"High Vacuum System for Recovering ^{3}He from Decommissioned Target Cells"

Senior Physics Research

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ABSTRACT

To learn more about the structure of the neutron, researchers at the Thomas Jefferson National Accelerator Facility (JLab) use glass cells filled with polarized ^{3}He to perform electron scattering experiments. To allow for better polarization, the cells are now using a mixture of K and Rb alkali metals to polarize the ^{3}He by a Spin Exchange Optical Pumping technique whereas before the cells only used Rb for that purpose. Due to this change in the target cells fabrication, the lab at the College of William and Mary was left with cells filled with ^{3}He that cannot be reused. Recent constraints on worldwide ^{3}He supply have further intensified the desire to recycle unused ^{3}He gas. For this research project, a technique was developed to recover the ^{3}He from the older, Rb filled cells.

I Introduction

^{3}He is used for electron scattering experiments to study the behavior of the neutron, medical research, low-temperature physics research, oil and gas detectors, and neutron detectors. The neutron detectors used for homeland security, as of 2002, have dominated the use of ^{3}He supply of the United States [2].

^{3}He is produced through tritium decay:

$$^{3}H \rightarrow {}^{3}He + \beta + \upsilon \qquad (1)$$

Where β represents a beta-particle and υ represents a neutrino. Because tritium is used in nuclear weapons to start a chain fission reaction, its storage in the weapons and subsequent

decay traps ^{3}He gas in the weapons. This allows one to gather ^{3}He gas when refurbishing or taking apart nuclear weapons containing tritium [2].

The United States supply of ^{3}He is due in large part to the supply of nuclear weapons previously developed during the Cold War with the Soviet Union and its stockpile of ^{3}He has dwindled in recent years due to the end of the Cold War and the depletion of that resource. This shortage has been exaggerated recently by a jump in demand for neutron detectors for security which are able to detect the smuggling of plutonium and other nuclear weapons and are put in strategic ports and borders around the United States [2]. The demand for these detectors has risen in the past few years since the terrorist attacks in 2001. The Department of Homeland Security claims it will need 20,000 to 25,000 liters of ^{3}He gas yearly in order to supply its detectors around the nation [2].

Before 2009, the Department of Energy (DOE) was able to provide an annual supply of 60,000 liters of ^{3}He at atmosphere to consumers. In fiscal year 2009, the DOE supplied consumers with only 35,000 liters of the noble gas [3]. Scientists at Pacific Northwest and Oak Ridge national labs have estimated worldwide demand for the ^{3}He gas at 65,000 liters/year. They've estimated supply to be only 10,000 – 20,000 liters/year [2].

All of these factors have combined to make the price of the gas rise from $100-$200 per standard liter just two years ago to around $2,000 per liter in 2009 [2].

^{3}He is an isotope of He containing 2 protons and 1 neutron. The ground state nuclear wave function of ^{3}He is dominated by the S-state where the two protons are anti-aligned. This configuration, the S-state, contributes mostly neutron spin to the spin of the nucleus [1]. Because of its most probable configuration, ^{3}He is a widely used substance for studying the behavior of the neutron which is a difficult particle to study by itself because it is unstable and has a short lifetime.

Due to these properties of ^{3}He, Todd Averett and his polarized ^{3}He lab at The College of William and Mary along with researchers at the Thomas Jefferson National Accelerator (JLab) use the gas for electron scattering experiments at JLab to study the behavior of the neutron. The lab at

William and Mary is responsible for the filling of the target cells used for the scattering experiments at JLab. Due to a recent discovery that the spin exchange optical pumping using K and Rb alkali metals together polarize the ^{3}He better than just Rb, a change in the fill procedure has included the combination of the two alkali metals.

The polarized ^{3}He lab at The College of William and Mary currently has 20-25 liters of ^{3}He gas in cells that contain only the Rb alkali metal. These cells, because of the change in fill procedure to include the two alkali metals, have made the older cells outdated and not useful for future electron scattering experiments.

Because of the large increase in the price of the gas and the overall shortage of supply of the gas, it has become increasingly important financially and environmentally to create methods for recycling ^{3}He gas from the unused cells. At the current price for ^{3}He, this amount of gas in the lab at William and Mary alone is worth approximately $40,000 to $50,000 and a successful method for recovering the gas would also benefit a lab at the University of Virginia with the ability to recycle gas from this project.

II Theory

Figure 1 shows a picture of the typical target cell filled with ^{3}He for electron scattering experiments.

