Design and analysis of the helium purification system for the NSRRC cryogenic system

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Abstract. Helium is an expensive consumable in cryogenic facilities and is used widely in space, medical and energy research. At NSRRC, liquid helium is used as a coolant for superconducting magnets and SRF cavities. Minor contaminants such as nitrogen, oxygen, moisture and oil will be picked up when liquid helium circulates in large scale cryogenic systems and such contaminants can crystallize and cause damage to the cold box turbo expanders resulting in system damage and failure. Therefore, a helium purification system is designed as an integral part of the cryogenic system to conserve helium by providing 99.9995% pure helium to the liquefier after eliminating contaminants. The NSRRC helium purification process is based on two principles, the first one being a cryo-sorption device using activated charcoal and a molecular sieve and the other being a cryo-condensation unit using a tubular heat exchanger. The purifier has been designed to purify impure helium with overestimated contaminants of as much as 2.5% nitrogen and 2.5% oxygen with a mass flow rate of 475 nm³/hr and delivering a pressure of 17 bar(a) of impure helium to the purifier, the actual impurity will be much lower than the actual design contaminants. In this paper, calculation and design of the helium purification system and components composed of one double pipe counter flow heat exchanger, one vessel and tube heat exchanger, one pre-cooler and one charcoal vessel will be discussed together with charcoal mass requirement calculations and design of other components. In NSRRC, helium is liquefied and is used as a coolant for the SRF system and for cryogenic undulators. During a cryogenic cycle in the cryogenic system, helium may pick up contaminants such as moisture, oxygen, oil, and nitrogen, which have a higher freezing point than liquid helium and will crystallize. These frozen impurities will then affect plant capacity and operation such as alternating flow characteristics, damaging moving parts like turbines in the cold box causing the overall cooling efficiency to drop. Eliminating the contaminants is therefore very important for the cryogenic system.

1. Introduction
The Helium Purifier for the NSRRC is split into two major parts as shown in Figure 1. The first part is to remove any excess moisture and the second to remove nitrogen, oxygen and oil. To remove moisture, a moisture separation vessel filled with a Molecular sieve was designed. The moisture separation vessel at 300K will remove most of the moisture in the system. Other parts such as the vessel, tube heat exchanger and the crystal filter will remove the remaining moisture ice crystals as shown in Figure 2.
Nitrogen, oxygen and oil contaminants are removed at 80K with carbon from coconut shells.

Figure 1. P&ID diagram of the NSRRC Helium Purifier System.

Figure 2. Vessel and Tube Heat Exchanger

2. Design of the Carbon Bed
To design the Carbon Bed, the carbon mass requirements, the size, length and diameter of the carbon vessel have to be calculated.
2.1 Mass Requirement of Carbon
We assume that the maximum impurity of the helium gas is 2.5% nitrogen and 2.5% oxygen. The saturation pressure of nitrogen is 1 bar and that of oxygen 0.2 bar at 77 K. According to Raoul’s law, partial vapor pressure of each component of an ideal mixture of liquids is equal to the vapor pressure of the pure component multiplied by its mole fraction in the mixture given by

\[ p = P x \]  

(1)

where \( p \) is the partial gas pressure, \( P \) its saturated pressure of the same gas at the same temperature and volume and \( x \) is the mole fraction. The partial pressure of air at 77K before entering the carbon bed will be

\[ p_{air} = p_{O_2} + p_{N_2} \]  

(2)

Due to the minimum purifier adsorption pressure \( p_{\text{min}} \) of 17 bar(a), the mole fraction of air \( X_{air} \) at 77K can be calculated by

\[ X_{air} = \frac{p_{air}}{p_{\text{min}}} \times 100 \]  

(3)

and the quantity of nitrogen \( D_{N_2} \) to be adsorbed is given by

\[ D_{N_2} = X_{air} \cdot Q \cdot t \]  

