Depleted Argon from Underground Sources

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Abstract. Argon is a strong scintillator and an ideal target for Dark matter; however ^{39}Ar contamination in atmospheric argon from cosmic ray interactions limits the size of liquid argon dark matter detectors due to pile-up. Argon from deep underground is depleted in ^{39}Ar due to the cosmic ray shielding the earth. In Cortez Colorado a CO_2 well has been discovered to contain approximately 600ppm of argon as a contamination in the CO_2. We first concentrate the argon locally to 3% in an Ar, N_2, and He mixture, from the CO_2 through chromatographic gas separation, and then the N_2 and He will be removed by continuous distillation to purify the argon. We have collected 26 kg of argon from the CO_2 and a cryogenic distillation column under construction at Fermilab to further purify the argon.

Keywords: argon, cryogenic, distillation, dark matter, scintillator, time projection chamber.
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INTRODUCTION

As a strong scintillator argon is an ideal medium for Dark Matter detection. However, a limiting contamination in atmospheric argon is the beta emitter ^{39}Ar ($^{39}Ar \rightarrow {}^{39}K + e^- + \bar{\nu}_e$ $(Q = 535\ keV)$). It is possible to reject beta events through pulse shape analysis, but ^{39}Ar limits the size of a detector due to event pile-up. ^{39}Ar is produced in the upper atmosphere in ^{40}Ar(n,2n) reactions, and is found in the atmosphere at a level of 8.1×10^{-16} ^{39}Ar/^{40}Ar; corresponding to more than 1 Bq/kg of atmospheric argon. A place to search for argon gas depleted in ^{39}Ar is deep underground where the cosmic ray flux is highly suppressed and therefore ^{39}Ar production rate is significantly reduced. We have identified a CO_2 well near Cortez, Colorado that contains argon at the 600 ppm level. The ^{39}Ar concentration in this argon has been measured to be reduced by more than a factor of 25. However, the argon must be extracted from the CO_2 and further purified in order to be used in a detector.

CHROMATOGRAPHIC GAS SEPARATION

It is possible to trap specific gases on an absorptive medium, while allowing other gases to pass. The absorption efficiency is directly proportional to the gas pressure, and this property can be exploited to create very efficient absorption devices, and to regenerate the absorptive medium (1). Outside of Cortez Colorado we have built a vacuum-pressure swing absorption (VPSA) unit to extract the depleted argon from a CO_2 well. CO_2, O_2, and other unwanted gases are trapped on Zeolite with high efficiency under pressure. Eventually the Zeolite will become saturated with the unwanted gases, and can pass through the column. Therefore flow is stopped through the column, and a vacuum pump is applied to the system to force the Zeolite to release the contaminants, and return them to the company. By using two columns we can have continuous production by absorbing on one column, while the other column is being regenerated. Figure 1 shows an illustration of the two column VPSA principle. Our unit consists of 2 stages, of 2 columns each. The first stage traps CO_2, O_2, and other unwanted gases, and while still under pressure the second stage continues to remove these gases as well as some of the nitrogen.

