

Darkside: The Cryogenic Distillation of Underground Argon

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Abstract

Darkside is an experiment designed to conduct direct searches for dark matter. Among the proposed candidates for dark matter are Weakly Interacting Massive Particles (WIMPs). These particles do not interact via the electromagnetic nor the strong nuclear force rather only through gravity and the weak nuclear force. To detect these low energy events an ultra low background environment needs to be established to achieve the high level of sensitivity that is required for direct searches. Darkside employs an array of background reducing techniques as well as underground argon (UAr) as target in the time projection chamber (TPC). Atmospheric argon is not suitable for direct searches as it contains the radioactive isotope ^{39}Ar , which is the limiting factor in multi-ton detectors. In the most recent analysis, underground argon from the Kinder Morgan carbon dioxide plant in southwest Colorado was found to contain at least 150 times less ^{39}Ar than is present in the atmosphere [1]. At Fermilab there has been an ongoing effort to purify underground gas to produce detector grade argon for the detection of dark matter. Our goal is to purify argon with contamination levels in the parts per billion (ppb). To successfully distill UAr and reach levels of high purity, the distillation system requires close monitoring for fine tuning. Since the fractions of the constituents in the gas vary, constant adjustments need to be made to maintain the efficiency of the process. This summer I had the distinguished opportunity of making a contribution to this marvelous effort. In this document I will detail the process of distilling argon and the operation of the multi stage distillation system used to produce pure argon for the Darkside experiment.

Introduction

Argon is a bright scintillator that has attractive features for use in experiments attempting to directly detect WIMP-type dark matter [1]. The relatively low cost of argon technology also makes the deployment of ton-scale target masses feasible [1]. However, atmospheric argon contains the intrinsic ^{39}Ar , a beta emitter giving a specific activity ~ 1 Bq/kg [2]. This level of radioactivity does not limit the ability of small detectors for direct searches, but it is the limiting factor in large multi-ton detectors due to pile up.

The discovery of underground sources with significantly less ^{39}Ar than atmospheric argon was an important step in the development of direct-detection dark matter experiments using argon as the active target [1]. Underground argon is shielded from cosmic rays, which are responsible for most of the ^{39}Ar production, therefore it is expected to contain lower levels of ^{39}Ar . A recent more thorough study determined that underground argon from our current source of

extraction contains at least 150 times less ^{39}Ar than atmospheric argon [1]. This reduction of ^{39}Ar in underground argon will greatly enable detectors using argon as target to outperform their counterparts using atmospheric argon for direct dark matter detection.

Argon, however, is not found naturally in a pure concentration. In fact, at our current source of extraction the underground gas contains an argon fraction of only ~ 300 ppm. The gas must therefore be extracted and purified from contaminants to be effectively used in the search for dark matter. There are several established methods for separating gases. One common method to accomplish this, is through cryogenic fractional distillation. The method works by exploiting the volatility of gases to effectively achieve separation. The gas is first cooled and condensed to a liquid by cryogenic means. It is well understood that the difference in volatility of the constituents in a multi-component fluid allows for separation through distillation [3].

The challenge for us has been separating a gas where the fractions of the constituents vary

while simultaneously learning to operate the distillation column. The distillation system was engineered at Princeton University and built at Fermilab. Since an operation manual was non-existent, much of this task has been an R&D effort. Up to this point, many modifications have been made to the system to improve the functionality and effectiveness of the process. The modifications that have been made thus far are not included in this document, which focuses on the current plant. This paper describes the methods by which we purify and distill underground argon for the purpose of directly searching for dark matter.

Underground source

The Ar collected comes from the Kinder Morgan carbon dioxide plant located in the southwest region of Colorado. It is extracted at an average rate of ~ 0.3 kg/day. With a concentration of $\sim 96\%$, the raw gas is primarily CO_2 , the rest is mostly N_2 and He. The Ar concentration is only ~ 300 ppm while trace amounts of contaminants are also present. The low concentration of Ar makes the shipment of gas obtained directly from the well impractical and prohibitively expensive [2]. Therefore the gas first undergoes preliminary processing at our specially developed Vacuum Pressure Swing Adsorption (VPSA) plant located onsite at Kinder Morgan. Pressure (and Vacuum) Swing Adsorption plants work by exploiting the different rates of adsorption of different species of gas at a given partial pressure [2]. The VPSA plant effectively removes all of the CO_2 while producing a gas with a higher argon concentration of $\sim 30,000$ - $50,000$ ppm [2]. The gas sent to

Fermilab is then a He, N_2 , and Ar mixture.