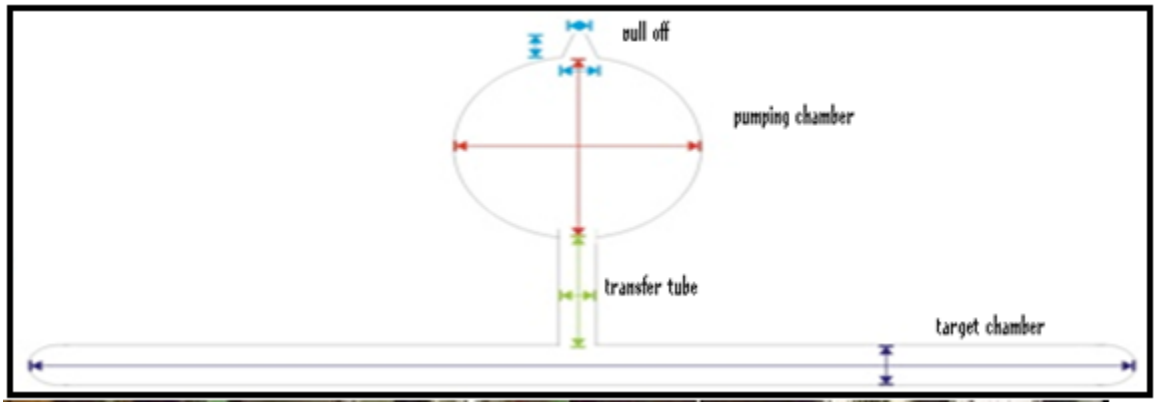

Figure 1: Visual of a target cell filled with ^{3}He. The pull-off is the upper-most piece of the cell (measured by blue arrows). The pumping chamber is next (measured by red arrows). The pumping chamber is attached to the transfer tube (green) which is then attached to the target chamber (purple) [6].

The ^{3}He recovered from the cells will ultimately be used in newer cells for electron scattering experiments that consist of mixtures of the alkali metals Rb and K.

For a gas relatively close to atmospheric pressure at room temperature, one can estimated that the gas is safely in the classical regime meaning that it is accurate to approximate the system with the statistical mechanical formula shown below. In this limit, its free energy can be written as:

$$F = N\tau[\ln\{N/(n_Q V)\}-1] \quad (2)$$

Using the relation:

$$P = -(dF/dV)_{\tau,N} \quad (3)$$

One can see that the pressure of an ideal gas is:

$$P = n\tau/V \quad (4)$$

where P is the pressure, V is the volume, n is the amount of gas, and τ is k_bT where k_b is the Boltzmann Constant (8.314472 J $K^{-1}mol^{-1}$) and T is the temperature (Kelvin) [5].

Solving for the pressure difference between the outside environment and the inside of the cell, it is possible to find the correct ratio of outside environment (considered to be water and oxygen) to ^{3}He inside the target cell. The two alkali metals Rb and K that are used for polarizing the ^{3}He inside of the new cells are very reactive with both oxygen and water. The oxygen corrodes the metals and the water can possibly release large enough amounts of energy to cause a fire. For this reason, it is ideal to keep the amount of oxygen and water to the 1 ppm range or lower [4].

The ideal gas law was applied to our system to ensure the 1ppm ratio of water and oxygen to ^{3}He gas in the chamber. The recovery system consists of an outside environment and the cell. Figure 2 shows a crude schematic of the system.

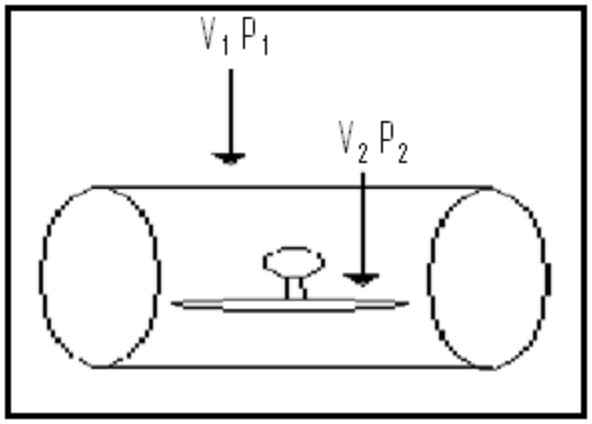

Figure 2: V_1 and P_1 correspond to the approximate volume and pressure of the system (chamber + after chamber) and V_2 and P_2 correspond to the volume and pressure of the target cell respectively. (Note: This is a crude picture of the system, used for estimation purposes only, a more in depth photograph can be seen in Figures 3 and 4)

To minimize the ratio of oxygen and water to ^{3}He, a vacuum system was used. Knowing the minimum allowed ratio, the ideal gas law was applied using the fact that the volume of the vacuum chamber, volume of the cell, and the cell's pressure are known (V_1 ~ 8,500 cm^3, V_2 ~ 200 cm^3, P_2 ~ 8 atm ~ 6080 Torr). Solving for P_1, the necessary pressure the chamber must reach to ensure a ratio of 1 ppm is:

$$P_1 = \frac{P2*V2*1E-6}{V1} \qquad (4)$$

Where, one can see the value of the variables in Figure 2.

