

Hybrid Alkali Production for Polarized ^3He Target Cells

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Abstract

Glass cells filled with polarized ^3He are used as targets in nuclear physics experiments. The ^3He is polarized using a Spin Exchange Optical Pumping technique (SEOP) using a hybrid alkali mixture of potassium and rubidium inside the cell. Nuclear experiments using these cells benefit from high polarization levels in part due to clean alkali mixtures of certain ratios. A glove box (with a control and filtration unit) was designed and constructed to reduce contaminants, control the pressure, and provide a clean and inert environment for creating usable and precise hybrid alkali mixtures.

1 Introduction

Polarized ^3He target cells (Figure 1) are produced in the William and Mary Polarized ^3He Target Lab (Pol ^3He). Completed target cells are taken to Thomas Jefferson National Accelerator Facility in Newport News, VA to be used as targets for electron beam collision experiments.

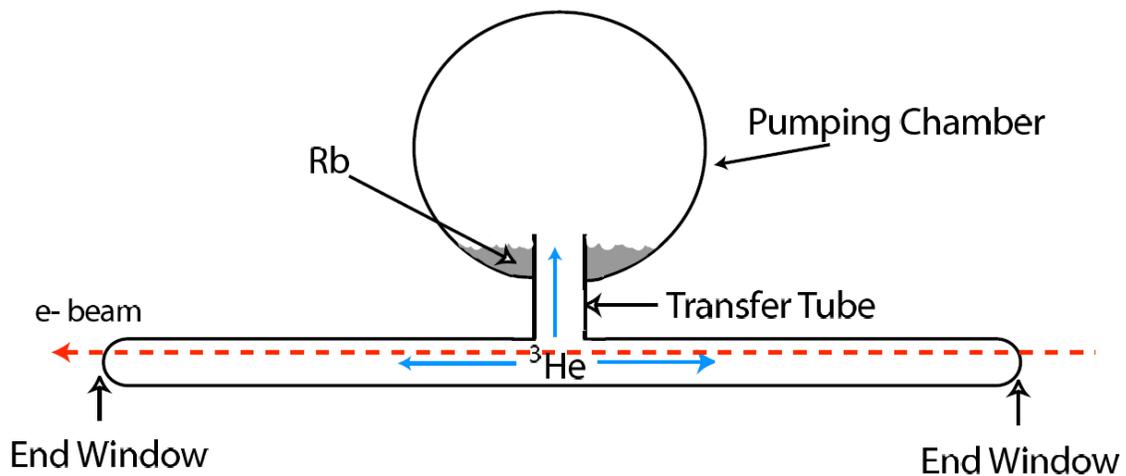


Figure 1: Schematic diagram of a Target Cell ([3] Buckley)

The ^3He nucleus consists of two protons and one neutron. Due to the anti-alignment of the proton's spin, the nucleus is left with an effective spin of the neutron, allowing for study of the structure of the neutron. Aligning all of the neutron spins of the ^3He nuclei results in a gas of polarized ^3He that simplifies these collision experiments. The goal of the William and Mary Pol ^3He lab is to create target cells with the highest possible levels of polarization. Factors such as the ratio of alkali metals within the cell as well as lack of contaminants affect the maximum attainable polarization. The role of these factors will be more evident after the production and polarization process is described.

First, glass cells with attachments for filling are blown in Princeton, NJ and are shipped to the lab. They are cleaned and evacuated through a baking process lasting several days and finally filled with ^3He and a small amount of a mixture of rubidium and potassium necessary for the polarization process is deposited within them. The cells are then sealed, completing the production of a target cell.

The cells are then polarized using a process called Spin Exchange Optical Pumping (SEOP). This can be broken down into two stages, the first of which is optical pumping. As stated before, a small amount of Rubidium is present within the cell. The cell is placed into a setup that includes two large Helmholtz coils (figure 3) to produce a magnetic field as well as an oven to heat the cell and vaporize the alkali. Optical pumping takes advantage of the Zeeman effect where energy levels of the $+1/2$ and $-1/2$ spin states split in atoms in the presence of a magnetic field [3]. With Rubidium, the presence of a magnetic field splits the 5p and 5s energy levels (where the outer electron will be) into $m = 1/2$ and $m = -1/2$ [3]. If the outer electron is not excited it will most likely be in the lowest of the split 5s levels. This electron is then optically pumped using a laser tuned to the exact energy necessary to bump it up to the higher of the two split 5p states [3]. Electrons are occasionally excited to other energy levels but will decay back down to the lower 5s state to be pumped up to the higher of the 5p states [3]. This ensures that a much larger than naturally occurring portion of the Rubidium outer electrons will be in the same spin state. The Rubidium then transfers this spin to the nucleus of the ^3He atom through collisions. An intermediary step has been recently added which requires the Rubidium to spin exchange with potassium atoms and then potassium to spin exchange with the ^3He . Cells polarized using this newer process are referred to as Hybrid Alkali cells and will be discussed at greater length later. The process of SEOP polarization takes several hours to polarize a cell and the polarization is only temporary. Once the laser is no longer optically pumping the Rubidium, the polarization slowly decays through internal collisions with the cell walls and the alkali metals. Therefore the polarization must be characterized while the cell is in a polarized state and hence still being pumped.