System overview

From left to right in Fig.1, the purification system consists of the condenser booster, carbon filters, cold traps and the distillation column. The purification starts at the left with the condenser booster for helium separation. The waste gas from the booster then flows through a carbon filter to capture any residual argon that may be in the waste. At this point the gas is a N_2 Ar mix that is ready to be processed in the distillation column for N_2 separation. To prevent residual contaminants from entering the column the gas is filtered through the cold traps to freeze out any contaminants that may be present. It is important to minimize contaminants from making it into the column as their presence can hinder the operation and distillation process.

The purification plant has intricate plumbing that provides versatility of operation. The purification plant is built to support continuous and batch operation. However, because the constituents of the gas fractions vary, batch mode has proven to produce Ar with higher purity. That is attributed to the correlation between the system operating parameters and the gas composition. It is difficult to maintain a continuous operation while maintaining a high purity when the gas fractions vary.

To monitor the quality of the gas we use a universal gas analyzer (UGA) made by Stanford Research Systems (SRS). The UGA is not capable of analyzing an unknown gas and making quantitative determinations of the constituent gas fractions, rather one must “tell” the UGA what

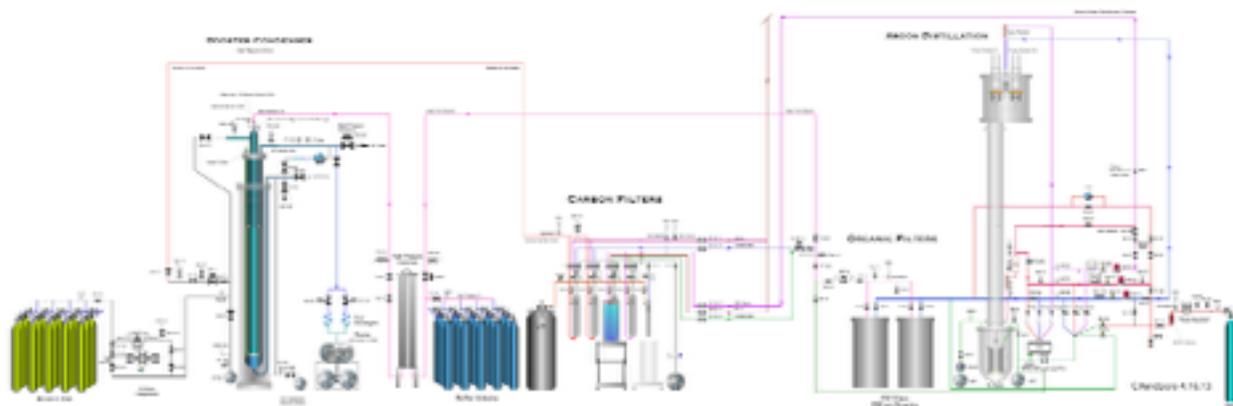


Figure 1. Purification Plant

gas to look for by giving it a molar mass [3]. The UGA then measures the current of ionized gas at a given mass to charge ratio and reports this as a partial pressure for a given mass [3]. There are multiple strategic locations throughout the system where the gas can be monitored. Each location has a sample port connected to the UGA. To view or monitor the gas, a flow is required by the UGA. It is not possible to simultaneously view multiple ports, for that reason it is important to ensure that the operator is monitoring the appropriate port. Failing to do so may result in the loss or contamination of argon.

To monitor and operate the purification system we use a LabVIEW control system which runs on a National Instruments CompactRio platform. LabVIEW is a data acquisition program commonly used to control processes by controlling instrumentation. LabVIEW stores data, which may then be read by the *history.vi*. The stored data is available for viewing at any given time. Since this is an R&D effort, it is useful to go back in time to correlate parameter settings and system behavior to determine optimal operating settings for a gas with specific gas fractions.

Condenser Booster

Upon arrival, the gas is sent through the first stage of the purification process for helium removal: the condenser booster. The system consists of a vacuum insulated pressure vessel for the purpose of condensing argon. Essentially, it is a 20 liter cylinder surrounded by a liquid nitrogen (LN_2) jacket with a vacuum jacket around it. At this stage of the process the composition of the gas is primarily helium with ~4% Ar and ~6% N_2 on average. The gas composition varies, the ratio of nitrogen to argon also varies, it can be higher or lower. This ratio is important as it will determine the temperature and pressure settings. Since helium boils at a very low temperature (4.2 K) it will remain in its gaseous phase while argon and nitrogen condense on the inner wall of the vessel, accumulating at the bottom as liquid. However, the efficiency of this process depends solely on the nitrogen to argon ratio.

