Cryo-Cooled Low Pressure Helium Purifier

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ABSTRACT

He gas recovered in laboratories from cryogenic instruments via hoses, He bags, compressors, and pressure storage, typically has up to ½ % of impurity gases such as Nitrogen. Purification of such recovery gas is typically achieved using liquid Nitrogen cooled molecular sieve purifiers.

Here we present a molecular-sieve-free, cryo-cooled purifier for flow rates up to 20 liter/min normal He gas and pressures up to 2 atm. The purifier is designed for large impurity capacity, quick regeneration, and removes impurities to better than 5 ppm.

INTRODUCTION

The purpose of the purifier device discussed here is to purify Helium gas for use in cryo-cooled small Helium liquefiers. Cryo-cooled liquefiers require a Helium gas stream of 1 or 2 atmospheres of pressure with a purity of about 5 ppm for good liquefaction performance and long term operation without fouling. With *fouling* we mean the buildup of water and nitrogen ice on the surfaces of the cryocooler yielding significant reduction of the liquefaction performance of the liquefier.

The majority of Laboratory Helium purifiers use cartridges filled with molecular sieves like zeolite or charcoal cooled to temperatures of about 77 K. When Helium gas is passed through these cartridges, non-Helium gases like Nitrogen are trapped in the molecular sieve and the resulting Helium typically has a purity level of below 15 ppm Nitrogen. The final purity of the Helium exiting these traditional molecular sieve type purifiers is strongly a function of the contamination level of the gas that is fed in. In our experience with commercially available purifiers, we achieve gas purities of around 15 ppm, when feeding with 0.3 % pure gas at a rate of 12 liters per minute.

The main drawbacks of using a molecular sieve as the main purification agent are:

1. The amount of impurities that can be stored by the molecular sieve is typically only 10 % of its volume. In other words, a 10 liter zeolite cartridge can only capture 1 liter of Nitrogen (liquid volume).
2. When the cartridge is saturated, impure Helium gas will exit the purifier and gas purity analysis may be required to prevent this.
3. If the molecular sieve is contaminated with water, only high temperature (>100°C) can regenerate the cartridge.
4. Depending on the size of the coolant (liquid Nitrogen) container, a refill operation of the coolant liquid is required frequently.

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5. Some Liquid Nitrogen cooled purifiers require a crane to pull the cartridge for regeneration. Others use an elaborate system to dump the coolant, heat up the cartridge and refill after regeneration. Automation can often be added for a price. Often a second purifier is used, so that one can be active and the second is in regeneration.

In this paper we will present design sketches for a working prototype of a cryocooler-based Helium purifier, which achieves purities better than 5 ppm Nitrogen at a gas flow rate of up to 25 normal liters per minute.

CRYOGENIC HARDWARE

The Purifier Cartridge

Inspired by traditional designs of oil coalescers, we experimented with cartridge designs that use glass wool as the condensation medium. In the case of Helium purification we use similar glass wool as the sublimation medium.

As illustrated in Figure 1, cold Helium gas (15 to 30 K) enters the purification cartridge radially through a perforated stainless screen, radially passed through about 1.7 inches of glass wool into a center plenum, which is a perforated tube of about ¾ in diameter. The glass wool is packed to about 10 % volume density, as is typical for oil coalescers. From the center plenum the gas passes into the second stage (seen on right) where it passes through another pack of glass wool axially.

Counter flow Exchanger

The Helium gas to be purified enters the purifier at room temperature. Inside the dewar, it is in contact with the jacket of the cryocooler and is cooled to the operational temperature of 15 to 30 K. The gas passes then through the purification cartridge and flows up the spiral shaped counter flow heat exchanger. The heat exchanger is a 10 mm stainless tube which is 6 m long and provides better than 95 % of the cooling work for the incoming gas for gas flow rates below 25 N-liters per minute. This is very important if one desires to use a modestly sized cryocooler for this application. The efficiency of the counter flow exchanger will degrade over time as it is coated with Nitrogen ice and other contaminants. See Figure 2 for the arrangement of the exchanger in the purifier.

Dewar

The dewar is a standard aluminum construction with a figerglass insert that serves as neck and belly. A radiation shield is also included since temperatures below 30 K are to be achieved.

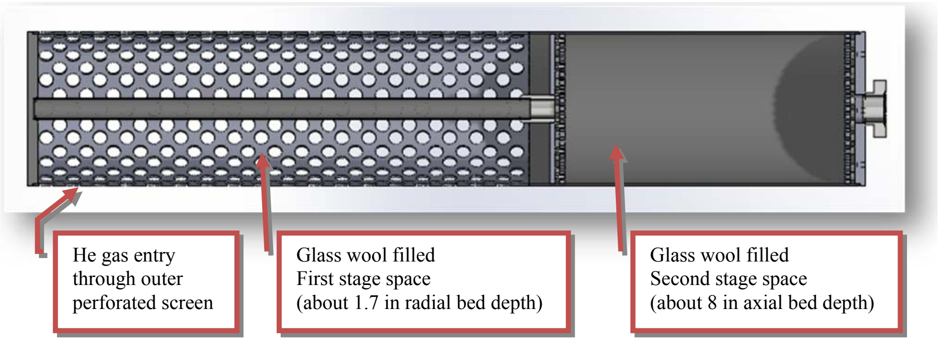

Figure 1. Current purification cartridge design (left is bottom)