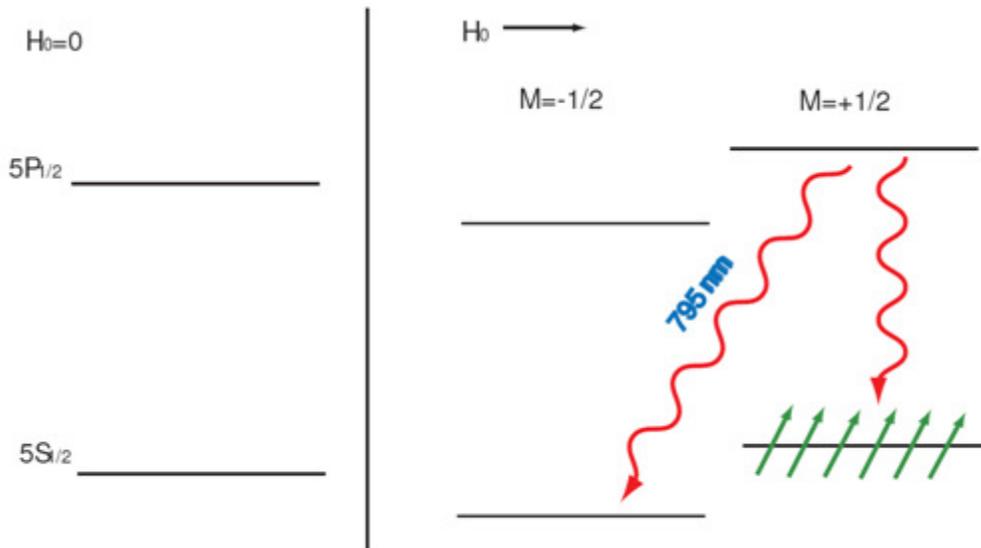


Figure 2: Zeeman Splitting of the 5p and 5s states of the rubidium outer shell ([3] Buckley)

Several tools exist to determine relative and exact levels of polarization within the cell. Nuclear Magnetic Resonance (NMR) is an older but still useful method of producing a relative measure of the amount of ^3He polarization within the cell. NMR requires another set of coils to produce an RF field tuned to the Larmor frequency for helium, inducing the ^3He nuclei to change states [3]. The movement of the magnetic moments of the collection of polarized atoms induces a voltage that can be picked up in a secondary set of coils. The magnitude of the induced voltage is directly proportional to the amount of polarized ^3He within the target cell [3]. The main disadvantage of this method is that in order to produce an exact calculation of the polarization, the system must be calibrated by measuring the induced voltage of an identical cell filled with water to acquire constants of proportionality. The same NMR process with water however results in a large amount of error so the NMR technique is used mostly as a tool to compare polarization levels between similar target cells [3].

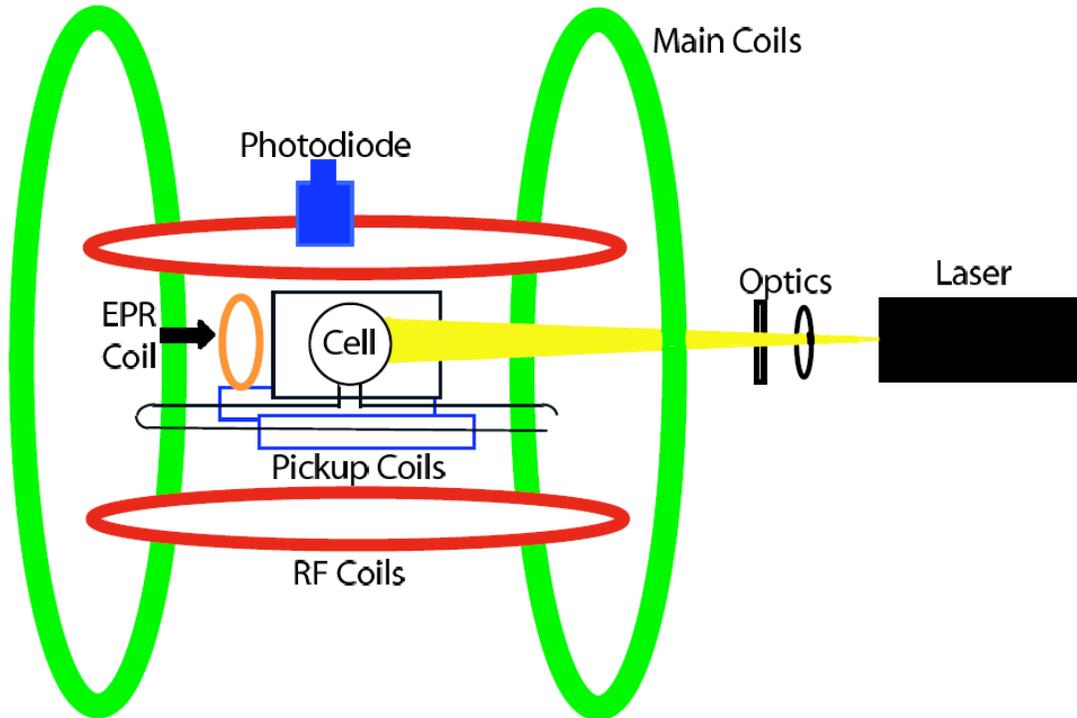


Figure 3: Cell and Laser setup for polarization, NMR and EPR ([3] Buckley)

A newer method of determining the polarization of a target cell is Electron Paramagnetic Resonance or EPR. This process once again takes advantage of the Zeeman splitting that occurs in rubidium in the presence of a magnetic field. The rubidium splitting is induced by the magnetic field applied by the coils but a further splitting is due to the magnetic field applied by the polarized ^3He [3]. The shift from the 795nm resonance of rubidium is directly proportional to the amount of polarization within the target cell [3]. These methods are used to characterize cells and thus find methods of creating cells with high levels of polarization that hold the polarization longer.