Solving for P_1, one can see that a value on the order of 1E-4 Torr in the chamber would be sufficient in obtaining a ratio of 1ppm oxygen and water to ^{3}He gas. This can easily be reached using first a roughing pump, and then a turbo pump.

III Method

In keeping with the spirit of this project, it was attempted to supply as much material as possible used in the construction by recycled or green sources. A stand for the system was made from recycled pressed wood and two recycled hose clamps. Most other materials, such as the chamber, the turbomolecular and roughing pumps, and the holding tank were recycled from other projects.

The design of the recovery system consisted of three independent sections; the chamber, the vacuum pumps, and the storage system for the recovered ^{3}He. Each system was conceived, designed, built and tested separately as to ensure that the assembled system is capable of keeping the oxygen and water to 1ppm or less. It was important to test these systems separately to avoid difficulty in diagnosing problems later on. If each part of the system is tested separately and in sequence, one can easily fix each problem as it arises.

The chamber used is a high vacuum cylindrical chamber. As previously mentioned, a vacuum system consisting of a roughing pump and a turbo pump are used to supply the correct vacuum of 1E-4 Torr or less. The roughing pump is attached by a bypass to the chamber and is able to pump the system down to sufficient values for the turbo to be turned on (see Figure 3). A gauge capable of reading values as low as 1 mTorr (or 1E-3 Torr) can be seen on the left of the chamber in Figure 3 (blue gauge with red lettering).

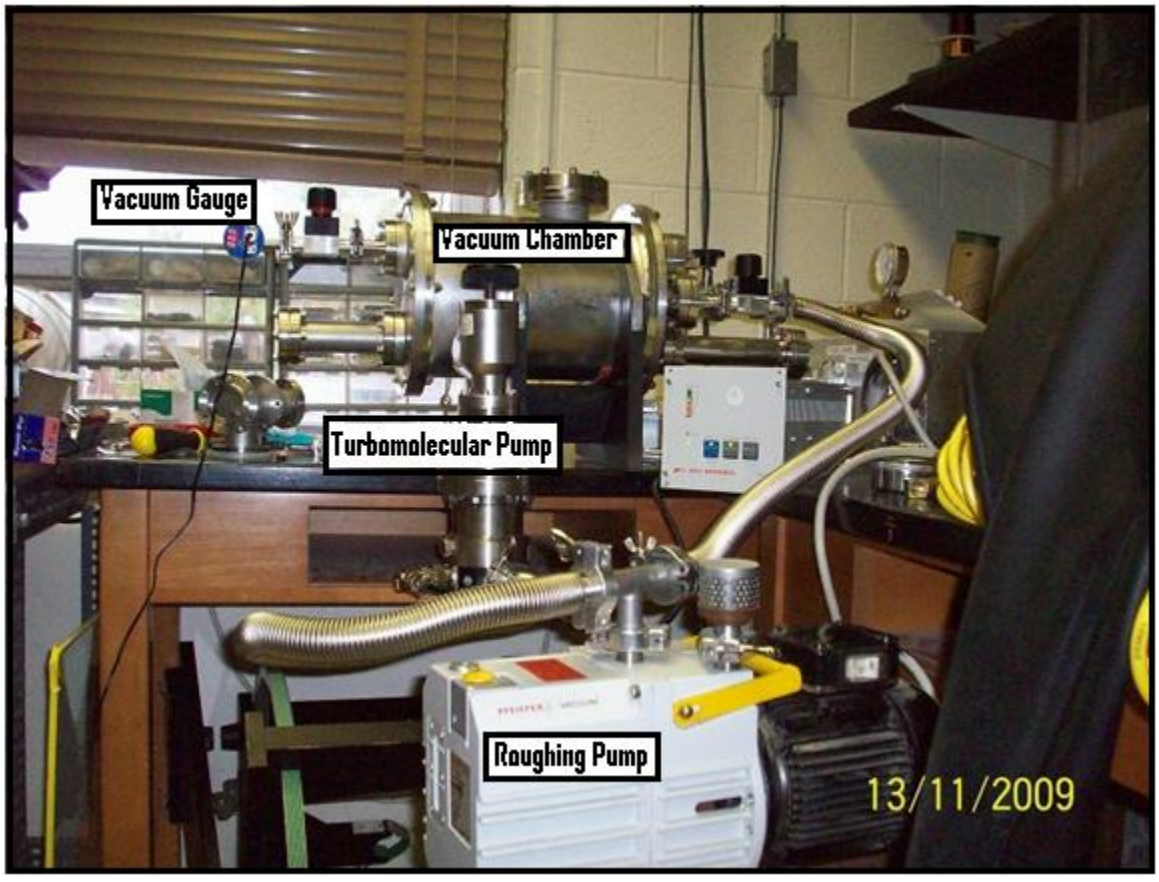

Figure 3: Full system assembled, without the tank for the storage of the ^{3}He.

