Freeze-out purifier for helium refrigeration system applications

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Abstract. Purification systems are necessary to support commissioning and operation of helium refrigeration and associated experimental systems. These systems are typically designed for a low level of impurity (i.e., in parts per million), since a 4.5 K or 2 K helium system will solidify, or freeze out every other substance. The trace impurities can block and/or change the flow distribution in heat exchangers and potentially damage turbines or cryogenic compressors operating at high speed. Experimental systems, such as superconducting magnets, require helium purification due to inherent characteristics in their construction. These are also used for the commissioning of sub-systems, like the compressors, and cold boxes. As known from experience, molecular sieves do not remove low-level moisture impurity sufficiently. Typical commercial freeze-out purifiers using molecular sieves have very short operating times between regeneration and are inefficient requiring substantial utilities like liquid nitrogen and high-pressure operation. Based upon proven experience from a freeze-out purifier design for Brookhaven National Lab (BNL) in 1983, a liquid nitrogen assisted freeze-out purifier has been designed. This design includes a multi-pass and multi-stream heat exchanger and an activated carbon bed. The heat exchanger design is expected to minimize the liquid nitrogen usage and extend the capacity and the operating pressure range, thereby the time interval between regeneration. The goal is to provide a simple design procedure to develop and operate an efficient purifier system.

1. Background
Helium has a wide range of applications in various scientific, space, medical and process industries. These industries take advantage of helium’s very low (cryogenic) boiling temperature and its chemically inert characteristic. The known helium reserves are depleting, and this is reflected in the recent price escalations. Hence, not only from a technical aspect, but economics too dictate the need for helium purification / recirculation and minimize wastage from the aforementioned applications. In 2015, more than one-third of the total helium consumption in US was in the cryogenic refrigeration sector [1]. Cryogenic refrigerators which utilize helium as a refrigerant are necessary for systems using superconducting devices, such as magnetic resonance imaging and particle accelerators. These refrigeration systems operate at 4.5 K (the just above normal boiling point of helium), down to 1.8 K (which requires helium with vapor pressure of 16 mbar). At these very low temperatures, the presence
of any other substances (contaminants) except helium will result in solidification. This can lead to
damage to moving parts of the cryogenic system and/or affect the flow distribution in heat exchangers
and flow blockage in valves. Obviously, these can have a deleterious effect on the refrigerator capacity
and operations. Although, usually, better than industrial Grade-A (also Grade 4.7) purity helium is used
in these refrigerators, contaminants are inadvertently introduced to the system through residuals leftover
from a clean-up, air in-leaks to systems operating below 4.5 K, and out-gassing from cooled devices
e.g., magnets). The constituents from the first two are of oxygen, nitrogen and moisture. After the initial
clean-up, these constituents are present in relatively low concentrations, of the order of 10 ppm or less.
Although, this seems small, it can (and does) build up over time and consequently pose threat to the
reliable and efficient operation of the equipment.

Traditional helium purification systems used for large-scale cryogenic refrigerator applications use
molecular sieve for moisture removal and an activated carbon bed cooled with liquid nitrogen (LN) to
remove air contaminates [2]. However, from operational experience at Jefferson Lab and the Spallation Neutron Source [2], it was found that the molecular sieve is unable to remove low level moisture
sufficiently, despite reasonable regeneration practices. This was evident from the pressure build-up in
the helium-helium-nitrogen heat exchanger used to cool the helium to liquid nitrogen temperatures. To
address these issues, several different methods of low level impurity removal [3-5] have been
investigated in the past, including freeze-out (or refrigeration) purification [4]. For the latter, a heat
exchanger specifically designed to accommodate the solidified moisture from a contaminated helium
stream is used, rather than molecular sieve. This is a very effective method for removing low level
moisture contamination due to the very low saturation vapor pressures. However, it requires a heat
exchanger design that is well suited for contaminant solidification distribution and minimal impact on
flow distribution. Typical commercially available freeze-out purifiers have a much shorter operating
time in between regenerations and are not optimized for low pressure operation or efficient LN usage.
As such, there is a need for fundamental improvements of this critical sub-system.