2 Theory

As mentioned above, a more recent development in the production of ^3He target cells is the hybrid alkali mixture. Up until a few years ago, maximum ^3He polarization was around 30 to 40% but with the inclusion of potassium (in addition to the rubidium) polarizations of up to 60% are regularly observed and newer lasers have pushed this to as high as 70%. This increase is due to the spin exchange rates of the alkali metals used in polarization. The rate at which a target cell is polarized in part determines the maximum polarization of a cell. The target cell has a variety of imperfections and naturally loses polarization, a process called polarization relaxation [2]. If the rate at which a cell is polarized is increased to overcome the rate of relaxation, higher polarizations can be achieved. Optical pumping polarizes the outer shell electron of the alkali metal that the laser is tuned to very efficiently and thus very quickly. The holdup in the process occurs

during the spin exchange of the rubidium with the ^3He . It has been found that the spin exchange efficiency and speed of potassium to ^3He is almost ten times that of rubidium to ^3He [1]. Lasers are affordable and available only in the range of the rubidium absorption so it is necessary to include both the rubidium and the potassium. This hybrid alkali SEOP process now involves a laser optically pumping the rubidium, the rubidium spin exchanging with the potassium and then the potassium spin exchanging with the ^3He . The spin exchange rate between the rubidium and potassium involves only an outer electron spin exchanging with another outer electron, occurs quickly, and hence the time is negligible compared to the much less efficient spin exchange between the outer shell electron of the alkali and the ^3He nucleus [1].

The ratio of rubidium to potassium within the cell also affects the efficiency and the maximum achievable polarization [1]. In addition, once ratios are properly adjusted, the final alkali ratios may end up different within the cell due to differences in vaporization temperatures between rubidium and potassium and the fact that they are heated when they are deposited into the cell. For these reasons it is paramount to be able to mix the two alkali metals in a clean environment so as to be able to create a vital constituent for the target cells and also to allow for experimentation with alkali ratio's for efficiency of polarization. Figures (4) and (5) show how the efficiencies and maximum polarization are affected by different potassium and rubidium ratios. The photon efficiency η_λ is the number of nuclei polarized per photon absorbed into the alkali vapor [1]. This is effectively a measure of the efficiency of the entire SEOP process. The spin exchange efficiency η_{SE} , is the ratio of the rate at which angular momentum is transferred to the ^3He nuclei to the rate at which alkali polarization is lost through other processes [1]. This is a measure of just the alkali-helium spin exchange and does not take into account the optical pumping necessary to polarize the rubidium. In figure (4) it is evident that the spin-exchange efficiencies increase as the potassium-to-rubidium ratio ($[\text{K}]/[\text{Rb}]$) increases, however the photon efficiency reaches a peak at a ratio slightly less than 40. The spin-exchange efficiency only takes into account the Rb-K- ^3He collisions and not the photon absorption and as stated above the spin-exchange efficiency for potassium is much greater than for rubidium. This upward trend in the spin-exchange efficiency for Rb-K- ^3He represents the fact that more potassium is present and allowing more ^3He to be polarized through more efficient K- ^3He collisions. The fact that the photon efficiency reaches a peak and then falls with increase in $[\text{K}]/[\text{Rb}]$ ratio can be explained by the trend in figure (5). As the $[\text{K}]/[\text{Rb}]$ ratio increases, less rubidium is present to be optically pumped. It is only the rubidium and not the potassium that is optically pumped so as the ratio increases, the maximum alkali polarization decreases as there are less polarized Rb atoms to spin-exchange with potassium. These two competing factors require the alkali ratio to be properly chosen in order to achieve the maximum ^3He polarization within the cells.

