of cooling at 4.2 K on the second stage while generating 50 W at 55 K on the first stage. The coolers must be located so that the magnet induction at the rotating valve motors and HTS leads is less than 0.3 T [2]. As a result, the coolers must be located some distance (~0.3 to 0.5 meters) from the magnet module being cooled. The coolers will be connected to the magnet cold mass through a two-phase helium thermal siphon cooling system [3], where the liquid helium is around the magnet cold mass. Thus the re-condensation of helium is of great importance.

The three focusing modules each contain a liquid hydrogen absorber [4] that is cooled using a single PT415 cooler that generates about 20 W of cooling at 20 K. The absorber cooler will be connected to the absorber through a hydrogen thermal siphon circuit [5]. It is hoped that the cooler will liquefy hydrogen into the absorber. For the MICE liquid hydrogen absorbers, both re-condensation and liquefaction of hydrogen is of interest.

RE-CONDENSATION OF HELIUM AND HYDROGEN USING A COOLER

The key to connecting the cooler cold head with the load immersed in a cryogenic fluid is creating a condenser circuit that causes the cryogen that cools the load to circulate. Simply immersing the cold head of the cooler into a bath of liquid cryogen does not work if one wants a low temperature drop between the source of the heat and the cold head. A re-condensing gravity thermal siphon circuit shown in FIGURE 1 will have a much lower ΔT between the magnet and the cooler cold head than any other method of connecting the cold head to the load [3]. Excellent vibration isolation can be achieved because the distance from the load to the cold head can be increased and the elements connecting the two can be flexible within the pressure constraints of the cooling circuit.
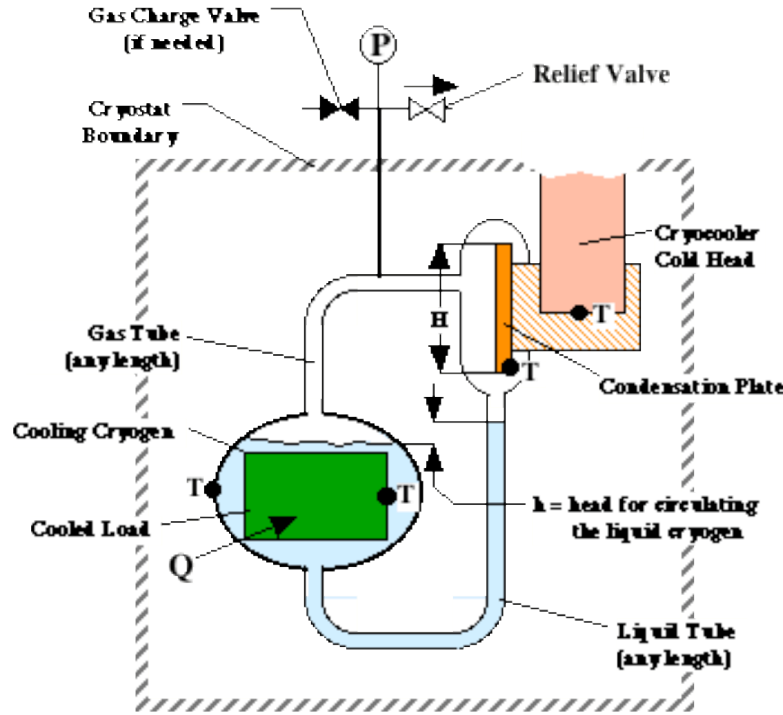

FIGURE 1. A schematic representation of a gravity loop thermal siphon system to transfer heat from the load (shown in the liquid cryogen) to the cooler cold head (attached to the condenser plate).

The temperature drop between the load being cooled (the surface magnet cold mass) and the condenser that is connected to the cold head is the temperature drop at the condenser ΔT_c added to the boiling temperature drop at the cooling surface ΔT_b

The temperature drop across the condenser ΔT_c can be estimated using the following expression [6];

$$\Delta T_c = \left[\left(\frac{Q}{A} \right)_c \left(\frac{\mu_l H}{\rho_l^2 g h_{fg} k_l^3} \right)^{0.25} \right]^{\frac{4}{3}}. \tag{1}$$

$(Q/A)_c$ is the heat flux on the condenser surface; μ_l is the viscosity of the liquid phase; H is the vertical height of the condensation surface; ρ_l is the density of the liquid; g is the acceleration of gravity; h_{fg} is the heat of vaporization of the fluid; and k_l is the thermal conductivity of the liquid phase. TABLE 1 shows the parameters for helium and hydrogen.

