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ScienceDirect
Nuclear Instruments and Methods in Physics Research A 587 (2008) 46–51
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH
Section A
www.elsevier.com/locate/nima

Discovery of underground argon with low level of radioactive ^{39}Ar and possible applications to WIMP dark matter detectors

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Received 9 October 2007; received in revised form 27 November 2007; accepted 14 December 2007
Available online 2 January 2008

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doi:10.1016/j.nima.2007.12.032

Abstract

We report on the first measurement of ^{39}Ar in argon from underground natural gas reservoirs. The gas stored in the US National Helium Reserve was found to contain a low level of ^{39}Ar. The ratio of ^{39}Ar to stable argon was measured to be $\leqslant 4 \times 10^{-17}$ (84% C.L.), less than 5% the value in atmospheric argon ($^{39}\mathrm{Ar}/\mathrm{Ar} = 8 \times 10^{-16}$). The total quantity of argon currently stored in the National Helium Reserve is estimated at 1000 tons. ^{39}Ar represents one of the most important backgrounds in argon detectors for WIMP dark matter searches. The findings reported demonstrate the possibility of constructing large multi-ton argon detectors with low radioactivity suitable for WIMP dark matter searches.

PACS: 23.40.−s; 07.20.Mc; 95.35.+d

Keywords: Dark matter; Low background detectors; Cryogenic noble gases

1. Introduction

The existence of dark matter is well established, but its composition is unknown. One possible candidate is a gas of Weakly Interacting Massive Particles (WIMPs) formed in the early history of the Universe. The WIMP particle is also motivated theoretically in extensions of the standard model based on supersymmetry and will be the subject of searches in upcoming experiments at the LHC at CERN. WIMP dark matter particles, if they exist, could be detected by observing their collisions with ordinary nuclei as the earth moves through the gas. Because of the low relative velocity between the target and the WIMPs, the nuclear recoils will have a small energy. For WIMPs with a mass of ~100 GeV and medium mass target nuclei, the recoil spectrum is continuous with a maximum kinetic energy of ~100 keV. The WIMP-nuclear cross-section is expected to be at the weak interaction scale, and thus expected rates are small, possibly as low as a few events per ton of target per year. Detecting WIMP dark matter could require a large detector with low background and a low threshold [1].

The noble elements—neon, argon, and xenon—are ideal targets for WIMPs searches as they allow detection of rare WIMP-induced nuclear recoils down to a few keV by scintillation and/or ionization. Liquid argon, in particular, is an excellent material for use as a detector of ionizing particles. It produces copious scintillation light, allows the drift of the ionization charge over long distances, and it has been used for large detectors [2]. Moreover, the difference in the stopping power between nuclear recoils and β/γ events leads to a significant difference in the pulse shape of scintillation light and in the ratio of ionization charge to scintillation light providing powerful tools to separate WIMP-induced events from natural radioactivity [3,4]. Studies of the beta/recoil discrimination with the WARP 3.2-kg liquid argon detector demonstrate that the discrimination by pulse shape alone permits a separation of 1 recoil in $10^8 \beta/\gamma$ events, and the β/recoil separation by the ratio of scintillation to ionization is 1 in 10^2 [3].

For a liquid argon detector, the separation of recoil events from β/γ events is particularly important because of the intrinsic background from β decays of ^{39}Ar, present in atmospheric argon. The specific activity of ^{39}Ar ($Q = 565\,\mathrm{keV}$, $t_{1/2} = 269\,\mathrm{y}$) is $\sim 1\,\mathrm{Bq/kg}$ of atmospheric argon [5,6]. ^{39}Ar is produced by cosmic ray interactions in the atmosphere, principally via the $^{40}\mathrm{Ar(n,2n)}^{39}\mathrm{Ar}$ reaction [7,8].