(4)

where \( Q \) is the flow rate (475 nm\(^3\)/h) and \( t \) is the time (6hrs). \( D_{N_2} \) can be calculated from \( V_a = 5.03 \times 10^6 \) cc. To calculate the mass of carbon needed, the Brunauer–Emmett–Teller (BET) theory is used [1] for which the equation is

\[ V_a \cdot m_a = \frac{u_m c x}{1-x} \left[ \frac{1-\left((n+1)x^n + n x^{n+1}\right)}{1+c(n-1)x^n+c x^{n+1}} \right] \]  

(5)

where \( V_a \) is the total gas volume adsorbed under standard conditions (\( T=273.15 \) K and \( P=101.3 \) kPa), \( m_a \) is the mass of adsorbent, \( u_m \) the gas volume of a monolayer of coverage, \( p \) the partial pressure of the gas being adsorbed, \( p_{\text{sat}} \) the saturation pressure of the gas being adsorbed at the temperature of the adsorbent, \( n \) is the number of layers, \( x = \frac{p}{p_{\text{sat}}} \) = 17 and \( c \) is a function of energy of adsorption and the temperature of the adsorbent, \( c = \exp \left( \frac{\theta_a}{P_{\text{low}}} \right) = \exp \left( \frac{300.2}{77} \right) = 49.338 \).

Equation (5) is reduced to the Langmuir equation (for \( n = 1 \))

\[ \frac{V_a}{m_a} = \frac{u m \left( \frac{p}{p_{\text{sat}}} \right)}{1+\left( \frac{p}{p_{\text{sat}}} \right)} \]  

(6)

Equation (6) is solved to find the mass of adsorbent \( m_a \)

Therefore, the mass requirement for activated charcoal \( m_a \) can be calculated as 27.75 Kg. As suggested by Haselden [2], the estimate of the adsorbent requirement is based on 70% saturation of the carbon bed and therefore the minimum amount of carbon required will be 39.64 Kg.
2.2 Design of the Carbon bed vessel

The operating pressure of the purifier is 17 bar(a) and the total flow rate is 4755 Nm$^3$/hr max.

Therefore, the helium flow rate at 17 bar(a) and 77 k is $0.0022\text{m}^3/\text{sec}$, which can be determined from the gas equation:

$$\frac{P_1V_1}{T_1} = \frac{P_2V_2}{T_2} \quad (7)$$

Where $P$ is the pressure of gas, $V$ is the volume of gas and $T$ is the absolute temperature of gas.

The dimensions of the carbon bed have been chosen to keep the gas velocity near $10-2\text{m/sec}$, because the gas approach velocity ($V_2$) through the fixed carbon bed ranges from $10-2$ to $1\text{ m/sec}$ [3]. The inner diameter $D_i$ of the carbon vessel can then be calculated to be $280\text{ mm}$ using the equation

$$V = (\pi/4)\times D_i^2 \quad (8)$$

A pipe should be selected with an inner diameter greater than $280\text{ mm}$ to obtain an optimum value for the gas velocity through the carbon bed. The carbon vessel will be immersed in LN$_2$ and will be warmed up to $70^\circ\text{C}$ during regeneration heating and therefore $316\text{ L}$ stainless steel is selected for the carbon vessel pipe material. We select a 12-inch pipe for the helium gas under a pressure of 17 bar(a) following ASME B 31.3 (1999). For the process piping, the allowable internal pressure is calculated by

$$P = (2 \times t_{\text{min}} \times S) / [\text{OD} - (2 \times Y \times t_{\text{min}})], \quad (9)$$

where, $P = 17\text{ bar or } 0.246\text{ ksi}$, the allowable stress $S = 16.7\text{ ksi}$ for SS316L, $Y = 0.40$ for $t_{\text{min}} < \text{OD}/6$, $Y = (\text{OD} - (2 \times t_{\text{min}})) / 2(\text{OD} - t_{\text{min}})$ for $t_{\text{min}} = \text{OD}/6$, $t_{\text{min}}$ is the minimum wall thickness, OD the nominal outside diameter. Applying Eq. (9), we get $t_{\text{min}} = 2.3713\text{ mm}$ showing that the pipe size for 12” Sch 5 (Outer Diameter (OD) = 323.85 mm, t = 3.96 mm) is suitable.