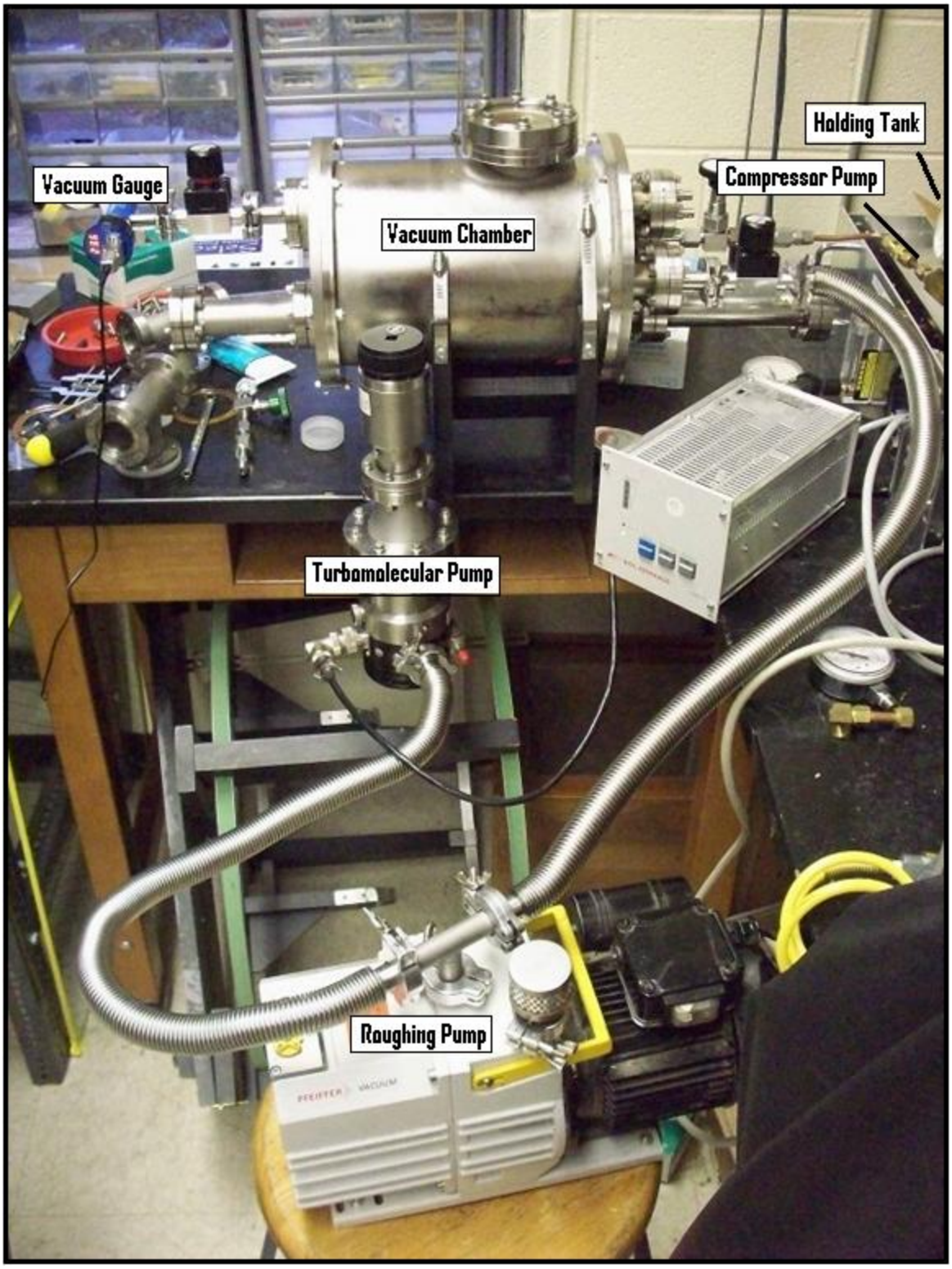

Figure 4: Full system assembled, top view.