The WARP 3.2-kg detector [3] published results from a first search for WIMPs obtaining a sensitivity comparable to the best current limits [9]. The high selectivity for argon recoils should permit a sensitive WIMP search with a 140 kg liquid argon detector employing atmospheric argon, currently under construction at Laboratori Nazionali del Gran Sasso [10]. However, in spite of its favorable β/recoil discriminating power, it is highly desirable to use argon with a much lower ^{39}Ar contamination for future, larger detectors. Based on the proven β/recoil discrimination, a 10-fold or more reduction of ^{39}Ar with respect to the atmospheric level would enhance the prospects of future multi-ton argon WIMP detectors.

The availability of large quantities of argon depleted in ^{39}Ar may also enable proposed experiments to study neutrinos from reactors and from high-intensity stopped-pion neutrino sources through neutrino-nucleus elastic scattering [11,12], with the potential of constraining parameters for non-standard interaction between neutrinos and matter, and of realizing precision measurements of the weak mixing angle and of the neutrino magnetic moment [12]. Thanks to the excellent properties of identification of nuclear recoils from β/γ events, depleted argon could be used for the development of small portable neutrino detectors to monitor reactor sites for non-proliferation efforts [11]. Depleted argon could also be used to develop neutron detectors for port security.

Centrifugation or differential thermal diffusion are established methods for $^{39}\mathrm{Ar}/^{40}\mathrm{Ar}$ isotopic separation, but such techniques could become extremely expensive and require a long production time on a multi-ton scale. Argon from natural gas wells is a promising source of ^{39}Ar-depleted argon because ^{39}Ar production induced by cosmic rays is strongly suppressed underground. Shielding of target materials in deep underground reservoirs has recently played a crucial role in solar neutrino physics: the Borexino experiment measured the sub-MeV ^{7}Be solar neutrinos in a liquid scintillator target [13], beating background from cosmic ray-induced ^{14}C thanks to the extremely low level of the ^{14}C/C ratio found in crude oil reservoirs ($\sim 10^{-18}$), six orders of magnitude below typical modern carbon values [14].

In the subsurface, ^{39}Ar can be produced by a number of reactions, mainly neutron reactions on potassium, such as ^{39}K(n,p)^{39}Ar [15]. Argon samples collected in groundwater from U and Th rich crystalline rocks revealed an enhanced ^{39}Ar activity, with ^{39}Ar/Ar ratios exceeding atmospheric values by up to 16 times [16]. The enhanced activity is likely due to an abundant local neutron flux, originating from fission or (α,n) reactions induced by decays in the U and Th chains. The ^{39}Ar/Ar ratio in subsurface gases thus depends on the local U, Th, and K concentration, on the porosity of the surrounding rocks, and may vary significantly among geological formations.

Prior to this work, measurements of the ^{39}Ar/Ar ratio in natural gas wells are not available in the literature, to the best of our knowledge.[1] The ubiquitous presence, in natural gas, of ^{4}He—a by-product of radioactive decays in the U and Th chains—and the correlation between ^{40}Ar and ^{4}He content [18] called for a measurement of the ^{39}Ar/Ar ratio in underground argon to assess its potential for use in ultra-sensitive WIMP detectors.

2. Discovery of underground argon with low level of radioactive ^{39}Ar

We report on the measurement of argon from the US National Helium Reserve, located in the Cliffside Storage Facility outside Amarillo, TX [19]. The facility stores crude helium separated by chromatography and/or cryogenic distillation from the nearby helium-rich natural gas fields. The separation processes employed also transfer a limited fraction of the argon contained in the natural gas along with the crude helium stream. Production takes place in nine separate plants (five in Kansas and four in Texas) so that the gas in the Reserve represents an average sampling of natural gas sites in the Texas Panhandle and southern Kansas. All plants are connected to the US National Helium Reserve by the Helium Conservation Pipeline, run by the Bureau of Land Management.

Crude helium samples originating from the National Helium Reserve were collected directly from the Helium Conservation Pipeline at the Kansas Refined Helium plant in Otis, Kansas. In order to minimize contamination from atmospheric argon in the samples, the booster pump used to transfer crude helium into the bottles was driven by a separate stream of crude helium. The composition of the crude helium, as measured by mass spectrometry, is given in Table 1.