TABLE 1. Various Parameters of Two-phase Helium and Hydrogen as a Function of Temperature [7], [8]

Fluid Parameter	Cryogenic Fluid			
	Helium		Hydrogen	
Triple Point Temperature (K)	2.17		13.8*	
Triple Point Pressure (MPa)	0.0052		0.0071*	
1 atm Boiling Temperature T_b (K)	4.22		20.4	
Liquid Density at T_b (kg m^{-3})	124.9		70.8	
Gas Density at T_b (kg m^{-3})	16.84		1.39	
Critical Temperature (K)	5.19		33.0*	
Critical Pressure (MPa)	0.224		1.29*	
Critical Density (kg m^{-3})	69.6		30.3*	
Heat of Vaporization at T_b (J kg^{-1})	20700		441500*	
Ortho to Para Transition at T_b (J kg^{-1})	------		531500	
Gas Specific Heat T > 20 K (J kg^{-1} K^{-1})	5200		~14200*	
Enthalpy Triple point to 300 K (J kg^{-1})	~1578000		~5040000**	
Temperature (K)	3.8	4.6	16	24
Liquid Density (kg m^{-3})	132.0	116.3	72.8	68.3
Heat of Vaporization at T_b (J kg^{-1})	22480	17930	449500*	417200*
Gas Density (kg m^{-3})	11.1	24.5	0.35	2.84
σ Surface Tension (N m^{-1})	1.35×10^{-4}	0.54×10^{-4}	2.65×10^{-4}	1.30×10^{-4}
Thermal Cond. Liquid at T_b (W m^{-1} K^{-1})	0.0183	0.0188	0.109	0.127
Thermal Cond. Gas at T_b (W m^{-1} K^{-1})	0.0078	0.0105	0.0123	0.0188
Viscosity Liquid at T_b (kg m^{-1} s^{-1})	3.63×10^{-6}	3.57×10^{-6}	1.97×10^{-5}	1.25×10^{-5}
Viscosity Gas at T_b (kg m^{-1} s^{-1})	1.08×10^{-6}	1.42×10^{-6}	0.86×10^{-6}	1.28×10^{-6}

* These values apply for para hydrogen.

** This includes the conversion heat from ortho hydrogen to para hydrogen.

It is clear that ΔT_c is a function of the $(Q/A)_c$ on the condensing surface. Extending the surface will reduce ΔT_c provided the surfaces are vertical so the condensed liquid can flow off of them. FIGURE 2 shows a condenser used for the LBNL cooling experiment.
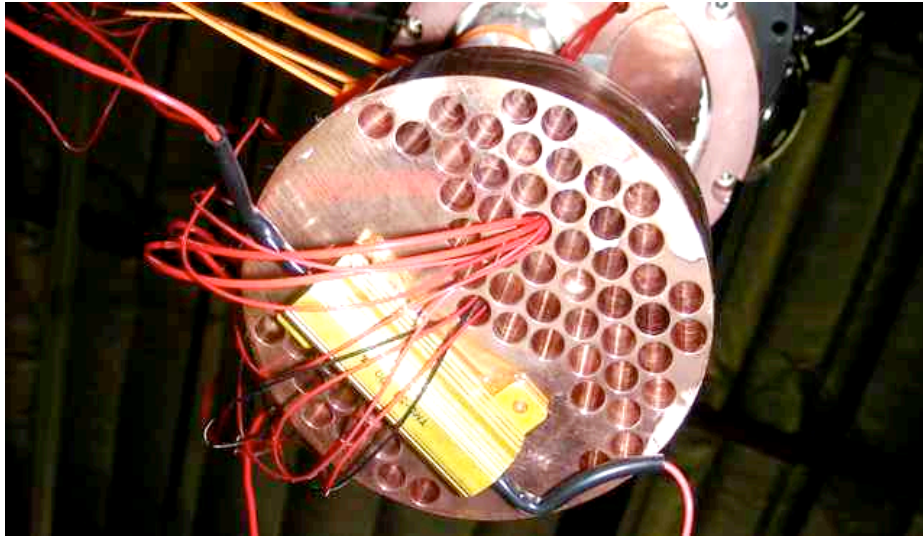

FIGURE 2. The Condensing Surface built into the Second Stage of a Cryomech PT415 Pulse Tube Cooler. (The cooler is used for the cooler experiment at LBNL and for the two MICE spectrometer magnet coolers.)