The container for the recovered ^{3}He is attached to the right of the vacuum chamber. It consists of a valve to the chamber attached to a bypass valve and the pump that will pump the gas into the holding tank. To minimize leaks, the valve and the pump are attached to another valve which, in turn, is attached to the tank for holding. A schematic of this system is shown in Figure 5.

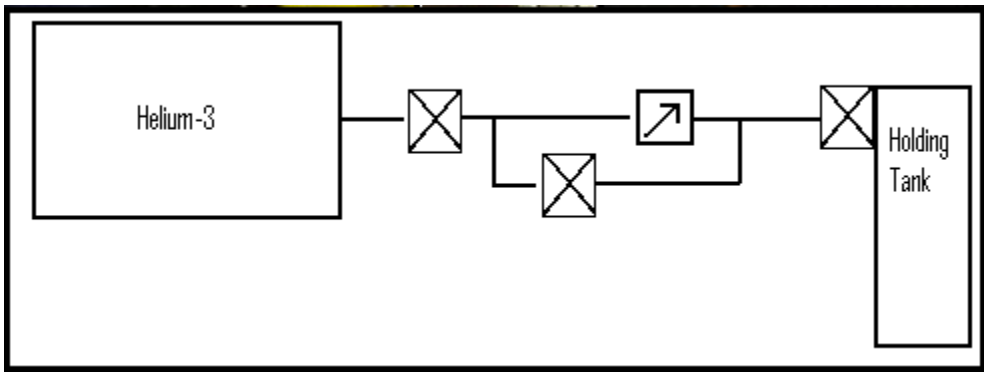

Figure 5: Schematic of chamber to holding tank for ^{3}He.

Below, in Figure 6, is shown an actual picture of the system.

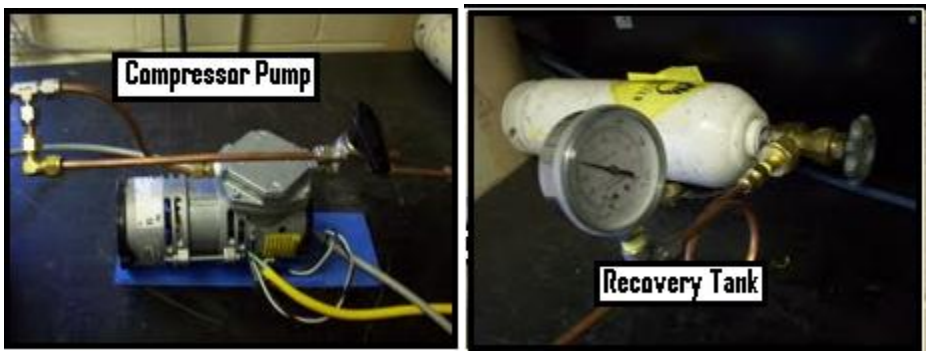

Figure 6: The "aft-chamber" where the pump transports the ^{3}He gas to the tank for holding. The tank and the pump were both recycled from an oxygen tank and a pump used for a previous project respectively.

To open the cell, made of GE-180 glass, high current resistance wire was used to attempt to melt the glass so that the target cells may be saved. The melting point of this glass is approximately 1000^{o}C, so the wire used is rated at 1100^{o}C when 3.75A are applied to it. The wire was wrapped repeatedly around the pull-off shown in Figure 1. To ensure insulation of the

wire to the surface of the cell, a small cap made of non-conducting insulating material was placed over the resistance wire which will be wrapped around the pull-off. A photo of melting a sample of the GE-180 outside of the vacuum chamber can be seen in Figure 7.