Table 1
Composition of crude helium extracted from the National Helium Reserve

Component	Concentration by volume
He	77.3%
N_2	20.3%
CH_4	1.6%
H_2	8000 ppm
Ar	680 ppm
CO_2	110 ppm

The crude helium was processed with a novel Pressure Swing Adsorption (PSA; see Ref. [20]) plant developed at Princeton. The unit is capable of processing the gas stream to remove the strongly adsorbing components (N_2, CO_2, H_2S, CH_4, and heavier hydrocarbons), concentrating Ar, He, and H. The concentration of strongly adsorbed gases—N_2, CH_4, and CO_2—was reduced by a factor $>10^4$ [21]. At the output of the PSA plant, argon was trapped at 76 K on liquid N_2-cooled activated charcoal and separated from helium and hydrogen. The gas de-sorbed from the charcoal trap was ~80% argon, the remaining 20% being mainly helium (with traces of N_2 and hydrogen). Contamination from atmospheric argon is negligible, the whole gas separation system being leak tight to 10^{-9} mbar l/s.

The PSA plant used for this work consists of two columns filled with zeolites 13X (Fig. 1). Zeolites have a good selectivity for Ar versus N_2: the adsorption capacity for Ar at 300 K and atmospheric pressure is of 2 l/kg, compared to an adsorption of O_2 of 9 l/kg [22]. The quantity of gas adsorbed depends linearly on the partial pressure for total pressures up to 100 kPa. CO_2, H_2S, CH_4, and heavier hydrocarbons are much more strongly adsorbed than N_2 on zeolites 13X. H_2 and He are adsorbed less strongly than Ar, and O_2 behaves in a very similar way to Ar. Our PSA plant exploit this difference in adsorption to separate the individual components in a product and a waste stream. Concentration of components with low adsorption is strongly enriched in the product stream. By alternating the pressure and direction of the gas flow, and running the two columns in opposite phases through consecutive high pressure feed cycles and low pressure purge cycles, the separation of the two streams can be performed in a continuous cycle. With this configuration, the adsorbent is self-regenerated during the low pressure purge cycle.

The gas flow in our PSA plant is directed by solenoid valves. The control system for the solenoid valves consists of a set of relays interfaced with a programmable automation controller, designed for industrial control. The software for the control system was developed in Princeton using the `LabVIEW FPGA` platform.

Measuring ^{39}Ar at or below atmospheric concentration is very challenging. A review of the methods used to measure trace levels of rare gas isotopes is given in Ref. [23]. The standard method, used in this work, is direct

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[1] While this work was being completed, we learned of a previous unpublished attempt to measure the ^{39}Ar/Ar ratio in subsurface gas [17].