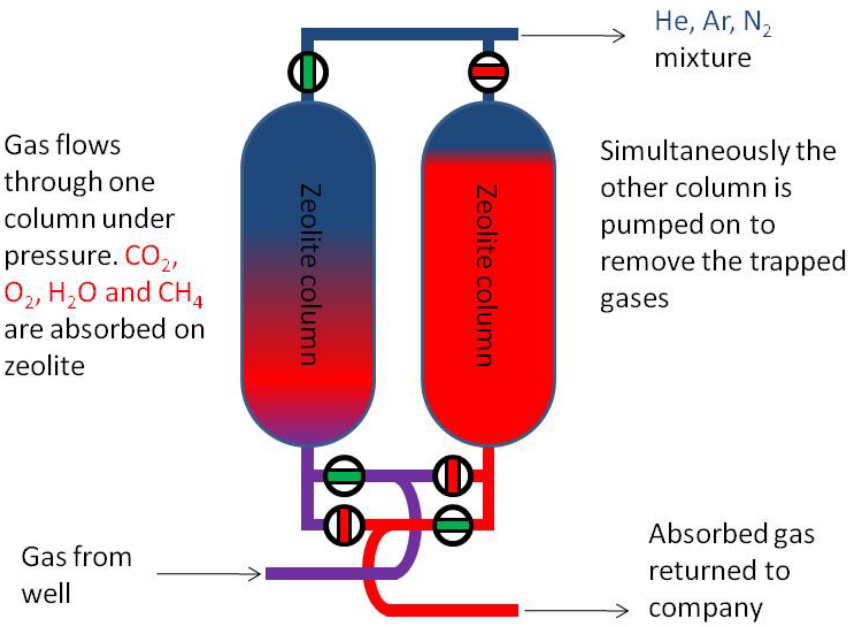

Figure 1. Illustration of the VPSA gas extraction principle.

Figure 2. VPSA system in Cortez, CO.

The VPSA system in Cortez, CO, has been operating since February 2010. It effectively concentrates argon, but there are some gases that are currently irreducible. Helium cannot be separated by using the VPSA, and currently nitrogen is not effectively removed by the columns. In Table 1 we see what the gas fractions are for the gas coming from the well and what the fractions are after the VPSA unit. As of the Fall of 2010 we have collected approximately 26 kg of argon in pressurized cylinders.

Table 1. Composition of the well gas before and after the VPSA extraction. The fraction of argon is increased significantly.

Gas type	Concentration from well	Concentration after VPSA extraction
Carbon Dioxide	96%	Below sensitivity
Nitrogen	2.4%	70%
Methane	5,700 ppm	Below sensitivity
Helium	4,300 ppm	27.5 %
Other hydrocarbons	2,100 ppm	Below sensitivity
Water	1,000 ppm	Below sensitivity
Argon	600 ppm	2.5%
Oxygen	Below sensitivity	Below sensitivity

CONTINUOUS DISTILLATION GAS SEPARATION

It is well understood that the difference in boiling points of the constituents in a multi component liquid allows us to perform separation through distillation. It is possible to perform distillation continuously in a column packed with a high surface area material, and a temperature gradient. The liquid will boil and the gas condenses continuously on the packing material. Gases rise and recondense, while liquids sink and reboil. The component with the lower boiling point rises preferentially and the component with the higher boiling point drops down to the lower volume. By maintaining a constant temperature gradient and a constant flow of liquid into the column, an equilibrium is set up, and very pure material can be extracted from the column.

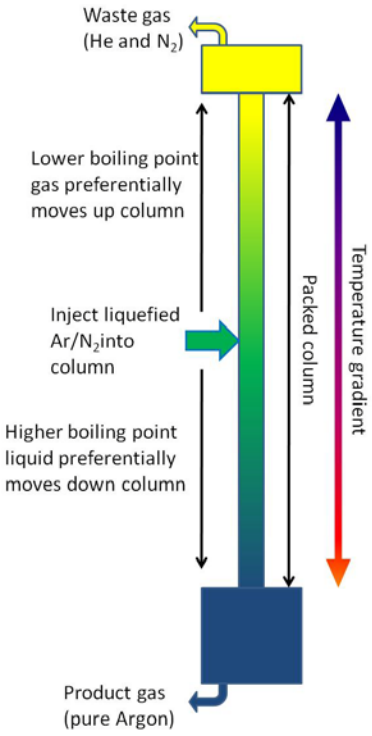

Figure 3. Illustration of the continuous distillation in packed column process principle.

At Fermilab a cryogenic distillation column has been constructed for the purification of the gas extracted for the CO_2 wells in Cortez, CO (Figure 4). The column is 320 cm tall and 2 cm in diameter, and is filled with a custom packing material (Figure 5), which makes this column equivalent to 40 theoretical stages. Two 600 watt cryocoolers that are regulated with 600 watt hears are used to liquefy the N_2 and Ar gas, which is injected into the column, and along with a heater on the lower volume (reboiler), the second cryocooler is used to set up the temperature gradient along the column to facilitate the distillation. Temperature monitors throughout the system are ready out by 2 LakeShore units, which are used to regulate the temperature with heaters in the system. When complete the cryogenic distillation column will be able to produce 99.9999% pure argon at a rate of 5kg/day.

Figure 4. Cryogenic Distillation Column at Fermi National Accelerator Laboratory.

Figure 5. Custom packing material in the distillation column.

ACKNOWLEDGMENTS

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REFERENCES

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