In the present paper, the development of a helium purification system utilizing freeze-out purifier
heat exchanger is reported. The purification system is designed to remove low level impurities (mainly
air), typically present in systems using superconducting devices at or below 4.5 K. The goal is to provide
a simple design procedure to develop an energy/utility efficient helium purifier with long operating
interval between regenerations. This purifier will serve as the primary helium purification system for
MSU-FRIB cryogenic refrigerator and superconducting magnet testing facility.

2. Process design

Referring to figure 1, the helium purification process in the freeze-out purifier begins with the
contaminated helium cooled to around 85 K in a counter-flow helium-helium-nitrogen heat exchanger
(HX-1 and HX-2 in figure 1). Any moisture in the contaminated helium stream is solidified on the HX-1
surface. The contaminated helium is then cooled to about 80 K in a LN boiler, after which it flows
through the activated carbon bed (also at 80 K) where the remaining contaminates (like oxygen and
nitrogen) are removed. Pure helium leaves the carbon bed, and its enthalpy is recovered in the counter
flow heat exchanger, exiting near ambient conditions from HX-1. Design goals for the freeze-out
purification system are listed in table 1.

| Table 1. Design requirements for the freeze-out purification system. |
|---------------------------------------------------------------|
| Mass flow rate (helium) | 30 g/s |
| Operating pressure (helium) | 6.0 to 16.0 bar |
| Design pressure (helium) | 18.0 bar |
| Design max. pressure drop (helium) | 0.25 bar |
| Design max. contamination (helium) | 30 ppm |
| Minimum time between regenerations | 14 days |
| Design LN usage | 8.0 g/s |
Figure 1. Simplified flow diagram of the freeze-out purification system.

Figure 2. Solid-liquid (S-L) saturation temperature of moisture as function of the mole fraction at different stream (operating) pressures [6]
3. Heat exchanger design

The heat exchanger is a major and critical component of the freeze-out purification system. Its effectiveness plays an important role in the purification capacity and LN consumption of the system. The type of heat exchanger is paramount to achieving the desired design requirements in a cost-effective manner. For this application, the coiled fin-tube heat exchanger type was selected, which is somewhat similar to those used in the small-scale refrigerators, and also known as a Collins heat exchanger. The model for this heat exchanger was developed following the work reported by Yuksek [7], studied for the Linde 1600 helium refrigerator. This type of heat exchanger is comprised of one or several tubes wrapped fin-to-fin, in a helix around a mandrel, and enclosed by an outer shell. There can be one or multiple passes that are arranged in one or multiple wraps (‘layers’). Multiple passes allow for higher volume (mass) flow, at a lower pressure drop and thus supporting low pressure operation to reduce compressor power. However, these multiple passes increase the heat exchanger mechanical design and fabrication complexity.

![Figure 3. Heat exchanger cooling curves for (a) HX-1 and (b) HX-2.](image)

The contaminated helium flows in the annular space in-between and over the finned-tubes in a locally cross-flow manner (although the heat exchanger is overall in a counter-flow configuration). This design inherently has the characteristics for high contamination holding capacity with lower impact on the heat exchanger performance. The purified helium stream flows through the tubes which are wound about a mandrel and bounded by the outer shell. For this design six parallel passes of coiled fin-tubes (12.7 mm outside diameter tube, 4.8 mm fin height and 0.5 mm fin thickness) are used. For geometrical compactness and segregation of the trapped contamination (moisture), the heat exchanger is physically split into two sections (HX-1 and HX-2 referring to figure 1). HX-1 is designed for freeze-out entrapment of the moisture from the contaminated stream. Figure 2 shows the calculated solid-liquid (S-L) saturation temperature of moisture at the stated (total) pressure. The S-L saturation temperature of moisture is calculated using Raoult’s law of partial pressures and polynomial fits to measured saturation temperatures obtained from [6]. It is observed that the S-L saturation temperature varies between 200 K and 240 K over the range of operating pressures. As such, HX-1 is designed to cool the contaminated stream from 300 K to 180 K. In this way, the trapped moisture stays in this section which facilitates the regeneration. An additional coiled fin-tubing is used in this heat exchanger section to recover the refrigeration from the nitrogen vapor stream (exiting the nitrogen boiler). HX-2 is designed to cool the
contaminated stream from 180 K to 80 K, recovering the exergetically more valuable the refrigeration from the purified helium stream. The calculated cooling curves for both of these heat exchangers are shown in figure 3. From the HX-1 cooling curve, it can be estimated that approximately 25% of HX-1 axial length (as indicated by the percent total NTU’s) is required to reduce (i.e., solidify) the contaminated stream moisture content from the design maximum of 30 ppm to 0.3 ppm (i.e., 1% of the initial value). The heat transfer surface area corresponding to this length is approximately 6.5 m².