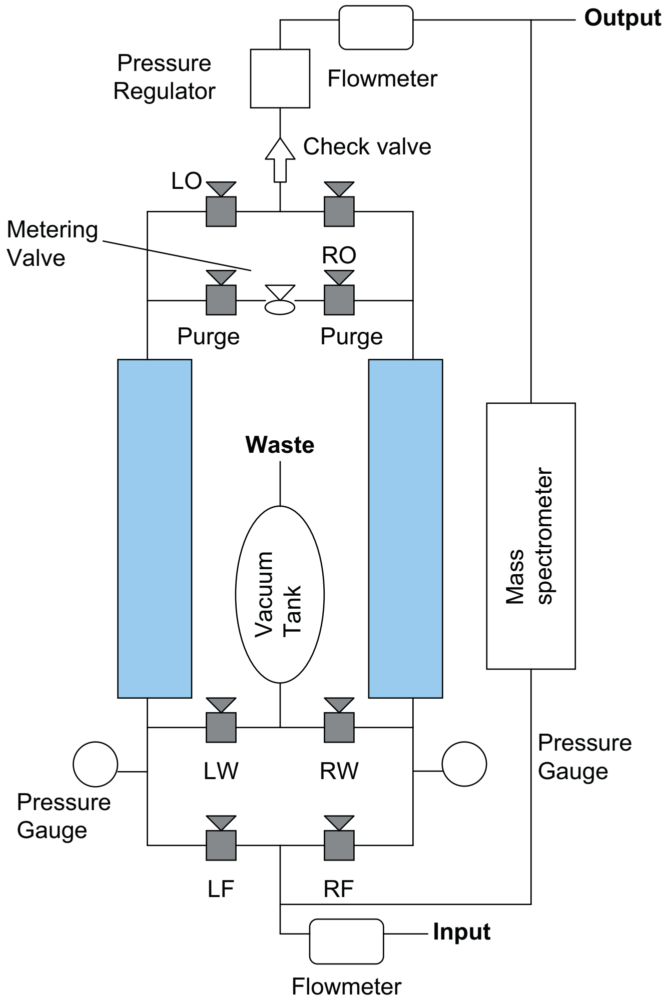

Fig. 1. Diagram of the PSA plant developed at Princeton. See text for details.

counting in underground, low-background proportional counters [24,28]. ^{39}Ar concentrations detectable by this method are typically ~5% of atmospheric value [23]. We note that activation of ^{40}Ar in the sample during the three months exposure to the surface flux of cosmic rays is negligible within the accuracy of our measurement.[2]

The measurements reported here were performed at the Low Level Counting Underground Laboratory at the Physics Institute at the University of Bern, located underground at a depth of 70 m water equivalent. This is, to our knowledge, the only facility where a number of low background proportional counters are dedicated to routine measurements of the ^{39}Ar/Ar ratio in samples of various origins for environmental and climatic studies [25] (Fig. 2).

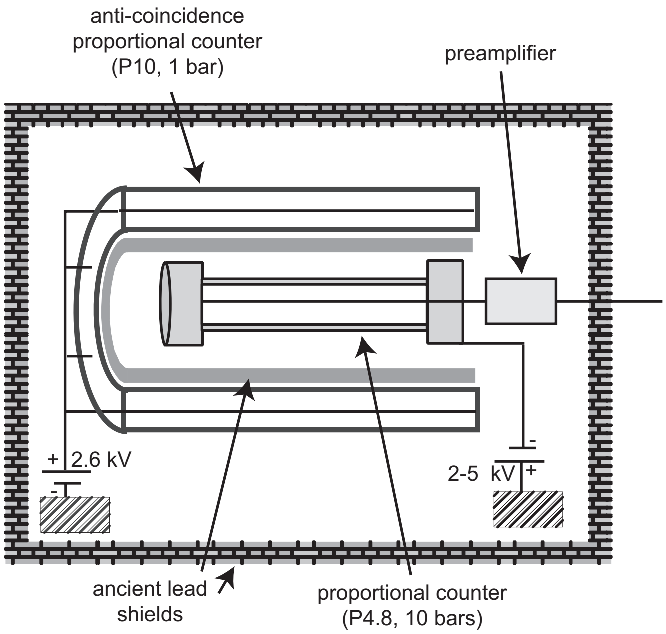

Fig. 2. Schematic of the setup utilized to measure the ^{39}Ar/Ar ratio at the University of Bern Low Level U. Lab. For ^{39}Ar measurements, the counters are filled with a P4.8 mixture (4.8% methane, 95.2% argon) at pressures in the range 5–25 bars. The measurements are performed at a depth of 35 m. Low activity lead shield and an anti-coincidence arrangement provide a further reduction of the background count rate.