The booster is pressurized to ~2700 psi (186 bar). The gas flows in through the top of the vessel and is cooled by the nitrogen in the LN_2 jacket. As the gas flows in at room temperature it causes the liquid nitrogen to boil and build pressure in the jacket. The pressure is controlled

by a manual back pressure regulator. The pressure is set to ~35 psia giving an operating temperature of 85 K inside the booster. At this temperature the vapor pressure of argon should be 0.7 bar or 0.38 % of the total pressure. However, when we vent the helium gas from the booster, we see over 1% argon in the waste stream. This means we are only condensing 80% of the input gas given a 5% argon concentration. For better condensation we can lower the pressure in the LN_2 jacket to effectively bring the temperature down. At atmospheric pressure argon freezes at 83.7 K, but because the gas is an argon nitrogen mix we are able to bring the temperature down to 69 K without freezing any argon (see Fig.2). The helium may be disregarded since it boils at a very low boiling temperature.

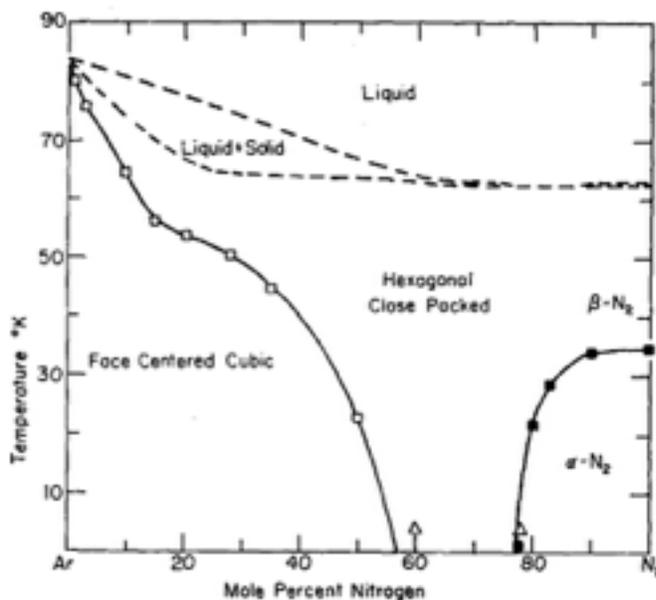


Figure 2. Argon nitrogen phase diagram

To bring the pressure in the LN_2 jacket down we pump on the jacket with a powerful vacuum pump. We bring the pressure sub-atmospheric to 1/3 bar. This gives us a temperature of 69 K in the booster. The vapor pressure at this temperature should be 0.07 bar or 0.038% of the total pressure. What we see in the helium waste is 0.27% argon concentration. The condensation is better but it is not perfect. The vessel is not completely engulfed by the LN_2 , therefore the temperature in the booster is not homogeneous. Also, the booster was originally

designed for a different purpose, but it was repurposed for helium removal.

The process of pressurizing and venting the booster is repeated until there is significant argon/nitrogen liquid accumulated to make a transfer. The liquid is transferred out of the booster by pressure to the distillation column. If the column is full we move to batch mode and a transfer is made to a buffer. Transferring liquid also makes available more surface area for greater condensation.

One of the problems we have encountered is monitoring the liquid level in the condenser booster. Along the length of the booster there are 4 PT1000 resistance temperature detectors (RTD) that we use to monitor the liquid level. However, they have failed to accurately report the level of the liquid. The resistance measured in the platinum RTD element is correlated to a temperature which has been well documented for platinum at atmospheric pressure. Therefore, based on the resistance measured by the RTD element we can determine if the sensor is in contact with liquid or gas. The problem is that while the booster is under pressure the PT1000's activate a signal on the control screen indicating there is liquid at that level, but once the pressure goes down the signal fades indicating there is no liquid at that level. After experimenting with PT1000's in high pressures we were able to determine they are not reliable and actually fail as liquid level monitors at high pressure.

Charcoal trap

To further prevent the loss of valuable underground argon the gas vented from the booster is filtered through activated charcoal to collect any residual argon that otherwise would be wasted. At cryogenic temperatures charcoal is known to be a good adsorbent of argon. For this reason it is kept submerged in LAr while venting. During this time the efficiency of the booster can be determined by viewing the incoming waste gas through the UGA port.

