Low-Radioactivity Argon for Rare-Event Searches

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Abstract. ^{39}Ar and ^{42}Ar produce irreducible backgrounds for several argon-based dark matter and neutrino experiments. The use of low-radioactivity underground argon (UAr) could be a solution to the problem. The DarkSide-50 experiment demonstrated that argon derived from underground sources can contain a much lower concentration of ^{39}Ar than atmospheric argon. Following this success, the Global Argon Dark Matter Collaboration is procuring hundreds of tons of UAr for the DarkSide-20k detector. To serve the broader community need, it is increasingly important to identify new sources of low-radioactivity argon. In addition, understanding the underground production mechanisms of argon radioisotopes and devising methods to measure them at ultra-low levels is necessary. The use of low-radioactivity argon could be crucial to expanding the physics goals and sensitivity of next-generation large-scale argon-based experiments.

INTRODUCTION

Low-radioactivity argon is crucial to several next-generation large-scale dark matter and neutrino experiments. The DarkSide-50 experiment demonstrated that argon derived from underground sources can be highly depleted of ^{39}Ar [1]. The measurements showed ^{39}Ar content to be a factor of 1400 times lower in underground argon (UAr) than in atmospheric argon (AAr). This discovery has broadened the physics reach of argon-based detectors and increased the demand for low-radioactivity argon in rare-event searches as well as in other related sciences. The Global Argon Dark Matter Collaboration (GADMC) [2] is planning to procure hundreds of tons of UAr for the DarkSide-20k detector. Current sources of low-radioactivity argon may prove to be insufficient for the broader needs of the community. Thus, it is important to pursue identification of new sources so that they will be available when needed. Large-scale commercial procurement of UAr from an argon-enriched gas stream is feasible. Kiloton-scale UAr-based experiments like the one proposed in Ref. [3] can be multipurpose detectors capable of carrying out a large range of neutrino measurements and dark matter searches. It is also important to have a good understanding of the production mechanisms of radioargon isotopes, notably ^{39}Ar and ^{42}Ar, and devise methods to measure their radioactivity to ultra-low levels.

BACKGROUNDS FROM RADIOACTIVE ARGON ISOTOPES IN RARE-EVENT SEARCHES

Argon is an excellent scintillator with intrinsic properties that enable particularly effective Pulse Shape Discrimination (PSD), which allows efficient background rejection. However, the presence of the long-lived radioactive isotopes ^{39}Ar and ^{42}Ar in argon poses a challenge in large-scale argon-based rare-event searches. Both ^{39}Ar ($Q = 565\,\mathrm{keV}$, $T_{1/2} = 269\,\mathrm{y}$) and ^{42}Ar ($Q = 599\,\mathrm{keV}$, $T_{1/2} = 32.9\,\mathrm{y}$) [4] are β emitters and have half-lives longer than a typical lifetime of experiments. In AAr, the specific radioactivity of ^{39}Ar is $\sim$ 1 Bq/kg [5], while that of ^{42}Ar is $\sim$ 50 µBq/kg [6]. A high rate of ^{39}Ar decays in the bulk argon can cause a high trigger rate and signal-reconstruction issues. ^{39}Ar decays can also be a background in searches for low-mass dark matter because PSD effectiveness is reduced at low energy. ^{42}Ar decays into ^{42}K ($Q = 3525\,\mathrm{keV}$, $T_{1/2} = 12\,\mathrm{h}$). ^{42}K is relatively short-lived and has two major decay channels: i) direct β decay ($Q = 3525\,\mathrm{keV}$) and ii) β decay ($Q = 2001\,\mathrm{keV}$) followed by prompt γ (1524 keV) emission from the resultant Ca isotope. ^{42}Ar has a β-spectrum and endpoint energy similar to that of ^{39}Ar. Given the higher activity of ^{39}Ar in argon compared to that of ^{42}Ar, more problematic backgrounds are from the decays of ^{42}Ar progeny, i.e.^{42}K, which produces energetic β's and γ's that can be a concern to large-scale argon-based neutrino experiments such as LEGEND [7].