The laboratory is located at a depth of 35 m, providing a reduction of the muon flux by a factor of ~10 [24]. The lab walls are constructed utilizing a special concrete selected for its low radioactivity content, in order to minimize the gamma-ray flux within the lab. The main components of the detector and of the counting system are shown in Fig. 2. A 100 cm^3 proportional counter (dimensions: 25 cm length and 2.2 cm diameter), built of high-conductivity oxygen-free copper, is filled with the sample gas at a pressure of 10 bars and placed in a cylindrical lead shield 5 cm thick (see Fig. 2). Only a fraction of the β-particle decay energy is released within the counter before they reach its wall. The deposited energy is recorded by a 7-bit Multi-Channel Analyzer (MCA), with a linear energy range of 0–35 keV; events with energies greater than 35 keV are recorded as saturated events in the last MCA channels and included in the analysis.

For background reduction, the proportional counter is constructed with low background materials and uses an anti-coincidence proportional counter to reduce background. The assembly is inserted in a second, larger, cylindrical proportional detector, which acts as an anti-coincidence counter. The passive shielding is complemented by an external lead shield 12 cm thick. The lead shields were built using lead from ancient ship wrecks with very low ^{210}Pb intrinsic activity.

The detector background is comparable to the rate due to ^{39}Ar in atmospheric argon and must be determined accurately. The background of the whole counting system is measured in a separate run using argon, depleted in ^{39}Ar by at least a factor of 20–50 through differential thermal diffusion at Monsanto Co. (Miamisburg OH, USA;

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[2] The activation on the surface is lower than the average activation time in the atmosphere, given that most of the ^{39}Ar production takes place in the upper layers of the atmosphere. The e-folding activation time in the atmosphere is the ^{39}Ar meanlife, i.e. 269 years. Therefore an upper limit for the activation in three months is given by: $3\,\text{mos}/269\,\text{year} = 0.1\%$.

see also Ref. [5]). The ^{39}Ar/Ar content of such sample was confirmed to be, at 99% C.L., lower than 3% of the ^{39}Ar/Ar ratio in the atmosphere by comparison with gas samples extracted from ancient ice—independently dated at more than 1000 old—and with gas samples from several aged groundwater sources [5,26,27].

Three measurements are performed to measure the ^{39}Ar content in the sample of interest: one with the ^{39}Ar-depleted gas to measure intrinsic background, one with standard atmospheric argon for reference, and one with the underground sample itself. The ^{39}Ar activity of the sample under investigation is evaluated by subtracting the detector background from the total measured activity and is then compared to the ^{39}Ar activity in atmospheric argon (background subtracted). Results are expressed in terms of the ^{39}Ar/Ar ratio relative to the atmospheric ratio.

The energy spectrum above threshold was analyzed with the routine procedure used for all ^{39}Ar measurements performed in the Low Level Counting Underground Laboratory (for a detailed review of the procedure, see Ref. [5]). Table 2 reports the count rate from the three samples in four regions of interest (ROI). The count rates in every ROI for the argon sample from the US National Helium Reserve are in good agreement with rates from the reference ^{39}Ar-depleted gas. The best estimate for the ^{39}Ar activity is obtained from the total count rate in the whole spectrum (Channels 20–127) and indicates a ^{39}Ar/Ar ratio $\leqslant 4\times10^{-17}$, which is less than 5% of the ^{39}Ar/Ar ratio in atmospheric argon. The depletion factor from the atmospheric activity of the sample is therefore $\geqslant 20$ at 84.1% C.L. ($\geqslant 10$ at the 97.7% C.L.).

3. Conclusions

This result is, to the best of our knowledge, the first measurement of the concentration of ^{39}Ar in subsurface gas. It represents an upper limit which is based on the background of the proportional counting system. It therefore motivates the development of new and potentially more sensitive techniques, such as Accelerator Mass Spectrometry (AMS) [28] and ^{39}Ar-decay counting in liquid argon detectors [6] for a more accurate determination of the ^{39}Ar/Ar ratio. AMS measurements at the ATLAS facility at Argonne National Laboratory are already capable of measuring ^{39}Ar/Ar ratios around 5% of the atmospheric levels, comparable to the best limits achievable with gas proportional counters. With improvements currently under development to reduce the ^{39}K background in the detector while still allowing a high beam current, a new limit of approximately 10 times lower is expected. ^{39}Ar-decay counting in the 3.2-kg WARP detector already reached an accuracy of 10% of atmospheric levels, and a similar, specially designed, low background, ~10-kg detector could achieve a sensitivity 100 times higher, down to 1 part in 10^3 of the atmospheric activity.