The peak nucleate boiling heat flux per unit area $[Q/A]_{np}$ and the peak temperature difference between the hot surface and the bath ΔT_{np} during nucleate boiling can be estimated using the following expressions [9];

$$\left[\frac{Q}{A}\right]_{np} = 0.16 h_{fg} \rho_v^{0.5} \left[\sigma g(\rho_l - \rho_v)\right]^{0.25} \tag{2a}$$

$$\Delta T_{np} = \Xi \left[1 - \frac{T_s}{T_c}\right]^m T_c \tag{2b}$$

where ρ_v is the density of the saturated vapor; σ is the surface tension; T_s is the saturation temperature of the fluid; and T_c is the critical temperature of the fluid. Ξ and m are constants that come from Van der Waal's equation. For diatomic fluids like hydrogen, nitrogen, or oxygen Ξ is about 0.5 and m is about 1.5. For helium, a monatomic gas, Ξ and m are about 1.0. Equations 2a and 2b are valid over a range of $0.56 < T_s/T_c < 1$. One may also use the measured values of $(Q/A)_{np}$ and ΔT_{np} [10]. Using nucleate boiling equations, for $Q/A < (Q/A)_{np}$, Q/A is proportional to $\Delta T^{2.5}$ [10].

The boiling temperature drop ΔT_b for the boiling side can be expressed as follows;

$$\Delta T_b = \left[\frac{\left(Q/A\right)_b}{\left(Q/A\right)_{np}}\right]^{0.4} \Delta T_{np} \tag{3}$$

From Equation 3, it is clear that the ΔT_b on the boiling surface is a function of $(Q/A)_b$. If wants to reduce the value of ΔT_b, one must increase the boiling area. As with ΔT_c, the value of ΔT_b can be set by controlling the heat transfer per unit area of the boiling surface.

TABLE 2 shows the heat transfer characteristics of the MICE coupling magnet [11] that is cooled using helium in tubes and the MICE liquid hydrogen absorber [4]. The condensing area given in the table is for the condenser that is shown in FIGURE 2. The increased temperature means that more heat is passing across the condenser surface and the boiling surface attached to the cold mass. Increasing the temperature of the load doesn't have a big effect on the helium system temperature drop. For the hydrogen, system there is a decrease in the temperature drop as the 2nd stage temperature increases.

TABLE 2. Heat Flow per Unit Area, Second Stage Refrigeration (PT-415 Cooler), and Temperature Drop between the Heat Load and the Cooler Second Stage as a Function of Temperature.

Fluid Parameter	Cryogenic Fluid			
	Helium		Hydrogen	
Liquid Surface Area at Source (m²)	1.18		0.47	
Condenser Vertical Area (m²)	0.042		0.042	
1 atm Boiling Temperature T_b (K)	4.22		20.4	
2nd Stage Refrigeration at T_b (W)	1.5		~20	
1st Stage Refrigeration (W)	50		15	
1st Stage Temperature (K)	53		36	
2nd Stage Temperature (K)	3.8	4.6	16	24
2nd Stage Refrigeration (W)	~1.1	~2.0	~15	~24
Liquid Surface Heat Flux $(Q/A)_b$ (W m⁻²)	~0.93	~1.69	~31.9	~51.1
Condenser Heat Flux $(Q/A)_c$ (W m⁻²)	~26.2	~47.6	~360	~550
Liquid Surface Temperature Drop (K)	~0.039	~0.014	~0.39	~0.12
Condenser Temperature Drop (K)	~0.015	~0.039	~0.08	~0.11
Total Temperature Drop (K)	~0.054	~0.053	~0.47	~0.23