GERDA [8] employed a a germanium (Ge) detector immersed in a Liquid-Argon (LAr) veto to search for a ^{76}Ge neutrinoless double beta decay ($0\nu\beta\beta$) signal (2039-keV signal). GERDA found that ^{42}K could drift in the electric field before decaying, and the energetic β's from the ^{42}K isotopes decaying close to the Ge detector posed a serious background. GERDA launched an extensive campaign to mitigate the ^{42}K decay backgrounds. GERDA's successor, the LEGEND experiment [7], could potentially employ UAr to mitigate the ^{42}K decay backgrounds. DUNE [9] [10] will employ tens of kilotons of argon in the Far Detector to carry out a wide range of neutrino measurements. However, ^{42}Ar/^{42}K backgrounds would limit the MeV-scale reach of the experiment, particularly affecting sensitivity to solar-neutrino and supernova-collapse neutrino searches. ^{42}K-decay backgrounds also make it challenging to perform ^{136}Xe $0\nu\beta\beta$ searches by doping the argon with xenon [3, 11, 12].

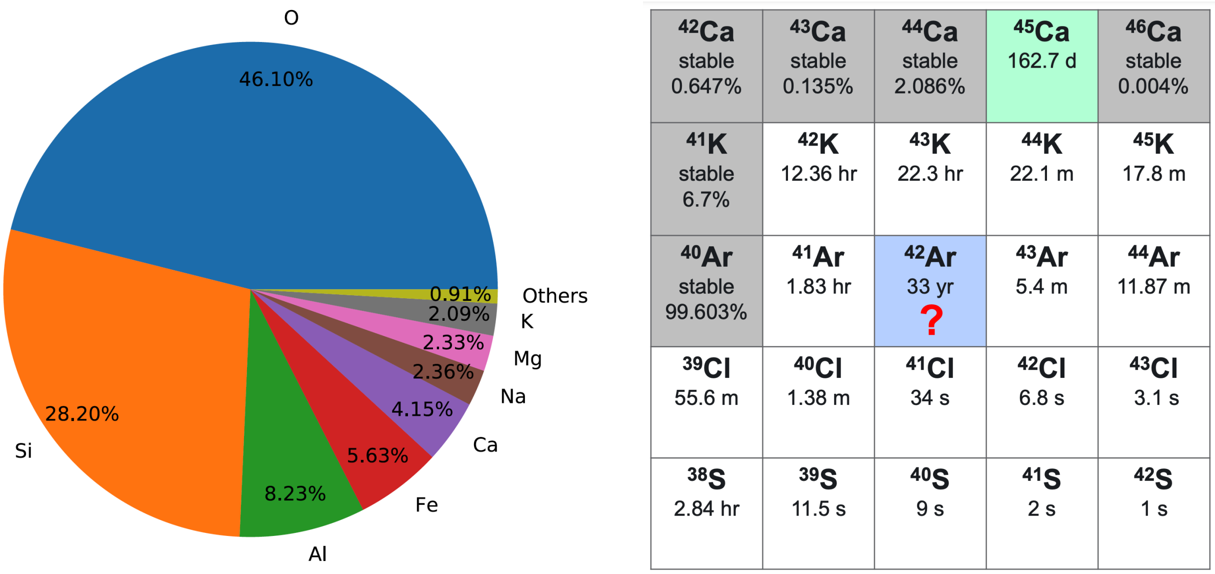
^{39}Ar AND ^{42}Ar PRODUCTION

In the atmosphere, ^{39}Ar and ^{42}Ar are produced primarily by cosmogenic activation on ^{40}Ar. The major channel of ^{39}Ar production is through the ^{40}Ar(n,2n)^{39}Ar reaction [13, 14, 15]. The ^{40}Ar(α,2p)^{42}Ar reaction is a major contributor to ^{42}Ar production in the atmosphere [16]. Two-step ^{40}Ar(n,γ)^{41}Ar, ^{41}Ar(n,γ)^{42}Ar can also produce ^{42}Ar, but its contribution is sub-dominant. Since ^{41}Ar is short-lived ($T_{1/2} = 1.8\,\text{h}$), any significant production through this mechanism requires high neutron fluxes like the ones released in nuclear tests [14, 17]. In AAr, the production of ^{39}Ar and ^{42}Ar is well-understood and their radioactivity well-measured.