The discovery of low ^{39}Ar/Ar ratio in the US National Helium Reserve is part of a larger program of exploration of several possible underground argon sources. During this investigation, the authors developed the technology required to separate and collect large quantities of argon from natural gas wells. The quantity of argon processed by the Kansas Refined Helium plant is 25 tons per year. Production of a 10 ton target of liquid argon for a dark matter search experiment would take a 1-year production campaign. The production would also require the construction of dedicated production plants, which could take a 1–2 years period after the project is funded and before the start of production.

These findings lead the way for future multi-ton, low background, argon detectors for WIMP dark matter, able to reach sensitivities for the WIMP-nucleon cross-section of 10^{-46} cm^2 or smaller. The availability of large quantities of argon depleted from ^{39}Ar will also be beneficial for studies of neutrino properties through neutrino-nucleus coherent scattering. It may also enable the construction

Table 2
Count rate for different ROIs: Channels 20–50, 51–102, 103–127, and the whole spectrum above threshold, 20–127

Argon sample from National Helium Reserve, Amarillo, TX	Count rate (μBq)			
	Ch. 20–50	Ch. 51–102	Ch. 103–127	Ch. 20–127
(1) Underground Ar	460 ± 21	480 ± 21	1096 ± 32	2036 ± 43
(2) ^{39}Ar-depl. Reference	454 ± 23	512 ± 25	1068 ± 35	2035 ± 49
(3) Atmospheric Ar	542 ± 30	855 ± 37	2228 ± 60	3625 ± 77
(4) (Under. Ar)−(Ref.)	6 ± 30	−33 ± 32	28 ± 48	1 ± 65
(5) (Atm. Ar−(Ref.)	88 ± 38	342 ± 45	1160 ± 70	1589 ± 91
(6) [(Under. Ar)−(Ref.)]/[(Atm. Ar)−(Ref.)]				0.00 ± 0.04
(7) $(^{39}\mathrm{Ar/Ar})_{\mathrm{und}}/(^{39}\mathrm{Ar/Ar})_{\mathrm{atm}}$				$\leqslant 0.05$

The rates are reported for the three gas samples under consideration (lines 1–3: argon from the US National Helium Reserve, ^{39}Ar-depleted reference, atmospheric argon). The majority of the counts are due to the detector background. This background is measured by using a ^{39}Ar-depleted reference gas and subtracted from the activities measured for the sample from the US National Helium Reserve (line 4) and for atmospheric argon (line 5). The ^{39}Ar/Ar ratio for the underground sample is 0.00 ± 0.04 relative to the atmospheric value, where the uncertainty comes from the statistical errors associated with the three measurements (line 6). The best estimate becomes $\leqslant 0.05$ when taking into account the uncertainty on the ^{39}Ar/Ar ratio for the reference sample, which is combined in quadrature with the statistical uncertainty (line 7).

of small and portable neutrino detectors for reactor monitoring in non-proliferation efforts and of neutron detectors for port security.

Acknowledgments

This work was supported by the US National Science Foundation under Grant No. 0704220. This work was supported in part by the US Department of Energy, Nuclear Physics Division, under contract DE-AC02-06CH11357. The WARP program is funded by the italian Instituto Nazionale di Fisica Nucleare and by the US National Science Foundation. A.M. Szelc has been in part supported by a grant of the President of the Polish Academy of Sciences and by the MNiSW Grant 1P03B04130. The authors express their gratitude to the Kansas Refined Helium Co., a division of Linde, for access to the Helium Conservation Pipeline and assistance with sampling and transport. The authors thank W. Brinkman, D. Marlow, J. Russell, and P. Wraight for their support and encouragement.

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