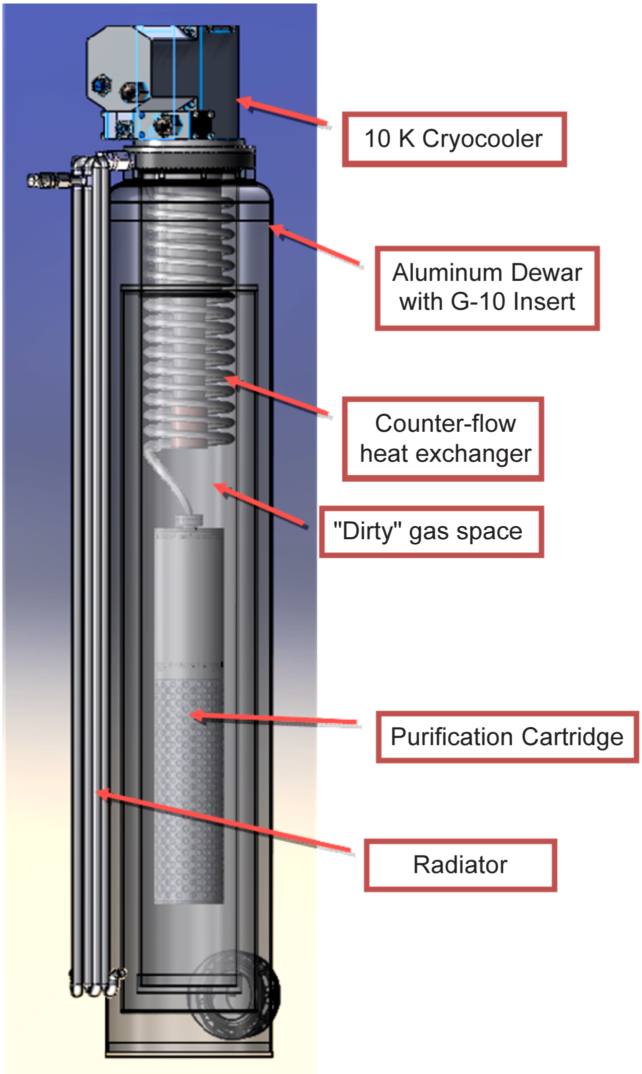

Figure 2. Model of entire cryogenic hardware

Radiator

As indicated in Figure 2, the inlet and outlet gas passes through a radiator on the outside of the dewar. This has no purpose during normal purification operation, but only helps to warm up the gas, which is pushed out of the dewar during the regeneration cycle.

OPERATIONAL DATA

We have had a prototype purifier operating for seven months since December 2011. It has been continuously feeding a liquefier at a gas flow rate of > 12 normal liters per minute. Gas purity of the purified Helium was measured by a residual gas analyzer (RGA). The main contaminant in the purified gas is Nitrogen gas at less than 5 ppm, near the analyzers detection limit. In the seven months of operation, the purifier has not been warmed up to room temperature. It is important to note, that the un-purified Helium gas is “dry”. Water vapors are removed from the Helium gas at the high pressure (170 bar) storage to a dew point below -30 Celsius.

The purity of the un-purified Helium gas was also measured with the RGA and, while fluctuating, is usually between 0.2 % and 0.5 % Nitrogen as the main contaminant.

We also note, that the prototype is fitted with a single (radial) stage purification cartridge with 2 liters of glass wool volume.

Regeneration

The purifier is regenerated weekly by warming the dewar insert to above 100 K to 120 K. The warming is accelerated with a 100 watt heater around the cartridge, and the cold head is turned off. While the cartridge thermometer diode is below 65 K, the inlet and outlet are connected to a Helium recovery system, then the inlet and outlet are vented to air. Once, the cartridge reaches 100 K, the dewar insert is optionally evacuated by a rotary pump. Subsequently, the cold head is turned back on, and the inlet is again connected to the un-purified Helium source. After the cartridge temperature drops below 30 K, the outlet is flushed for 15 minutes to the recovery system, and then is reconnected to the liquefier. The whole process takes about 3 hours.

About 0.3 to 0.5 liters of solid Nitrogen ice are sublimated weekly in the purification cartridge, which represents up to 25 % of the cartridges volume.

CONCLUSION

A simple but effective design has been presented for a cryo-cooled Helium gas purifier. All of the drawbacks of traditional Liquid Nitrogen cooled purifiers, as listed in the introduction, are positively addressed by this design.

Our experiments showed that as long as the cartridge temperature is kept below 30 K and the gas flow below 30 normal liters per minute, the quality of the purified Helium gas is good (< 10 ppm). If these conditions are met, the purifier will plug before producing un-pure gas.

PATENT PENDING

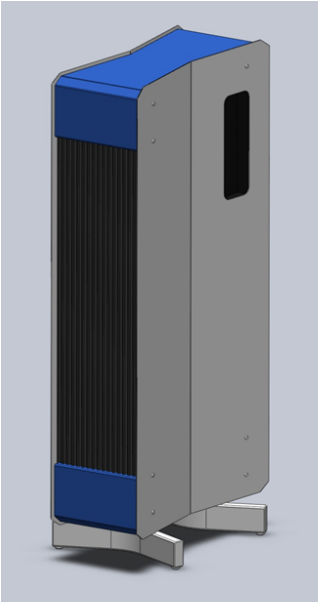

Figure 3. Purifier enclosure