THE LBNL RE-CONDENSATION AND LIQUFACTION EXPERIMENT

LBNL tested four PT415 pulse tube coolers in the drop-in mode [12]. These coolers had a press-fit between the cooler first stage and a copper sleeve that carries heat from the shield and leads. The heat into the cooler 1st stage must pass across this joint. The LBNL experiment was operated in three configurations. The first configuration has condensed LHe taken to the bottom the liquid tank; the boil off gas enters the condensing chamber above the condenser. The second configuration is similar to the first, except the boil-off gas enters just below the condenser. The third configuration has the condenser sitting in an open 100 mm diameter tube. LHe drips off the condenser into the pot; the boil-off gas goes up along the tube wall to the condenser. The three cases are shown in FIGURE 3.

All three configurations shown in FIGURE 3 exhibited helium re-condensation down to temperature of 3.5 K or so. The upper-limit temperature for re-condensation was always limited by the relief valve on the helium tank. The test was done with about 50 W going into the sleeve around the cooler first stage. The ΔT from the sleeve to the cooler first stage was from 0.5 to 3.0 K depending on whether high conductivity vacuum grease was used between the sleeve and the first stage. Arrangement A was able to liquefy helium gas at 300 K despite the lack of a heat exchanger on the first stage. Liquefaction was not tried with arrangements B and C. Arrangement A was able to cool down the experiment before liquefaction started. Both arrangements A and B have a fatal flaw. The pipe entering the bottom of the tank should not look like a sink trap. The trap can collect water, air, and dirt causing the flow from the condenser to stop.
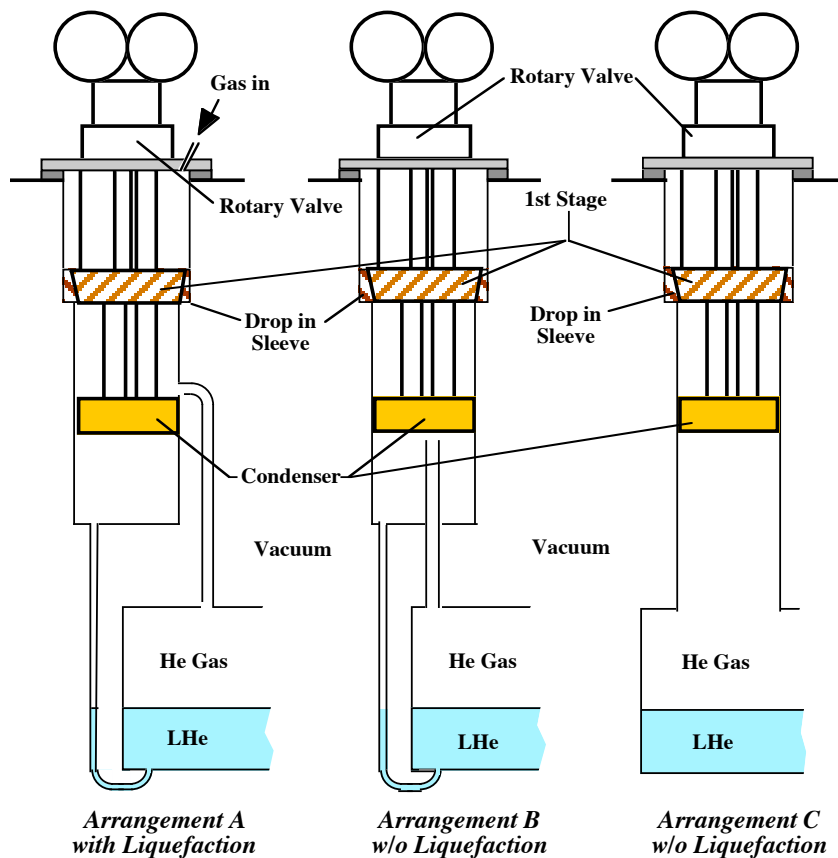

FIGURE 3. Three Arrangements of the Cooler Experiment performed using Four PT415 Coolers. (The experiment is designed to verify the cooler performance measure the ΔT between the drop-in sleeve and the first stage cold head, and measure the re-condensation characteristics of the coolers.