^{39}Ar is produced in the earth primarily by muon-capture on ^{39}K and by the ^{39}K(n,p)^{39}Ar reaction [18, 19]. At depths greater than $\sim$ 2 km water equivalent (kmwe), ^{39}Ar production from radiogenic-neutron-induced ^{39}K(n,p)^{39}Ar reactions become dominant [18]. Because ^{39}Ar production by cosmic-ray interactions is suppressed, argon extracted from underground sources is expected to be depleted of ^{39}Ar. While ^{39}Ar production in rock is well-understood, argon diffusion from rocks to gas fields and ^{39}Ar production in the gas fields is not. The DarkSide-50 experiment used UAr extracted from SW Colorado gas wells to search for dark matter and measured its ^{39}Ar activity to be 0.73 mBq/kg [1]. An analysis of other gas species in the argon processing stream suggests that the residual ^{39}Ar may have been due to an air incursion; thus, the ^{39}Ar content could be smaller [20].

There has been little study of ^{42}Ar production underground or the ^{42}Ar content in underground argon [21]. A summary of our study of underground ^{42}Ar production is presented here. For greater details, readers should consult Ref. [22]. We studied ^{42}Ar production in the Earth's crust. The composition of the crust and the elemental abundances in the modelled crust is taken from [23] and shown in Figure 1. The mass-number table in Figure 1 suggests that ^{42}Ar production should be significantly suppressed underground. All of the isotopes directly neighboring ^{42}Ar are short-lived. Our estimate based on TALYS [24] suggests ^{42}Ar production in the earth's crust from particle interactions on those short-lived isotopes is insignificant. Furthermore, production of ^{42}Ar from particle interactions on the closest stable isotopes are suppressed energetically because the reactions have high energy thresholds. The highest flux available for isotope production underground, where cosmic-ray secondaries are reduced, is that of neutrons and alphas that result from decays of radioisotopes in the uranium and thorium decay chains present in the rocks. Neutron- and alpha-yield energy distributions fall sharply above a few MeV. Based on our TALYS-based estimate and recording of isotope production in the simulated crust using particle transport code FLUKA [25, 26], the radiogenic production of ^{42}Ar in the earth's crust is insignificant and likely subdominant to cosmogenic production up to large crustal depths.

Cosmic-ray muons that reach underground have a hard energy spectrum extending to hundreds of GeV. Therefore, the energetic collisions from the cosmic-ray muons and the cosmic-ray muon-induced secondary particles can overcome the high thresholds to produce ^{42}Ar in the earth's crust. Our study shows that a major contributor to the ^{42}Ar production in the earth's crust is the interaction of cosmic-ray muon-induced neutrons and heavy-ion collisions on ^{44}Ca. This is primarily because calcium is relatively abundant (4.15% by a mass fraction) in the earth's crust and the isotopic abundance of ^{44}Ca is 2.06%. Based on our study, at 3 kmwe depth in the crust, the ^{42}Ar production rate is 10^{-3} atoms per ton of crust per year. For comparison, at 3 kmwe depth for similar crust composition, the ^{39}Ar production rate is $\sim 10^4$ atoms per ton of crust per year [18, 19].



^{42}Ca stable 0.647%	^{43}Ca stable 0.135%	^{44}Ca stable 2.086%	^{45}Ca 162.7 d	^{46}Ca stable 0.004%
^{41}K stable 6.7%	^{42}K 12.36 hr	^{43}K 22.3 hr	^{44}K 22.1 m	^{45}K 17.8 m
^{40}Ar stable 99.603%	^{41}Ar 1.83 hr	^{42}Ar 33 yr ?	^{43}Ar 5.4 m	^{44}Ar 11.87 m
^{39}Cl 55.6 m	^{40}Cl 1.38 m	^{41}Cl 34 s	^{42}Cl 6.8 s	^{43}Cl 3.1 s
^{38}S 2.84 hr	^{39}S 11.5 s	^{40}S 9 s	^{41}S 2 s	^{42}S 1 s

FIGURE 1. Left: The elemental abundance in the continental crust as considered from [23]. **Right:** The table showing isotopes neighboring ^{42}Ar with respect to mass number. Only very short-lived isotopes (white) directly neighboring ^{42}Ar (light blue), one long-lived (light green) and several stable isotopes (dark grey) are in the table, with ^{44}Ca most important for production of ^{42}Ar.