After several venting cycles, depending on the argon vapor in the waste, the charcoal becomes saturated and can no longer adsorb argon. Saturation becomes clear when argon rises above the background and appears on the RGA scan when monitoring the output of the charcoal trap. Regenerating the charcoal causes the charcoal to release the argon captured. This is done by heating the charcoal trap to 120 °C. The

gas released is put back into the booster to maximize UAr production.

Distillation column

Once helium has been removed, the gas is ready to be processed in the distillation column for argon nitrogen separation. The column is a 318 cm long stainless steel tube insulated by a high vacuum jacket to minimize heat transfer. The inside of the column is filled with 56 elements of Sulzer Chemtech 80mm DX packing material to increase the surface area for heat exchange. To monitor the temperature in the column there are 5 temperature monitors along its length. There are two volumes at the top, the condenser (C) and the condensing volume (CV) and one at the bottom, the re-boiler. Before the gas enters the column it is filtered through the cold trap to freeze out any residual contaminants that may be present. This step is necessary not only to maintain the purity of the argon but to minimize the risk of freezing the input line with impurities, which may cause impedance to the flow of gas.

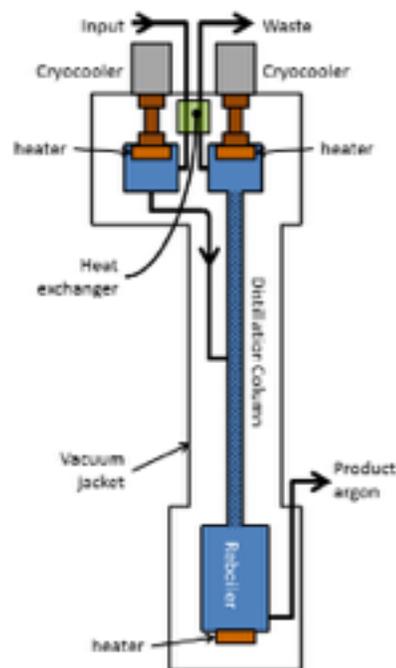


Figure 3. Distillation column

The gas flows into the condensing volume (CV) where it is cooled and condensed allowing for it to be injected at the mid point of the column as liquid. As it enters the column some of the liquid falls to the re-boiler while some vaporizes up to the condenser. Some of the nitrogen at the top exits while some condenses and falls back down through the packing elements. Meanwhile, the argon in the re-boiler is boiling, rising up through the packing elements. Nitrogen and argon meet and exchange heat in the packing elements. The warm argon gas heats the cold nitrogen causing it to re-vaporize. Argon loses its heat and condenses, falling back down to the re-boiler. This continuous up and down cycle determines the purity of the argon and it is known as the reflux. The reflux is defined as the flow rate inside the column divided by the flow rate out of the column. Minimizing the flow rate out increases the purity of the argon.

At full reflux the heater maintaining the temperature of the cryocooler of the condenser outputs 0 Watts. In this mode, the heater has no work to do because the cryocooler is maintained at its temperature set point by the heat in the nitrogen gas. In essence the power from the heater in the re-boiler is conserved. Since nitrogen balances the distillation process we try to maintain its concentration in the column. At the condenser (C) some nitrogen condenses while some exits through the waste line. Naturally the concentration of nitrogen goes down. We balance the distillation process by flowing in a constant stream of nitrogen argon gas.

It turns out that the temperature gradient (from top to bottom of the column) is non linear. Rather, the temperature gradient behaves more like a step function. The horizontal component of the step function is what we call the temperature transition. This temperature transition is an indicator of where nitrogen and argon are in the column. We monitor the 5 temperature sensors along the column to maintain the temperature transition in a relatively safe location. When the transition falls it is because cold nitrogen is pushing down the column of rising argon gas. When the temperature on the monitors start to rise we know warm argon is pushing up. We adjust the duty of the heater in the re-boiler to control the pressure. Allowing the transition to rise too high in the column risks losing argon in the waste line. Allowing the transition to drop too low will risk contaminating the purity of the argon with

nitrogen, the first scenario being the more catastrophic of the two.