All three arrangements operated well in the cooler experiment. Arrangement B was used in the 1st spectrometer solenoid [13]. Re-condensation did not occur because nitrogen ice (from LN$_2$ used for the magnet cool-down) plugged the line from the condenser pot to the magnet cold mass pot, cutting off the circulation of helium. The 1st spectrometer magnet configuration had the trap shown in arrangements A and B. When the spectrometer solenoid was modified, arrangement C was used.

It is clear that arrangement A (without a trap) is the best from a number of standpoints. If the pipe from the condenser to the bottom the cold mass pot is large enough (~20 mm in diameter), a cooler can cool down a load from 300 K using natural convection. Arrangement A did liquefy helium (up to 0.3 liter per hour) into the pot.

LIQUEFACTION OF HELIUM AND HYDROGEN USING A COOLER

In order for a cooler to liquefy helium or hydrogen, the cooler cold heads must remove the sensible heat from the gas as well as the latent heat. A re-condenser removes only the latent heat as it re-liquefies the gas. For helium, the sensible heat removed from 300 K gas during liquefaction is seventy-five times the heat of vaporization. For hydrogen, the sensible heat (from 300 K to 20 K including ortho to para transition) is about eleven times the heat of vaporization. Most of the sensible heat must be removed by the upper stages of the cooler. Either a GM cooler or a pulse tube cooler can be made into a liquefier. FIGURE 4 shows the configuration of a liquefier made from a GM cooler and a pulse tube cooler. Because a pulse tube cooler can get a lot of cooling from the regenerator and pulse tubes between the stages, the liquefier is more efficient [14]. Pulse tube coolers can have low refrigeration-to-liquefaction (R/L) coefficients (~2 times the heat of vaporization). The R/L coefficient for conventional liquefiers is not nearly this low.
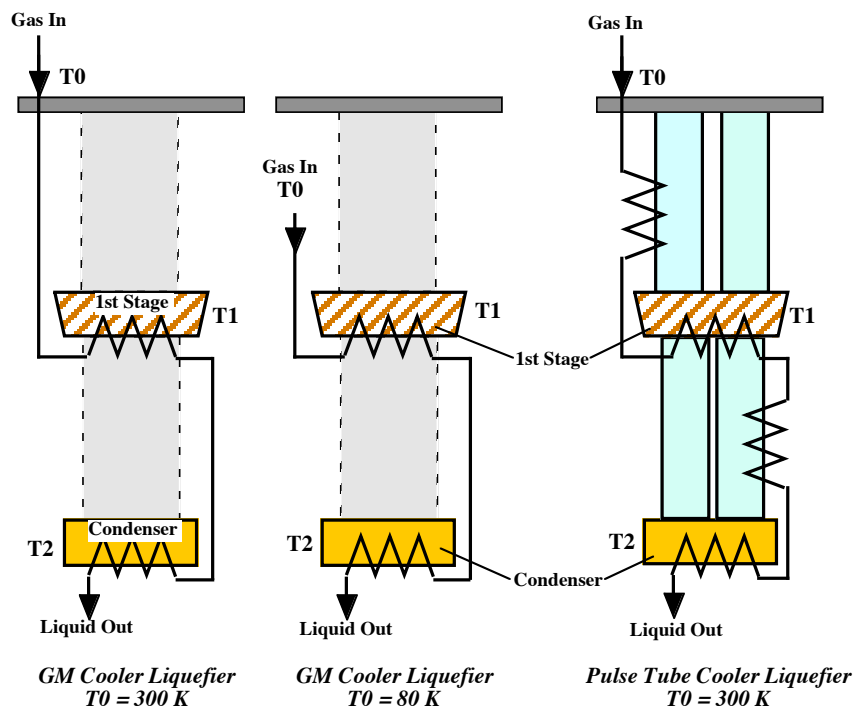

FIGURE 4. Cooler and Heat Exchanger Configurations that can turn a GM Cooler and a Pulse Tube Cooler into a Helium or Hydrogen Liquefier. (Note the GM cooler gets very little cooling from the section between stages. A pulse tube cooler can get cooling from the tubes as well as the stages themselves. The two GM cooler cases represent no gas pre-cooling (T0 = 300 K) and gas pre-cooling with liquid nitrogen (T0 = 80 K))