2.3 Length Calculation of the Carbon vessel

To calculate the carbon vessel length, the density of the carbon is needed. For the NSRRC purifier, active charcoal with a density of $550\text{ kg/m}^3$ is used. For an inside diameter of the charcoal vessel (size 12” Sch 5) of $d_i = 0.31593\text{ m}$, an activated charcoal mass of $39.64\text{ kg}$ and a vessel length of $0.9\text{ m}$ can be calculated using the equation below.

$$\pi/4 \times d_i^2 \times L \times \text{density}(\rho) = \text{Mass of activated charcoal} \quad (10)$$

Due to the area of a 3-inch pipe in the vessel, $3.1\text{Kg}$ of charcoal needs to be added and thus a 1m long 12-inch pipe will be needed.

3. Pressure Drop

The pressure drop of the NSRRC purifier within a packed bed containing particles is calculated as $62.333 \times 10^{-5}\text{ bar}$ by using the Ergun Equation [4]

$$f_p = \frac{150}{Gr_p} + 1.75 \quad (11)$$

where $f_p$ and $Gr_p$ are defined by

$$f_p = \frac{\Delta p}{L} \frac{D_p}{\rho u_2^2} \left(\frac{\epsilon^3}{1-\epsilon}\right) \quad (12)$$

and
Here $Gr_p$ is the modified Reynolds number, $f_p$ is the packed bed friction factor, $\Delta p$ is the pressure drop across the bed, $L$ is the length of the bed, $D_p$ is the equivalent spherical diameter of the packing, $\rho$ is the density of the fluid, $\mu$ is the dynamic viscosity of the fluid, $v_s$ is the superficial velocity and $\epsilon$ is the void fraction of the bed. It is evident from the Ergun equation that the pressure drop within the carbon vessel is $62.333 \times 10^{-5}$ bar, which is not significant in comparison with the system pressure.

4. Design of the Pre-cooler

The pre-cooler of the NSRRC purifier is placed between the LN$_2$ Vessel and the carbon vessel and is fully submerged in liquid nitrogen. The purpose of the pre-cooler is to cool the helium gas to 80K before entering the carbon bed as shown in Figure 3.

![Figure 3. Pre-Cooler.](image)

To find the length $L$ and diameter $D$ of the copper tubing for the heat exchanger, some assumptions need to be made as follows: $T_s$ is a constant at 77 K, the helium gas parameters are ($Cp$=5.2 kJ/kg K; $\mu$= $15 \times 10^{-6}$ Pa s; $\rho$= 0.3 kg/m$^3$, $k$ = 0.1 W/m K), the allowed pressure drop, $\Delta p$ = 10 kPa and the helium mass flow rate is 15 g/s.

A total heat transfer $Q$ of 17160W at a mean temperature of $\Delta T_{lm} = 51$K is calculated by

$$Q = \dot{m}C_p(T_{in} - T_{out}) \quad (14)$$

$$\Delta T_{lm} = \frac{\Delta T_f(x=0) - \Delta T_f(x=L)}{\ln(\frac{\Delta T_f(x=0)}{\Delta T_f(x=L)})} \quad (15)$$

where

$$UA = h\pi DL = \frac{Q}{\Delta T_{lm}} = 336.5 \text{ W/K} \quad (16)$$

The heat transfer coefficient is a function of Reynold Number $Re_D$ and Prandtl number $Pr = 0.67$.

Assuming a fully developed turbulent flow and using the Dittus Boelter correlation

$$NU_D = \frac{hD}{K_f} = 0.023Re_D^{0.8}Pr^{0.3} \quad (17)$$
\[ Re_D = \frac{4m}{\pi D \mu} \]  \tag{18}

Where \( h \) is the convective heat transfer coefficient of the flow, \( D \) is the diameter of the pipe and \( K_f \) is the thermal conductivity of the fluid.