LOW-RADIOACTIVITY ARGON FOR LARGE-SCALE ARGON EXPERIMENTS

The use of low-radioactivity argon extracted from underground sources would benefit several experiments, as presented in Table I. For instance, in the case of DUNE, the use of UAr could expand the physics reach to the MeV scale. Further, using UAr in the LEGEND veto would eliminate background events from ^{42}K decays in their $0\nu\beta\beta$ searches. In addition, a multipurpose detector capable of exploring both dark matter and neutrino physics (such as the one discussed in [3]) could be realizable using kton-scale underground argon (UAr).

However, the only known source for UAr is the CO$_2$ gas wells in SW Colorado, used by the DarkSide-50 experiment for dark matter searches [1]. The URANIA [27] plant will produce crude UAr (with 99.9% purity) from the gas wells at a rate of $\sim$300 kg of argon per day. The GADMC collaboration plans to use the UAr for DarkSide-20k and its proposed 300-ton successor ARGO. The crude UAr obtained from the URANIA plant will be transported to Sardinia, Italy where the ARIA plant will further purify and isotopically enrich the UAr. A dedicated detector DArT [28] will measure the ^{39}Ar depletion factor of the UAr.

^{40}Ar can be produced underground by electron capture on ^{40}K ($T_{1/2} = 1.25 \times 10^9$ y) [4]. The ^{40}K isotopic abundance is 0.117%. The ^{40}Ar/^{36}Ar ratio has been used as a tool for geochronology and for understanding

TABLE I. Experiments that benefit from the use of UAr for measurements of WIMP dark matter (DM), neutrinoless double-beta decay ($0\nu\beta\beta$), coherent elastic neutrino-nucleus scattering (CEνNS), or neutrino measurements.

Experiment	Physics	Argon use	Ar-related backgrounds	Required argon mass
DarkSide-20K [27]	DM	Target material	^{39}Ar	50 tons
ARGO [2]	DM	Target material	^{39}Ar	300–500 tons
LEGEND [7]	^{76}Ge $0\nu\beta\beta$	Veto and Shield	^{42}Ar/^{42}K	$\sim$ 10 tons
Coherent CAPTAIN-Mills [29]	CEνNS	Target material	^{39}Ar	$\sim$ 10 tons
DUNE [10]	neutrinos	Target material	^{39}Ar, ^{42}Ar/^{42}K	$\sim$ 40 kilotons

geochemical processes. Its ratio in the atmosphere is 296 [30]. Argon of mantle origin has been found to have a significantly higher ratio [31]. The bulk of the atmospheric argon originated from mantle degassing [31]. There is an abundance of argon underground. Identifying the argon-rich gas streams and commercially extracting the argon on a massive scale can make kiloton-scale UAr-based experiments such as the one discussed in Ref. [3] feasible.

A major challenge is to identify and procure UAr in time to benefit next-generation argon experiments. In addition, cost-effective and commercial production would be desirable. Initial findings reported in Ref. [3] suggest commercial production of UAr with a reasonable cost is possible. Measurement of radioactivity levels of ^{39}Ar and ^{42}Ar in the argon of underground origin is vital to ensure if commercial production and use in detectors are viable. Monitoring of the radioactivity levels is also necessary during extraction, transport, storage, and deployment. Addressing some of these challenges requires a facility like the one discussed in Ref. [32].

CONCLUSION

The use of low-radioactivity argon can expand the sensitivity and physics reach of several rare-event search experiments. The DarkSide-50 measurements and the study of underground ^{39}Ar production suggest argon derived from underground sources is depleted of ^{39}Ar. Based on our study, ^{42}Ar production is also significantly suppressed underground. Identifying argon-rich underground gas streams and producing argon commercially can be a significant step in realizing a multipurpose neutrino and dark matter experiment like the one discussed in [3]. Major challenges are procuring underground argon on a large scale and timely manner for next-generation experiments and devising techniques and/or building infrastructure to measure ultra-low level radioactivity levels in argon.

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