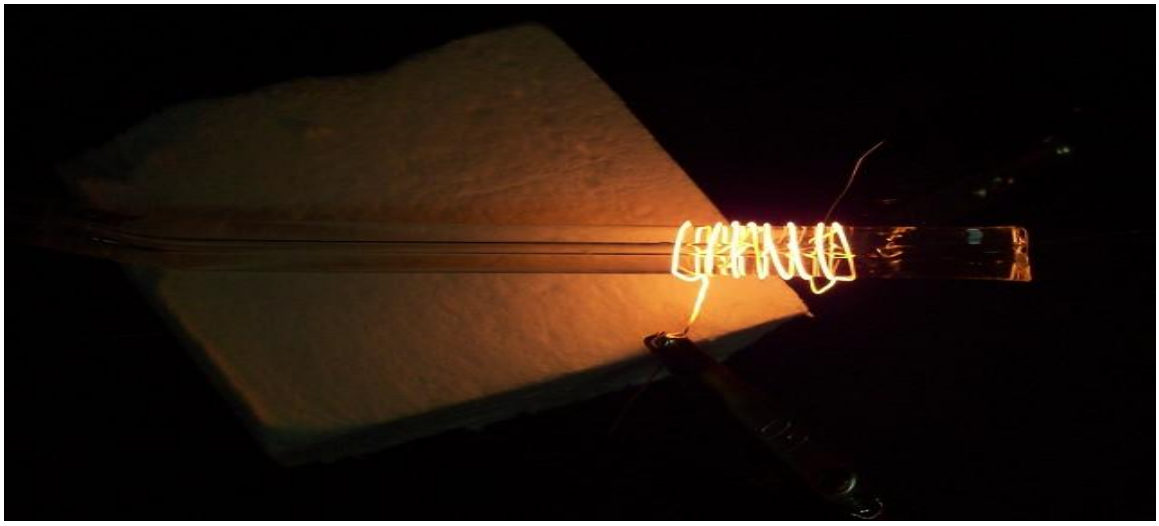

Figure 7: Approach to take for wrapping wire around pull-off. The more wraps, the hotter the glass will become.

To provide the current for the wire, a conflat BNC feed through was used (seen in Figure 8).

Figure 8: Right side of the chamber. Seen here are: at top of faceplate, the BNC feed through; on the left, valve which goes to the roughing pump; on the right, valve to ^{3}He holding tank; not seen, but on the bottom, nipple for the target chamber of the cell to rest in. On the top of the chamber is the viewport.

Once it was decided not to use the high resistance heating wire for cell bursting (discussion of this can be seen in the results section), it was decided to use a powerful (60 lbs of force) solenoid to attempt to break the cell off at the pull-off. A picture of the new method and its setup can be seen in Figure 9 below.

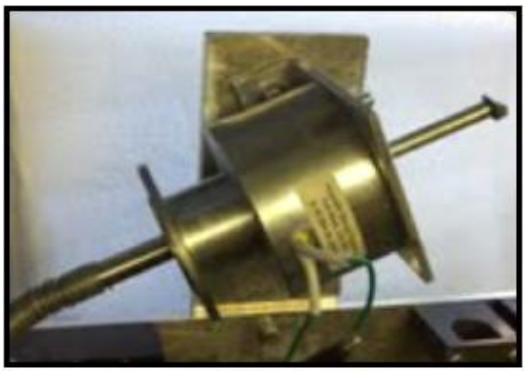

Figure 9: Solenoid used to break the cells.

To be able to adjust the solenoid in the north and south directions, two semi-circular shaped drilled adjustment orifices were installed allowing the solenoid to be angled appropriately for different sized cells.

One can also see in Figure 9 that a spring was attached to the solenoid and the chamber so that the solenoid could be activated without opening the chamber to reset it in the non-energized position.

IV Results

First tested was the vacuum limit of the entire system. This is important to know because of the previously mentioned limit of 1 ppm ratio of oxygen and water to ^{3}He. With the aft-chamber valve turned off, the roughing pump was able to get the chamber to 45.7mTorr. To test if the chamber had any leaks, the roughing pump was turned off over the weekend (68 hrs and 40 min) the chamber rose to 4.71 Torr in that time. This rise in vacuum was expected for a system of this type. But, to be sure that there was no significant leaks, the chamber was backfilled with helium to create a positive pressure variance between the inside of that chamber and the outside to be tested with snoop (a liquid that bubbles up when a leak is present. No leak was found in the system.

The turbomolecular pump was then turned on and the chamber reached 0.0mTorr (the limit of the gauge). The vacuum rose from 0.0mTorr to 8 mTorr at a rate of 0.1 mTorr/sec. This range allows the system to get in the range necessary to keep the ^{3}He to 1ppm or below.

Upon testing the resistance heating wire outside of the chamber, we were able to melt the GE-180 glass using insulation.

The method of melting the cell was not feasible because of the amount of out-gassing. With three resistance heating wires braided (so the amount of current applied to the wire could be higher), a current of 2.5 Amps was applied to the wire. The vacuum reading, with all valves closed and pumps off, went from 0.0mTorr to 28mTorr once the current was turned on. This increase in vacuum with outgassing from the wire immediately takes the chamber out of the region where the ^{3}He would be below 1ppm.

The wire also did not melt the GE-180 glass well enough for us to be confident in its ability to melt a cell enough to release the ^{3}He gas. With a current of 4 Amps applied to the resistance wire, the GE-180 glass did not melt at all and the vacuum quickly rose to 100 mTorr. Upon further testing, the GE-180 glass did not melt inside the chamber under vacuum with an applied current of 10.37 Amps.