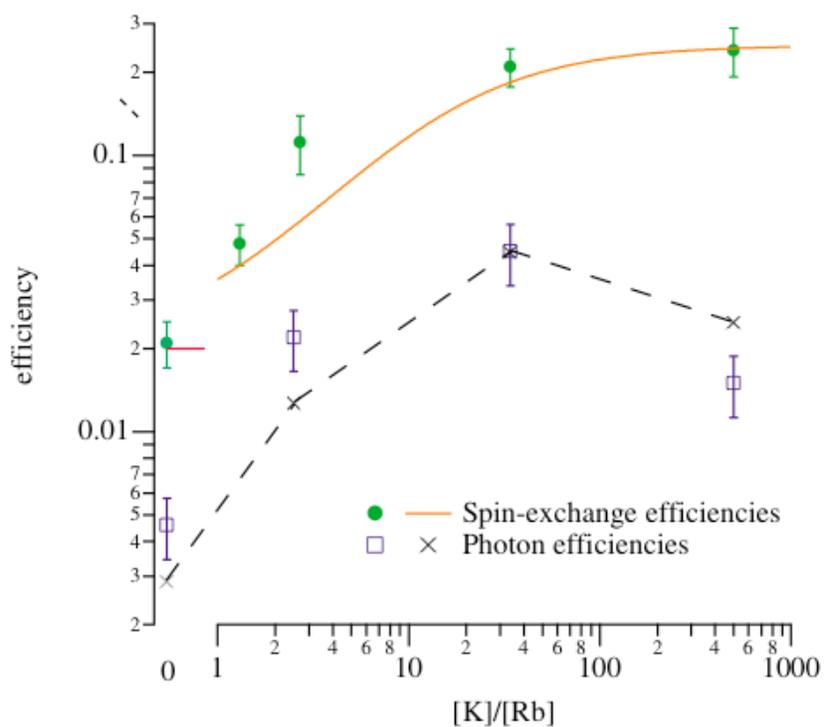


Figure 4: Spin exchange and photon efficiency vs. potassium to rubidium ratio ([1] Babcock)

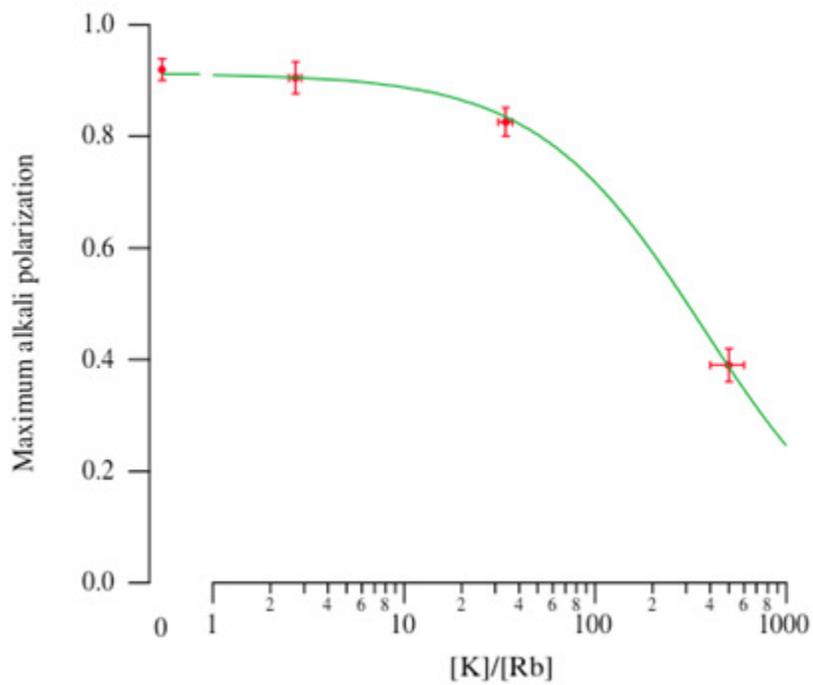


Figure 5: Maximum alkali polarization vs. potassium to rubidium ratio ([1] Babcock)

Rubidium and potassium are both very reactive alkali metals. Oxygen causes corrosion making them useless and reactions with water release large amounts of energy potentially starting fires. For this reason it is important to be able to mix rubidium and potassium to create the proper ratios in an inert and clean environment. A nitrogen glove box that filters out oxygen and water is the solution to this problem that was pursued. Ultimately the environment must be within the 1-5ppm range of oxygen and water but preferably lower. Oxygen sensors in the part-per-million (ppm) range are very expensive so a simple and cost effective method using light bulb lifetimes was used to determine the oxygen and water content in the glove box.

In an inert gas such as helium, filament lifetimes have found to be predictive of oxygen and water contaminant levels [4] and can be calculated by $\tau_x = AC_x^{-\gamma}$, where t_x is the filament lifetime in seconds, C_x is the contaminant level in ppm, and A and γ are constants. For multiple contaminants the lifetimes compound as follows: $1/\tau_{\text{overall}} = 1/\tau_{\text{O}_2} + 1/\tau_{\text{H}_2\text{O}}$. For $x = \text{O}_2$, $A = 3.0 \times 10^{4 \pm 0.09}$ and $\gamma = 0.80 \pm 0.03$, and for $x = \text{H}_2\text{O}$, $A = 2.0 \times 10^{3 \pm 0.14}$ and $\gamma = 0.78 \pm 0.11$. Figure (6) (from [4] Mao) shows the data taken using an argon environment and several different types of light bulb filaments. Despite small differences in between different filaments, the lifetimes still follow a power-law relationship.

