Published by IOP Publishing for Sissa Medialab

Received: *December 9, 2023*
Accepted: *May 21, 2024*
Published: *June 17, 2024*

Light Detection In Noble Elements (LIDINE 2023)
Madrid, Spain
20–22 September 2023

The DarkSide-20k underground argon procurement chain

V. Cocco on behalf of the DarkSide-20k collaboration

INFN Cagliari,
09042 Cagliari, Italy

E-mail: valentina.cocco@ca.infn.it

Abstract: The DarkSide-20k experiment searches for dark matter by looking for interactions of WIMPs in a 50-tonnes target of liquid argon using double-phase time projection chamber (TPC) technology. The key component of the experiment is low radioactivity argon depleted in the isotope ^{39}Ar. Unfortunately, ^{39}Ar is naturally present in atmospheric argon and it is constantly produced due to the interaction with cosmic rays. Finding a source of depleted argon was the first step of the Collaboration when it started to size the DarkSide-20k detector.

The procurement chain begins with the Urania plant in Colorado, which can produce argon with a purity of 99.99% from a crude CO_2 stream extracted from a deep well, at a rate of about 250 kg per day. The plant has already been fabricated while the site is being prepared for installation. After the extraction and purification of 120 t of underground argon (UAr), it will be transported to Sardinia, Italy, where the ARIA plant, consisting of a 350 m cryogenic distillation column, will further remove impurities up to 2 orders of magnitude. The final purity is expected to be 99.9999%. After the ARIA purification stage, the ultra-pure UAr will be delivered to the Laboratori Nazionali del Gran Sasso, L'Aquila, Italy where it will fill the DarkSide-20k TPC.

The ARIA plant has already been fully fabricated and is now in the installation phase in a mine shaft. A shorter version, about 26 m high, has been tested over the last three years with very positive results. We were able to measure the separation factor between two isotopes of nitrogen ($^{29}N_2$, $^{28}N_2$) and three of argon (^{40}Ar, ^{38}Ar, ^{36}Ar) thus validating the capability of performing isotopic distillation with ARIA.

Keywords: Detector design and construction technologies and materials; Gas systems and purification

https://doi.org/10.1088/1748-0221/19/06/C06019

2024 JINST 19 C06019

Contents

2024 JINST 19 C06019

1 Introduction

The Global Argon Dark Matter Collaboration (GADMC) is advocating for the development of new experiments to search for dark matter based on liquid argon (LAr) detectors. One of the most promising experiments is DarkSide-20k, which is designed to search for Weakly Interacting Massive Particles (WIMPs) by direct detection via WIMP-nucleus elastic scattering in LAr [1]. DarkSide-20k poses its base in previous experiments, the most important is DarkSide-50, a two-phase LAr detector that obtained very interesting results [2].

Argon is the third-most abundant gas in Earth's atmosphere, comprising roughly 0.93% of the atmosphere by volume. Atmospheric argon (AAr) consists primarily of the stable isotopes ^{40}Ar, ^{36}Ar, and ^{38}Ar. However, because of the interactions with cosmic rays, AAr also contains three radioactive isotopes: ^{39}Ar, ^{37}Ar, and ^{42}Ar.

Even if the concentration of ^{39}Ar is considerably low, this isotope is a beta-emitter whose activity of about 1 Bq kg^{-1} raises background and pile-up concerns [2]. It has been found that argon extracted from some underground wells, called Underground Argon (UAr), has a greatly reduced ^{39}Ar content and is therefore pivotal to the physics potential of dark-matter search experiments. A CO_2 reservoir where the concentration of ^{39}Ar is estimated to be 1400 times lower than in AAr has been discovered in Colorado; the UAr procurement chain starts from there. The Urania plant will first separate UAr from the crude CO_2 stream, then the UAr will be delivered to Sardinia, Italy, where a second purification step will be performed by the ARIA plant, further improving the quality of the UAr. Finally, the ultra-purified UAr will be sent to the Laboratori Nazionali del Gran Sasso (LNGS), L'Aquila, Italy, where the DarkSide-20k time projection chamber (TPC) is being constructed. A total amount of 120 tonnes of argon is going to be extracted and purified.

A significant effort will be dedicated to UAr transportation and storage tasks, as improper storage practices could cause a dramatic reduction in Ar purity.

All along the UAr procurement chain, the concentration of ^{39}Ar will be measured by another experiment, DArT. It has been constructed in Laboratorio Subterraneo de Canfranc (LSC), Canfranc, Spain, and will ensure high-quality standards.

Table 1. Stable and long-lived isotopes of argon, along with their typical abundances and specific activity in atmospheric argon [3, 4].

Isotope	Mole fraction	Specific activity (Bq/kg_{Ar})
^{40}Ar	0.9960	Stable
^{36}Ar	0.0033	Stable
^{38}Ar	0.0006	Stable
^{39}Ar	8.2×10^{-16}	1.0
^{37}Ar	$\approx 1.3 \times 10^{-20}$	$\approx 4.5 \times 10^{-2}$
^{42}Ar	6.8×10^{-21}	6.8×10^{-5}

2 Argon characteristics

In table 1 the abundance and the activity of each isotope have been reported for AAr [3, 4].