As an illustration of how a cooler can be made into a liquefier, a simple model was developed to show that one can estimate the maximum amount of liquefaction that is possible using a GM cooler. (See the left and center parts of FIGURE 4.) Two cases were investigated. The first case is when there is no pre-cooling from the cooler first stage and all liquefaction and pre-cooling comes from the condenser. There is a single simplified equation that can be used to estimate the mass flow M of the liquid produced (provided the area of the condenser is infinite). This expression is as follows:

$$M\left[c_p(T0 - T2) + h_{fg}(T2)\right] = Q_2(T2) = \Gamma_2(T2 - T_{M2}) \tag{4}$$

where c_p is the specific heat of the gas being liquefied. T0 is the temperature of the gas entering the system; $h_{fg}(T2)$ is the heat of vaporization of the gas at temperature T2; Q(T2) is the net refrigeration developed on the condenser that is at temperature T2; T_{M2} is the minimum temperature for the condenser cold head when Q2 = 0, and Γ_2 is the linear coefficient of refrigeration for the cooler second stage. Equation 4 can be solved directly for M, if one knows the other constants.

If most of the gas is pre-cooling is done by the first stage of a two-stage GM cooler, the following simplified equations apply (provided the heat exchanger area is infinite):

$$Mc_p(T0 - T1) = Q(T1) = \Gamma_1(T1 - T_{M1}), \text{ for stage-1;} \tag{5}$$

$$M\left[c_p(T1 - T2) + h_{fg}(T2)\right] = Q(T2) = \Gamma_2(T2 - T_{M2}), \text{ for stage-2,} \tag{6}$$

where T1 is the temperature of the first stage heat exchanger. Q(T1) is the net refrigeration generated on the first stage at temperature T1; T_{M1} is the minimum temperature on stage-1 when Q1 = 0; and Γ_1 is the linear coefficient of refrigeration for the first stage. Equations 5 and 6 can be solved simultaneously to calculate M and T1 provided the other constants are known. TABLE 3 presents the results of calculations for M and T1 for helium and hydrogen for various values of T0, with and without a first-stage heat exchanger.

TABLE 3. The Liquefaction Rate M and the 1st Stage Heat Exchanger Temperature T1 for Helium at 4.2 K and Hydrogen at 20.4 K as a Function of the Input Gas Temperature T0 with and without a Heat Exchanger cooled to Temperature T1. For He, c_p = 5.2 J g^{-1} K^{-1}, and h_{fg} = 20.7 J g^{-1}. For the H$_2$, c_p = 14.2 J g^{-1} K^{-1}, and h_{fg}= 441 J g^{-1}. For both gases, Γ_1 = 2.2 W K^{-1}; Γ_2 = 1.1 W K^{-1}.

Gas	T0 (K)	T1 (K)	T2 (K)	M (g s^{-1})
He	300	----	4.22	0.0010
He	300	35.5	4.22	0.0081
He	80	----	4.22	0.0036
He	80	31.8	4.22	0.0094
H$_2$	300	----	20.4	0.0044
H$_2$	300	59.0	20.4	0.0190
H$_2$	80	----	20.4	0.0148
H$_2$	80	37.8	20.4	0.0290

TABLE 3 shows the maximum liquefaction that can be achieved using a GM cooler. A pulse tube cooler, which has additional cooling from the regenerator and pulse tubes, will liquefy more helium or hydrogen than is shown in TABLE 3. Cryomech PT415 liquefiers will liquefy at least twice as much helium as is shown in the TABLE 3 [14].

The key to having the high liquefaction rates in TABLE 3 is good heat transfer between the heat exchanger surfaces and the gas. At mass flows M that apply for liquefaction, the heat exchanger gas flow is laminar. The liquefier heat exchangers should be laminar flow heat exchangers similar to those used in gas-cooled magnet leads [15].

The space around the tubes of a pulse tube cooler may not be in laminar flow. For laminar flow, the flow around the tubes should be within a thin annulus. Laminar flow heat exchangers generally have small passages that increase both the heat transfer area and the U factor. As a result, one cannot expect that any cooler will liquefy impure gases.