We substitute \( Re_D \) and solve for \( h \)

\[ h = 0.023 \frac{K_f}{D} \left( \frac{4m}{\pi D \mu} \right)^{0.8} Pr^{0.3} = \frac{0.63}{D(m)^{1.8}} \]  \tag{19}

Substituting equation (16) and (19), equation (20) may be formulated as shown below.

\[ \frac{L}{D^{0.8}} = 169.95 \text{m} \]  \tag{20}

The pressure drop equation provides the other equation for \( L \) and \( D \)

\[ \Delta P = f \frac{L}{2 \rho D} \left( \frac{m}{A_{flow}} \right)^2 \]  \tag{21}

where \( A_{flow} = \frac{\pi D^2}{4} \) and \( f \approx 0.02 \). Substituting for \( Re_D \) and \( f \)

\[ \Delta P = 0.016 \frac{m^2}{\rho} \frac{L}{D^5} = 1.2 \times 10^{-5} \left( \frac{L}{D^5} \right) \]  \tag{22}

From equations (20) (22) with \( \Delta p = 10,000 \text{ Pa} \), we can calculate that \( D = 0.026 \text{m} \) and \( L = 9.17 \text{m} \).

5. Design of a Double Pipe Counter-Flow Heat Exchanger

A double pipe counter flow heat exchanger was designed to cool impure helium from 300 K to 120 K and to heat pure helium from 77 K to 300 K as shown in Figure 4.

![Figure 4. Double Pipe Counter-Flow Heat Exchanger.](image-url)
To calculate the length of the Double pipe counter flow heat exchanger, equation (19) is used. From the previous section, a 26mm inner diameter pipe has been selected for the outer pipe and a 17mm inner diameter pipe for the inner pipe. Using the same diameter pipe and substituting in equation (19), $h$ is calculated as 449.2 W/m² K and the overall heat transfer coefficient can be calculated to be 224.6 W/m² K using the equation below while $\Delta T_{lm}$ is determined by equation (15).

$$U = \left( \frac{1}{h} + \frac{1}{h} \right)^{-1} \quad (23)$$

The length of the double pipe counter flow heat exchanger can be calculated by substituting $q$, $U$, $D$ and $\Delta T_{lm}$ in equation (24).

$$L = \frac{q}{U \pi D \Delta T_{lm}} = 77m \quad (24)$$

6. **Design of a Phase Separator Vessel**

The phase separator is fully submerged in the LN$_2$ vessel and uses a cyclone motion inside the vessel for efficient separation of the liquid phase from the gas stream, which is collected at the bottom due to gravity. For design purposes, let us consider 2.5% nitrogen impurity instead of air impurity in the helium gas flowing at a rate of 475 nm$^3$/hr and a delivery pressure of 17 bar(a). The remaining nitrogen after the phase separator vessel is only 0.18%. Hence, the amount of nitrogen separated in the phase separator is 2.32%, which is about 11.02 nm$^3$/hr of nitrogen gas. This amount of nitrogen will be separated as LN$_2$ which is about $4.37 \times 10^{-6}$ m$^3$/sec. Using 4” Sch 10 pipe, for example, the volume of LN2 produced in 15 min is around $3.933 \times 10^{-3}$ m$^3$ and, the length of the vessel can be calculated to be 427.74 mm. Adding the length for cyclonic movement which is two times the inner diameter of the pipe (0.216m), the minimum vessel length needed will be 0.643m.

7. **Conclusion**

A helium purifier for the NSRRC cryogenic system has been designed and is now being manufactured. The purifier has been designed for impure helium with contaminants of 2.5% nitrogen and 2.5% oxygen at a mass flow rate of 475 nm$^3$/hr and delivery pressure of 17 bar(a). After the helium purifier is constructed, experiments will be conducted.

8. **References**

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