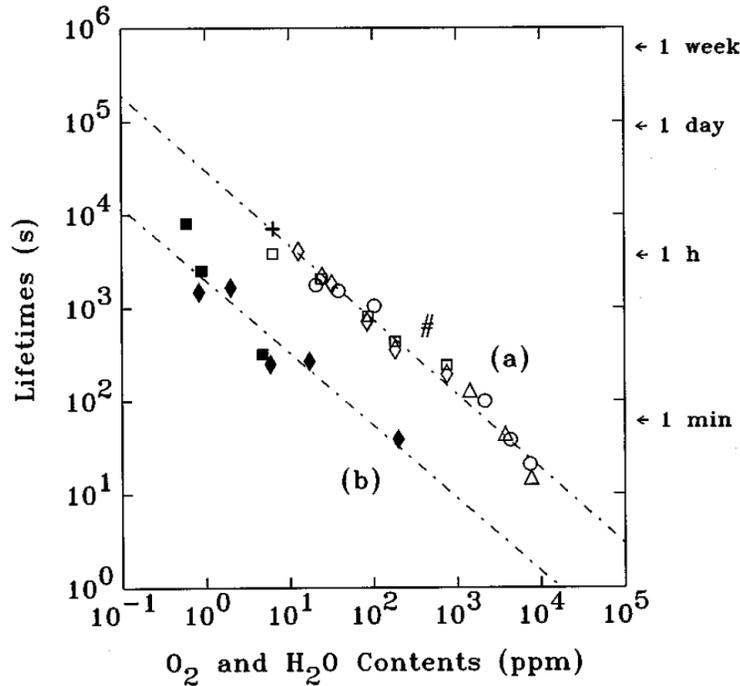


Figure 6: Filament lifetime in an inert gas with H₂O and O₂ contaminants. Water contaminant levels appear in (b) and oxygen in (a) ([4] Mao)

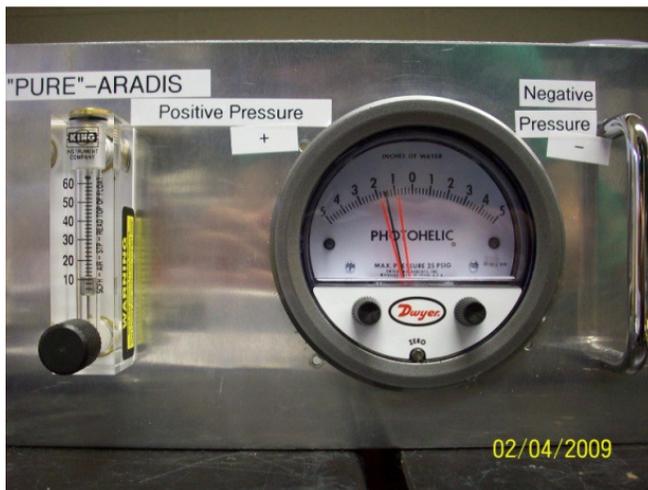
3 Method

The design and construction of the alkali mixing glove box system involved two main components: the glove box itself and an external control and filtration unit. The control and filtration unit was constructed first. The requirements of this unit were that it must be able to filter nitrogen within the box to a level of less than 5ppm of oxygen and

other contaminants as well as maintain positive glove box pressure despite possible leaks and movement of gloves in and out of the glove wells. The filtration requirement was fulfilled with an air purification system utilizing a Matheson Tri-Gas PUR-Gas filtration system. This consists of a baseplate and two filter cartridges, designed for Liquid Chromatography (LC), that filter out moisture and oxygen which contaminate the box for alkali mixing processes. A 1/16 horsepower compressor/vacuum pump was acquired to flow nitrogen from inside the glove box into the filtration system. After flowing through the two LC filters the nitrogen flows into a (King 2-60 standard cubic feet per hour) flow meter with an adjustable flow control. This allows us to view and control the flow rate through the air filtration and back into the glove box. The process of creating a clean glove box involves the flowing of nitrogen straight from a dewar for an initial purge lasting several hours followed by a continuous filtering of the nitrogen within the glove box for several days to remove contaminants to an acceptable level.



Figures 7: Filtration Pump



Figures 8: Flow meter and differential pressure gauge (right)

The second requirement was fulfilled using a (Dwyer A3000 Photohelic) differential pressure gauge. This gauge compares and displays the pressure difference between atmospheric pressure and a pressure sensing line. It has two internal relays that can be programmed through wire connections to activate under certain conditions in reference to upper and lower pressure set points. These relays were then attached to solenoid valves to eventually be connected to the glove box. The solenoid valves were connected and the differential pressure gauge was programmed (through jumper connections) such that when the pressure drops below a low pressure set point, the valve connected to the nitrogen source opens allowing nitrogen to flow into the glove box while the other valve remains closed. Additionally, when the pressure rises above a high pressure set point, the exhaust valve opens allowing pressure release. With the set points chosen properly, this guarantees that the pressure will equalize between the two set points in the event of any leaks or pressure disturbances. This is especially vital because the glove box gloves are continually entering and exiting the glove box and hence change the pressure. If the pressure within the box is not higher than the atmospheric pressure in which the glove box is operating then any small leaks will leak contaminants into the box rather than clean nitrogen out.