Based on a design goal of maximum pressure drop of 0.25 bar and the surface area available to capture the moisture, it is estimated that up to 2.5 kg of moisture can be captured by HX-1. A parametric study on the effect of the operating pressure on the purifier operating time was performed and the results are shown in figure 4. From this figure, it is observed that with a moisture concentration of 30 ppm (at 30 g/s), the operating period of the purifier (i.e. time before HX-1 reaches a pressure drop of 0.25 bar) is about 30 days or longer.

The nitrogen boiler and the absorber bed are the other major components of the purification system. The design of these two components were performed following [8]. Based on an estimated LN consumption of 8.0 g/s (approx. 0.05 m³ of saturated liquid nitrogen at 1.01 bar, per hour), a 0.17 m outside diameter (OD) vessel (approx. 0.05 m³ volume) was selected for the nitrogen boiler. The adsorber bed design is also conventional, and is based on the operational experience from the purifier designed in [8]. A 0.33 m OD vessel with an aspect ratio of approx. 7.0 was selected. Activated charcoal (coconut shell carbon) is selected to be the adsorbent material. Following a detailed analysis (using the methods described in [8]), it is found that approximately 0.12 m³ of the adsorbent is required to remove the nitrogen from the contaminated helium stream. The estimated break-through period for the absorber bed is found to be slightly over 20 days (with 30 ppm of nitrogen in 30 g/s helium stream). The effective freezing surface can also be increased by influencing the cooling curve with increased LN boil off; thus increasing the time in between the regenerations as demonstrated in the BNL purifier.

![Figure 3. Calculated cooling curves for both of these heat exchangers.](image)

**Figure 3.** Calculated cooling curves for both of these heat exchangers.

4. Mechanical design
The purifier design described in this paper has three major pressure vessels – freeze-out heat exchanger, nitrogen boiler and absorber bed. All these pressure vessels operate at cryogenic temperatures and are
enclosed in a vacuum insulating shell. The complete purifier assembly along with its major components are shown in figure 5.

Mechanical design of the purifier piping and pressure vessels were performed following American Society of Mechanical Engineers (ASME) B31.3 Code and ASME Boiler and Pressure Vessel Code (BPVC), respectively. Piping flexibility analysis for the cryogenic process piping was performed for design in accordance with the ASME B31.3. Pressure design of different novel components were carried out using finite element analysis and following ASME B31.3 and BPVC (as applicable). Basic dimensions of the major components of the purifier are listed in table 2, and their design details are discussed in the following sub-sections.