While waiting for the solenoid to arrive, the aft-chamber was tested. We evacuated the chamber and the storage bottle. We then filled the chamber with 720Torr of Nitrogen. Turning the recovery pump on, the chamber vacuum reading went to 312Torr and equalized while the gauge to the container reads 17 psi (~880 Torr). This shows that the pump is pumping some ^{3}He into the holding tank, but not nearly a satisfactory amount of the gas is being recycled.

Using the solenoid, we were able to burst William and Mary cell #2. Seen in Figure 10 is the burst pumping chamber.

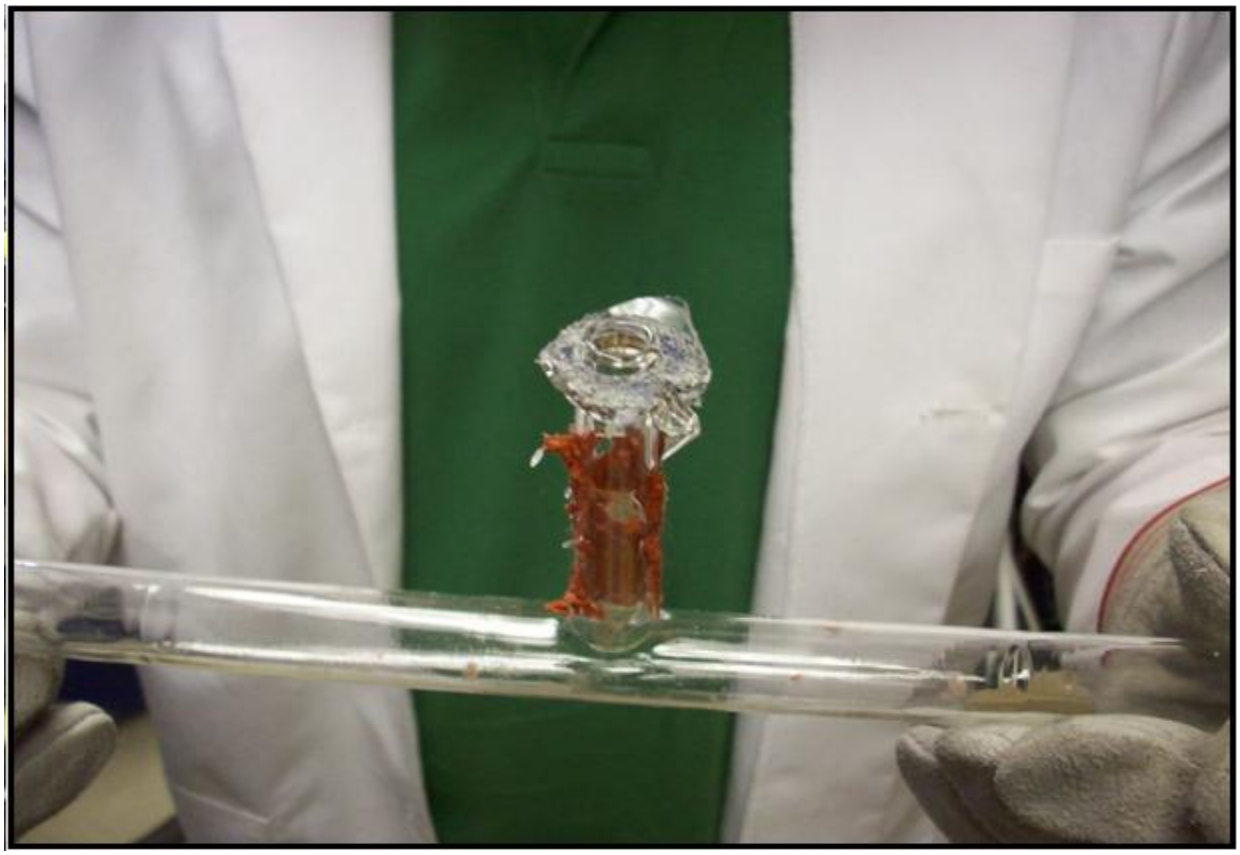

Figure 10: William and Mary Cell # 2. The pumping chamber exploded during solenoid impact.

With the vacuum rising at a rate of 0.16 mTorr/sec, we were able to break a cell applying 35 volts and 10 Amps to the solenoid. As one can see in the above picture, the pumping chamber burst. The vacuum gauge is not meant to be used with ^{3}He and the initial shock of the cell blowing up partnered with this fact caused the gauge to become not calibrated correctly. And, in effect, we were not able to be confident in the purity of the ^{3}He in the tank, nor were we able to measure the amount recycled.

After William and Mary Cell #2, we were successfully able to burst William and Mary Cell #3 (A picture of the burst cell can be seen in Figure 11). This cell exploded, leaving no part of the target cell intact to be able to recycle. Bits of glass were scattered everywhere. We were able to pump an undetermined amount of ^{3}He into the holding tank.