Figures 9: Solenoid exhaust valve with relay connections, compression fit plastic tubing from box and atmospheric opening

The glove box used was a 76.2 x 61 x 61cm (Sidentry) glove box. The glove box must fulfill two main requirements. First it must be able to flow gas for filtration and feed electricity to power the supplies needed for the glove box operation, and second, it must be able to stay pressurized and not leak any gas in or out. A series of holes were drilled in the glove box to allow for all the necessary connections into and out of the box. A strip of acrylic was first attached with acrylic cement to one side with of the box to provide support for the drilling process. A total of seven holes were drilled with special acrylic drill bits sensitive to the conditions of the acrylic. Six of the holes were for ¼”

compression fitting feed-throughs for gas entry and exit and a seventh hole was drilled for a 1/2" feed-through for the electricity entry. Soap water was applied to the holes as they were drilled to lubricate and heat sink the drilling process. Holes were drilled very slowly and carefully because high drill speeds heat and melt the acrylic causing it to grab the drill bit and shatter the surface. Acrylic cementing and drilling was practiced at great length on test strips to find acceptable techniques and speeds so as not to damage the delicate glove box.

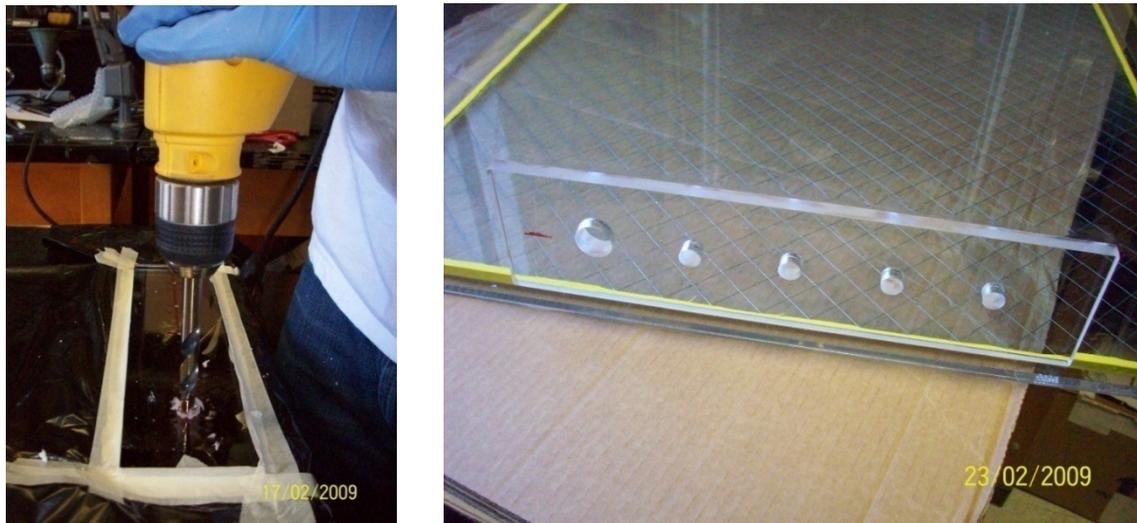


Figure 10 and 11: Acrylic drilling (Left) and five completed holes along with glass base with protected edges (right)

The above mentioned feed-throughs were mounted and sealed with silicon aquarium sealant. The plug of a power strip was cut and the wire was fed through the 1/2" feed-through. A new plug was then attached and the feed-through was sealed off with the silicon sealant. The remaining six 1/4" feed-throughs were connected using 1/4" plastic tubing and compression fitting to the following components: the solenoid valve for atmospheric exhaust, the solenoid valve connected to the nitrogen source, the differential pressure gauge pressure sensor, the pump intake, the filtration system intake, and finally a hand-turned valve open to atmosphere. Additionally, the pump output was connected with the same plastic tubing and compression fitting to the filtration system intake. The gloves were then attached using circular clamps that screw tighten. It was found that these did not create a perfect seal so the silicon sealant was used to seal the crack where the gloves and glove box meet. Two small holes found in the glove box door's foam seal were also filled with silicon to prevent gas leaks. Pressure was applied to the inside of the box and Snoop liquid leak detector was applied to find all possible leaks. Any leaks that were found were cleaned and resealed with silicon sealant. A schematic of the connected system can be seen in figure (13);