CONCLUDING COMMENTS

The LBNL cooler experiment demonstrated that re-condensation of helium can occur over a range of temperatures when the condenser has a vertical area of 0.042 m^2. One can increase the area of the condenser shown in FIGURE 2 by increasing the thickness of the copper block attached to the second stage. For helium the temperature drop is ~0.054 K over a range of 2nd-stage temperatures from 3.8 to 4.6 K. For hydrogen the temperature drop decreases from 0.47 K to 0.23 K as one increases the 2nd-stage temperature from 16 K to 24 K. Liquefaction of hydrogen has been demonstrated using a GM cooler. Pulse tube coolers can use the regenerator and pulse tubes as additional heat transfer surface for pre-cooling the gas. For helium, the liquefaction rate is increased at least a factor of two.

ACKNOWLEDGEMENTS

REFERENCES

1. Gregoire, G,, Ryckewaert, G., Chevalier, L., et al, "MICE and International Muon Ionization Experiment Technical Reference Document," http://www.mice.iit.edu, 2001.
2. Green, M. A., Yang, S. Q., Cobb, J., Lau, W., et al, "The Effect of Magnetic Field on the Position of the HTS Leads and the Cooler in the Service Tower of the MICE Focusing Magnet," *IEEE Trans. Appl. Supercon.* **18**, No. 2, p 1447, (2008).
3. Green, M. A., "How the Performance of a Superconducting Magnet is affected by the Connection between a Small Cooler and the Magnet," *IEEE Trans. Appl, Supercon.* **16**, No. 1, p 1330, (2006).
4. Baynham, D. E., Bish, P., Bradshaw T., et al, "A Liquid Cryogen Absorber For MICE," *Advances in Cryogenic Engineering* **51**, p 1068, AIP Press, Melville NY (2006).
5. Baynham, D. E., Bradshaw, T., Green, M. A., et al, "Cooling of the MICE Liquid Absorber Using a Small Cooler," *Advances in Cryogenic Engineering* **51**, p 1076, AIP Press, Melville NY (2006).
6. Kreith, F., *Principles of Heat Transfer*, International Textbook Co., Scranton PA. (1958).
7. Arp, V. D. "Thermophysical Properties of Helium-4 from 0.8 to 1500 K with Pressures to 2000 MPa," NIST Technical Note 1334 (1989).
8. Johnson, V. J. "Properties of Materials at Low Temperature," Pergamon Press, New York, (1961).
9. Frederking, T. K. H., "Peak Heat Flux and Temperature Difference in Nucleate Boiling of Liquefied Gasses," *Advances in Cryogenic Engineering* **8**. P 489, Plenum Press, New York, (1962).
10. Brentari, E. G., Smith, R. V., "Nucleate and Film Pool Boiling Design Correlations for O_2, N_2, H_2, and He," *Advances in Cryogenic Engineering* **10**, Vol, 2, p 325, Plenum Press, New York (1964).
11. Wang, L., F. Xu, F. Y., Wu, H., et al, "Magnetic and Cryogenic Design of the MICE Coupling Solenoid Magnet System," to be published in *IEEE Trans. Appl. Supercon.* **19**, No. 3, p 1344, (2009).
12. Green, M. A. and Wang, S. T., "Tests of Four PT-415 Coolers Installed in the Drop-in Mode," *Proceedings ICEC-22*, Seoul Korea, 21 to 25 July 2008, p 105 (2009).
13. Virostek, S. P., Green, M. A., Li, D., et al, "Preliminary Test Results for the MICE Spectrometer Superconducting Solenoids," *IEEE Trans. Appl. Supercon.* **19**, No. 3, p 1364, (2009).
14. Wang, C. Cryomech Inc. private communication concerning pulse tube cooler liquefiers.
15. Green, M. A., Jia, L. X., Addessi, L. J., et al, "A Design Method for Multi-tube Gas Cooled Electrical Leads for the g-2 Superconducting Magnets," *Advances in Cryogenic Engineering* **41**, p 573, Plenum Press, New York (1996).