Figure 11: William and Mary Cell # 3 after cell burst and ^{3}He recovery.

V Discussion

Because the gauge is not meant to measure He and the initial shock of the explosion uncalibrated it, it is difficult to know how much ^{3}He was recovered from the first and second cell bursts. On the second cell burst attempt, a valve was put between the chamber and the vacuum gauge. This will alleviate the effects of the initial shock from an explosion inside the chamber to the gauge. But, even in guarding the gauge from the initial shock, the gauge was still unable to accurately read the vacuum as it wasn't meant to measure ^{3}He.

Another problem was the amount of glass shattering everywhere in the chamber. If glass were to drop down into the turbo pump, the pump would be ruined. To avoid this, a mesh screen was glued to the conflat copper piece between the turbo pump and the chamber. This mesh screen (shown below in Figure 12) will block glass from future explosions.

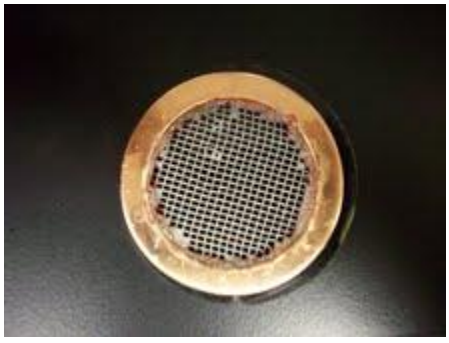

Figure 12: Mesh attached to conflat copper to avoid glass shards getting into the turbo pump.

Another problem with the current system is the power of the pump. The pump seen in Figure 6 is rated at a maximum compression of 50 psi (the pressure that the holding tank reaches is limited by this relatively small value, so the amount of ^{3}He gas capable of being put in the holding tank is directly dependent on this value). From the results of the nitrogen fill, one can see that the 312 Torr the system gets down to after the pump is turned on could be drastically improved saving a significant amount more ^{3}He.

One way to improve this setup where it would allow the experiment to recycle more ^{3}He would be to have a cooler filler with liquid nitrogen capable of holding the holding tank. This would cool the tank to temperatures near 77K. Using the ideal gas law (Equation 4), and assuming the

rest of the system is kept at room temperature (300K), the pressure in the tank would decrease by approximately four times. This would allow about four times more ^{3}He gas to be pumped into the tank than before with a tank at 300K. A schematic of this setup can be seen in Figure 13.

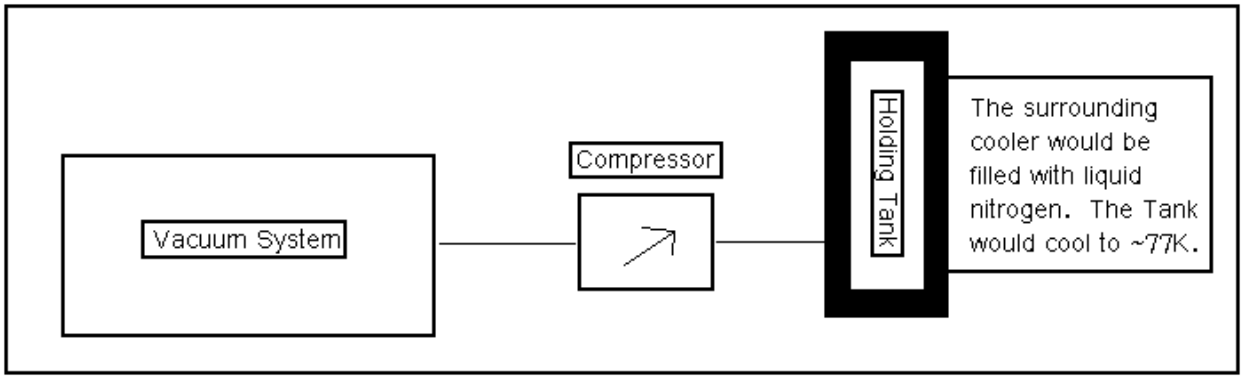

Figure 13: Schematic of the system after improvements have been made. Ideally, the compressor pump would be replaced for a more powerful model.

To measure the purity of the recovered ^{3}He, in the future, the lab will use the residual gas analyzer already located in the lab to be sure that the recovered gas is pure enough to be reused (1 ppm or less outside gases to ^{3}He).

The ability to recover ^{3}He gas from unused target cells has the potential to save the lab thousands of dollars due to recent supply shortages of ^{3}He. The system is functional now, but it is not recovering the desired amount of ^{3}He.

References

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