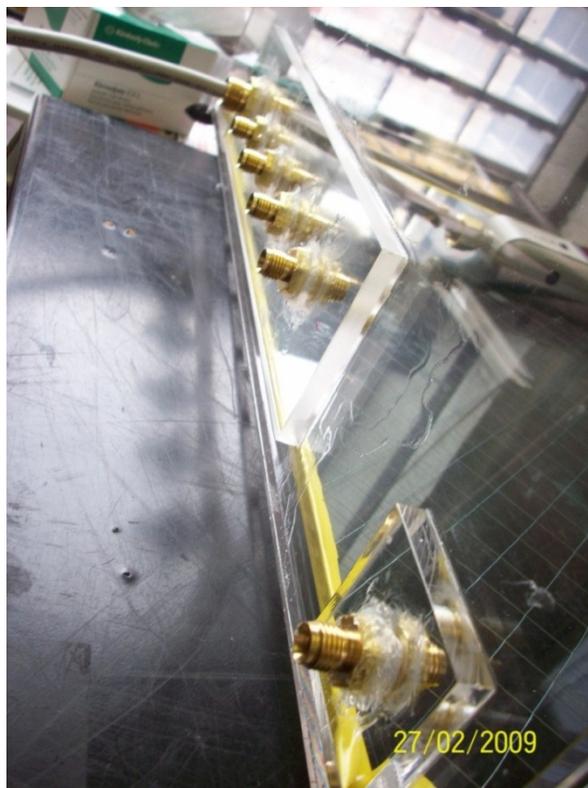


Figure 12: Silicon sealed feed-throughs on glovebox

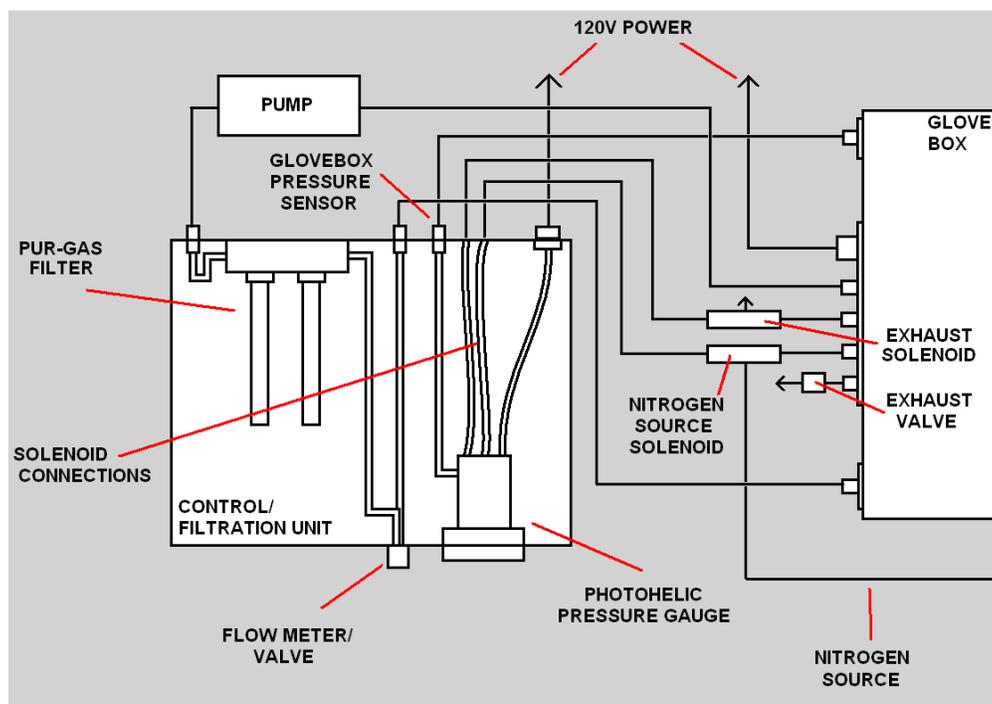


Figure 13: Control/filtration unit and glovebox schematic with connections

A procedure for creating a clean glove box atmosphere was created and is as follows. The filtration system must be turned off and the external exhaust valve opened. Nitrogen must be flowed into the box from a dewar or tank for several hours in an effort to cycle approximately 10 volumes of nitrogen through the glove box. The time this takes depends on the flow rate into the box. The exhaust valve must then be closed and the control unit must be turned on. The differential pressure gauge's set points must be set slightly above and below a positive pressure. The gauge goes from positive to negative 5 inches H₂O. Too high of pressure can result in a tear in the gloves or breaking of any of the other seals but 5 inches H₂O is less than 0.2 psi so any positive pressure set point should not be above the .25 psi recommended maximum pressure for the glove box. The glove box must then be allowed to equalize to within this range at which point the pump must be turned on. The filtration system intake and exhaust valves must be opened slowly until they reach the desired flow rate. The filtration system must then be allowed to circulate for several days until the glove box has achieved the desired level of contaminants. For alkali mixing this should be <1ppm oxygen and water.



Figure 14: Constructed lightbulb holder with light bulb without glass bulb for the light bulb test

An alkali mixing process was also designed to be implemented in the Pol³He lab. A trip was made to a lab performing the same alkali mixture process at the University of Virginia to observe their setup and method. The two alkali metals must be melted, weighed and mixed in the proper ratio, and then resealed within the box. For this process a (Fulcrum Torbal AD50) scale with precision to .001g, a (Corning Scholar 170) hot plate, (Kaplan) heater tape, (Pasteur) pipettes, 10mL ampoules, 1mm glass rods, ring stands, and several variable power supplies have been purchased and collected. After the gloves have been placed on, an ampoule that will be used to mix the alkalis is weighed

and the scale is zeroed to that mass. Kaplan heater tape is to be used to melt an ampoule of potassium to be poured into the final ampoule which is also heated with heater tape to ensure that no metal cools and accumulates on the walls. A syringe is to be used to force the rubidium out of its initial ampoule. Bent Pasteur pipettes will be used to allow air to escape from the final ampoule ensuring that the alkali makes its way and settles at the bottom of the ampoule. The ampoule is then weighed to find the mass of the potassium within it and the necessary amount of rubidium is calculated. An ampoule of rubidium is then melted with more heater tape and rubidium is tapped out onto a Petri dish. After the rubidium has cooled slightly, small amounts are added and the final ampoule is weighed until the desired combined alkali mass is achieved. A small rubber stopper will then be placed inside the ampoule so that the alkali is protected when it is removed from the glove box. Outside of the glove box, the rubber stopper is replaced with a rubber stopper attached to a line with argon. The ampoules are then heated with a torch and the end is pulled and melted to create a glass seal to ensure no leaking in of atmospheric contaminants. The ampoule must be removed and sealed by melting the ampoules outside the glove box because no flame can be used inside the box and the rubber stoppers only function as temporary seals. This process will be further developed and adapted when the glove box is fully functioning.