Argon methane distillation

Normally the gas we receive from Colorado has small trace amounts of contaminants in the ppm. However, we had a batch that was high in methane contamination with more than 1%. Although the column was built to separate nitrogen and argon, for the first time in June 2014, we proved that it was capable of separating argon and methane. Since methane has a higher boiling temperature than argon we reversed the distillation and collected argon through the waste line at the top while methane was collected at the bottom in the re-boiler. The separation was very effective since the difference in their boiling temperatures is relatively higher when compared to that of argon and nitrogen. The exact level of the purity of the argon is yet unknown as we are awaiting results from Pacific Northwest National Laboratory (PNNL). All of the gas we purify and distill is sent to PNNL for a thorough professional analysis. The sensitivity level of the UGA is not sufficient enough to provide an accurate measurement of the small contamination levels.

Results and future plans

The dual phase TPC of the Darkside experiment employs 152 kg of UAr. At the time this paper was written 87.6 kg of UAr have been purified through the distillation column. We have ~21 kg of UAr that have high levels of oxygen (from the underground source), which we are currently working to reduce. Thus far we have managed to reduce the oxygen level by a factor of 10 through distillation. A total mass accounting of UAr including the purified mass, the oxygen contaminated mass, and the UAr in transit shows we have ~132 kg. The 152 kg goal is expected to be reached by this fall. The purification process described in this paper was able to purify argon to at least 99.9%. A complete analysis was done by PNNL (see Fig.4).

The next and final stage of purification is currently under construction. To reach levels of contamination lower than ppb, the UAr will be processed through a heated zirconium gettering system. The system has been built and is now in the final stages of completion. Since there is a limit to the amount of oxygen that the heated

zirconium getter can process, an oxygen scrubber system will be retrofitted to further lower the oxygen concentration before the gas can be sent through the gettering system. The oxygen scrubber system is in its early stages of

construction but it is expected to be up and operating in the next couple of months.

Cylinder	Mass	Ar	N ₂	O	CH	CO
UAr#2	6.6 kg	99.97%	240 ppm	80 ppm	<10 ppm	<10 ppm
UAr#3	10.7 kg	99.993%	30 ppm	<10 ppm	40 ppm	<10 ppm
UAr#4	10.7 kg	99.917%	30 ppm	740 ppm	60 ppm	<10 ppm
UAr#5	10.7 kg	99.944%	30 ppm	420 ppm	110 ppm	<10 ppm
UAr#6	10.7 kg	99.956%	50 ppm	250 ppm	120 ppm	20 ppm
UAr#7	9.2 kg	not avail.	not avail.	not avail.	not avail.	not avail.
UAr#8	8.0 kg	not avail.	not avail.	not avail.	not avail.	not avail.
UAr#9	12.9 kg	not avail.	not avail.	not avail.	not avail.	not avail.
UAr#10	8.1 kg	not avail.	not avail.	not avail.	not avail.	not avail.

Figure 4. Purification results

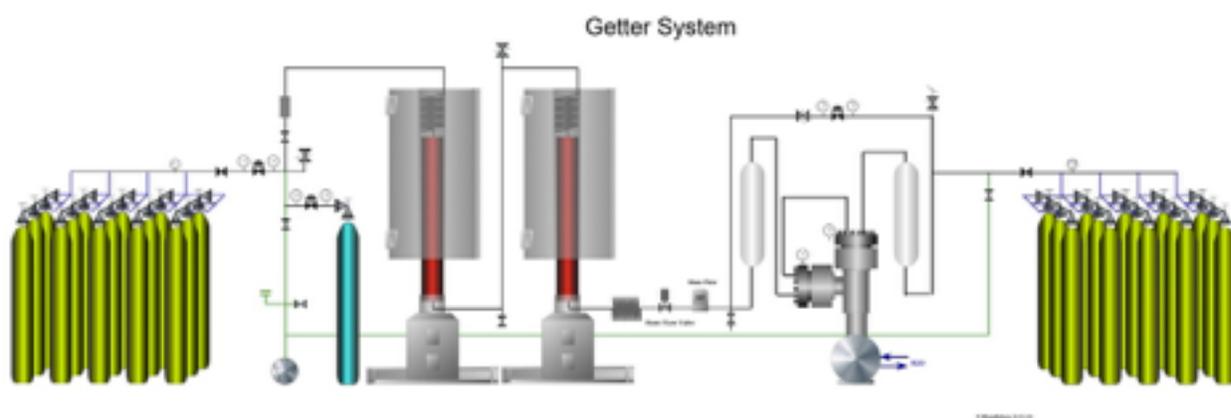


Figure 5. Gettering system

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