**Table 2. Basic dimensions of the major components of the purifier.**

| Components                  | Outside Diameter (m) | Shell Thickness (mm) |
|-----------------------------|----------------------|----------------------|
| Insulating vacuum shell     | 0.91                 | 7.92                 |
| HX-1 Mandrel                | 0.27                 | 6.35                 |
| HX-2 Mandrel                | 0.36                 | 7.92                 |
| HX-2 Shell                  | 0.41                 | 4.78                 |
| Nitrogen Boiler Shell       | 0.17                 | 3.40                 |
| Carbon Bed Shell            | 0.33                 | 4.57                 |

*Figure 5. Sketches showing (a) complete purifier assembly, (b) nitrogen boiler, (c) freeze-out heat exchanger (without outermost shell) and (d) carbon bed.*
4.1. Freeze-out heat exchanger
As mentioned in section 2, the freeze-out heat exchanger is physically split into two sections. Each of these sections (HX-1 and HX-2) consist of copper finned-tubes coiled around a mandrel and are enclosed by a shell creating an annular vessel. For geometrical compactness, HX-2 is concentrically nested inside HX-1. Annular flat heads at the ends of both heat exchangers serve as headers for tube (pure helium) and shell (contaminated helium) flows. As shown in figure 6 (a), the outlet of HX-1 allows for extraction of collected moisture during regeneration, without carrying it over into HX-2 inlet.

4.2. Adsorber bed
The adsorber bed is comprised of two pressure vessels, one nested inside the other. The outer vessel holds the adsorbent (activated carbon) in a fixed bed, while the inner vessel is mounted at the centerline of the fixed bed, supported by the inlet and outlet process piping. LN flows through the inner vessel keeping the adsorbent at a constant temperature (approx. 80 K). The adsorbent is held in place within the fixed bed using layers of wire mesh screens and fiber-glass filter. In addition, sintered metal filters are used at the inlet and outlet nozzles to the adsorber bed to prevent any carry-over dust from the exiting pure helium. Band heaters are mounted to the outer vessel shell for the regeneration process. A detailed cross-sectional view of the adsorber bed assembly is shown in figure 6 (b).

Figure 6. Detailed cross-sectional view of (a) freeze-out heat exchanger assembly, (b) adsorber bed assembly and (c) nitrogen boiler assembly.
4.3. Nitrogen boiler and miscellaneous components
As shown in figure 6 (c), the nitrogen boiler consists of six parallel passes of stainless-steel tubing coiled inside a vessel. Contaminated helium from the freeze-out heat exchanger (HX-2) outlet flows through the coiled tubing submerged in the liquid nitrogen and is then fed to the adsorber bed. The nitrogen boiler is nested inside the annular space of HX-2 for compactness and minimizing radiation heat in-leak to the liquid nitrogen bath.

There are several other key components in the purifier. The insulating vacuum shell is constructed from a 36 NPS pipe section and has a standard ASME dished head. All the components inside this shell are mounted from this head. The insulating vacuum shell is attached to the head using a flanged connection, allowing access to the inner cryogenic components without cutting the vacuum shell. Cryogenic valves, instrumentation and maintenance ports are mounted to the top and side of the dished head. All cryogenic components are wrapped with multi-layer insulation (MLI) to minimize radiation heat in-leak to the process. In addition, there is an external valve and instrumentation panel (not shown). A recirculation blower will be used for warm up of the heat exchangers from 300 K end circulating helium in the tube side and band heaters for carbon bed warmup. The LN vessel in the carbon bed will assist the cool down following regeneration.

5. Summary
The design of a helium purification system utilizing freeze-out heat exchanger for application in systems requiring helium refrigeration is reported. The purification system is designed to remove low level air impurities. Key features of the process and mechanical design are discussed in this paper. Detailed analysis of the purification system demonstrates an effective and efficient design for supporting the 6-16 bar operation, with operating period greater than 30 days at design contamination level of 30 ppm and an LN consumption of approx. 0.05 m$^3$/hr. at full capacity. This design and analysis has shown, the procedure developed can be a good tool to serve as the primary helium purification system for MSU-FRIB cryogenic refrigerator and superconducting magnet test facility.

6. References
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Acknowledgments
This material is based upon work supported by the U.S. Department of Energy Office of Science under Cooperative Agreement DE-SC0000661, the State of Michigan and Michigan State University. Michigan State University designs and establishes FRIB as a DOE Office of Science National User Facility in support of the mission of the Office of Nuclear Physics.