4 Results

The control and filtration unit and the glove box were completed and investigated by testing one function at a time. First, the differential pressure system was tested by closing the glove box door, the exhaust valve as well as the valves for the filtration system intake and exhaust valves. The nitrogen source was attached via compression fit plastic tubing and the pressure gauge set points were set slightly above and below positive 2 in. H₂O. When the unit's power was turned on, the box filled and the gloves bulged out until the high set point was reached. Small leaks in the glove box resulted in the internal pressure slowly dropping below the low setpoint at which point the box began to fill again until it reached the high point. This indicates correct and expected functionality out of the differential pressure system. To test the filtration system, we turned on the pump and slowly opened both the intake and exhaust valves on the filtration system. A flow of about 12 SCFH was observed, well within the operating specifications of the motor and the filtration system.

The entire glove box system was then tested to see if it was capable of creating the oxygen and contaminant-free environment necessary to expose alkali metals without reaction. To test this, the above procedure for creating a clean glove box atmosphere was followed using a tank of welding nitrogen already available in the lab. This was only for a test and an actual dewar of liquid nitrogen will be used in the final setup. The box was allowed to filter for 24 hours when the first light bulb test was performed. Compared to a lifetime of one second outside the glove box, the light bulb stayed lit for approximately 30 seconds before burning up. This showed a massive improvement in the oxygen levels in the air but indicated that much more filtration would be necessary to reach the desired levels. Light bulb lifetimes in the 10⁴ to 10⁵ ranges are necessary to indicate desired containment levels. After this initial light bulb test, we allowed the box to filter for

approximately 96 hours and proceeded with an additional light bulb test. This time, the bulb lasted 1 minute and 13 seconds showing an additional improvement but still nowhere near the time indicative of a clean atmosphere. Performing the light bulb test requires the operator to place his or her hands into the gloves to turn on the light bulb. Upon removing the hands from the gloves, the pressure control unit will fill the glove box with nitrogen directly from the tank or dewar. It was observed during the light bulb test that when this filling occurred, the light bulb produced smoke indicating higher levels of oxygen in the nitrogen from the tank. This was cause for concern considering that the tank nitrogen alone was rated at oxygen levels around what is required for alkali production. A few modifications were made such as widening the exhaust solenoid valve to allow for quicker pressure release and the pump was moved inside the glove box to prevent possible contamination. A new set of bulbs was placed inside the box and the box was once again purged and filtered for about 60 hours. The light bulb test was repeated and burned for 1 minute and 15 seconds still indicating some problem filtration system or the nitrogen supply. The nitrogen supply tank was replaced with a 240L dewar of liquid nitrogen which provided the nitrogen gas supply via liquid nitrogen boil off. After 26 hours of purging and no filtration we performed a light bulb test and observed a lifetime of 495 seconds indicating 100-300 ppm oxygen or 3-15 ppm of water. We have no way of determining the ratio of oxygen to water but just maximum concentration of each contaminant assuming the absence of the other. After 1 hour of filtration the lifetime fell to 250 seconds indicating 250-650 ppm oxygen or 7-36 ppm of water and finally after a week of filtration we observed 95 seconds. This downward trend in filtration lifetime is a serious cause for concern since it is not only indicating that instead of purifying the nitrogen our filtration system is contaminating it. Currently, it is supposed that this increase is due to either the filters or the pump. The filters were quoted for nitrogen purification systems but may fail to remove oxygen and water from the nitrogen. New filters have been ordered that will likely solve this problem. Additionally, the pump's location inside the glove box may be lead to contamination of the nitrogen. Any sparking or internal corrosion could introduce oxygen or other contaminants into the glove box atmosphere. Though these factors are likely the larger causes of contamination, the current purge and filtration periods being used are ballpark time estimates based on other similar systems and due to the differences in construction and function between these systems, the procedures may have to be adapted to suit this glove box. Upon arrival of new filters and relocation of the pump light bulb testing will continue and hopefully allow for determination of the necessary periods of purging and filtration.

Below are pictures of the constructed control and filtration unit, the glove box, and the connections between the two:



Figure 15: Control/Filtration unit front view



Figure 16: Control/Filtration unit back view



Figure 17: Glovebox feed-throughs with connections



Figure 18: Glovebox under pressure



Figure 19: Glovebox front view under pressure

5 Conclusions

The ability to produce and experiment with hybrid alkali mixtures allows the William and Mary Polarized ^3He Lab to further investigate the properties of hybrid alkali SEOP in an effort to maximize polarization as well as provide the lab with its own source of cleanly produced hybrid alkali mixtures. The glove box system completed this year is a vital portion of the alkali production process hopefully leading to higher polarizations. Presently the system has the ability to maintain desired pressure and flow in and filter nitrogen to a level of purity much higher than atmospheric levels, though unfortunately it is still far from the necessary 1-5ppm oxygen range. Light bulb tests indicate that efforts to reduce oxygen levels have shown improvements in the purging and filtration process and with further modifications and supplies, a clean glove box atmosphere is expected in the near future.

6 References

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