In the atmosphere, ^{39}Ar is mostly produced by interactions with cosmic rays, therefore it is expected that argon from underground sources would have less cosmogenic ^{39}Ar radioactivity. Unfortunately, ^{39}Ar can be also produced underground by a sequence of nuclear reactions starting from the decay of uranium and thorium. Because the concentration of these two elements is at the ppm level for the Earth's crust and ppb level for the Earth's mantle, it would be reasonable to search for deep underground sources of argon that come directly from the Earth's mantle [5]. A natural CO_2 reservoir, where most of the CO_2 has mantle origin, was discovered in Doe Canyon, Colorado (U.S.A.). Among all the analysed samples, the Doe Canyon sample showed the highest ^{40}Ar concentration (33.1×10^{-4} cm^3 $_{STP}/cm^3$) [6]. For this underground sample, the most important fact that was discovered is that the calculated ^{39}Ar activity corresponds to a reduction factor of 1400 relative to AAr [7, 8].

The oil company Kinder Morgan operates the Doe Canyon Deep unit, producing CO_2 that is used for oil drilling in New Mexico and Texas. A new extraction plant called Urania has been designed and it is going to be installed in Doe Canyon, beside the Kinder Morgan facility. The plant will collect the CO_2 gas stream coming from the Kinder Morgan pipeline, separate argon from the gas stream and finally send back the residual flow to Kinder Morgan.

3 Urania facility

The UAr procurement chain starts from the Urania plant, Cortez, CO, U.S.A. Urania is the first extraction and chemical purification plant of UAr. The initial concentration of Ar in the inlet CO_2 stream has been measured to be around 440 ppm. The design recovery fraction will be 95% or higher. The ultimate goal of this plant is to purify UAr at a rate of at least 250 kg d^{-1} (330 kg d^{-1} max) from the CO_2-rich gas stream coming from Kinder Morgan deep wells and purify the UAr to a purity of 99.99%. The plant will operate continuously 24/7.

The plant is quite complex, and it is schematically shown in figure 1. It has two separate sections: a first section for the separation of a raw argon current from the crude CO_2 gas and a second argon purification unit, based on cryogenic distillation technology, designed to reach the required purity. Some more details about cryogenic distillation technology will be given in section 4.

2024 JINST 19 C06019

All the components of the plant have already been manufactured and shipped to a warehouse in Houston. The plant was fully built in Italy by an Italian company Polaris.

Although the purity of UAr obtained by Urania is quite high, for the DarkSide-20k experiment the concentration of the contaminants needs to be at the ppm level or even lower. For this reason, the UAr need to be further purified by the ARIA plant.

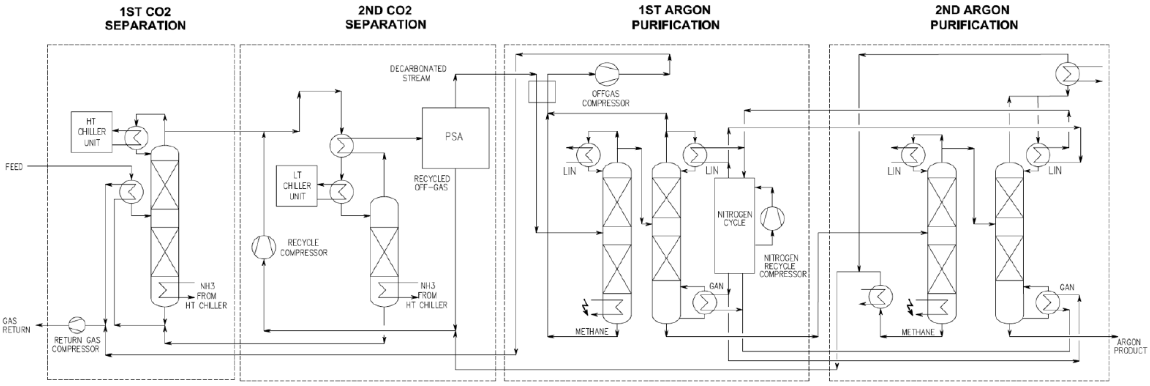

Figure 1. Schematic of Urania plant; from the left to the right the first two stripping columns working at different temperatures, the 1^{st} cryogenic distillation unit and the 2^{nd} cryogenic distillation unit.

4 ARIA plant, state of the art

The cryogenic isotopic distillation plant ARIA was designed to further reduce the ^{39}Ar isotopic fraction of UAr by a factor of 10 per pass, with a production rate of few kg d^{-1}. The column is currently in the installation phase in a mine shaft at Carbosulcis S.p.A., in Nuraxi-Figus, Sardinia, Italy.

Distillation columns separate different components from a fluid based on their different boiling points. For ideal mixtures, the relative volatility is given by the ratio of the components' vapour pressures at a given temperature. Figure 2 shows the concept of a distillation column: heat is constantly provided from a bottom heat exchanger, called the reboiler, that vaporizes the liquid. On the other side, heat is constantly extracted from a top heat exchanger, called the condenser, that condenses vapour. Therefore, the liquid drops down and the vapour raises up in a counter-current exchange at thermodynamic equilibrium. Saturation conditions are kept stable by controlling the pressure (or the temperature) inside the column. Due to the physical characteristics of Ar, to maintain the column pressure in a range of 1–2 bar, the saturation temperatures must be between 87–94 K. For this purpose, LN is used as the refrigerant in the auxiliary system.

In the case of isotopic distillation, the elements we try to separate are isotopes, thus the difference in volatility is very small and it makes the separation process harder. For a two-component distillation at total reflux, i.e. without a significant amount of argon entering or leaving the column during distillation, the separation between isotopes i and j between the top (T) and the bottom (B) of the column is given by:

$$S_{i-j}^{TB} = (\alpha_{i-j})^N$$

where α_{i-j} is the relative volatility between the lighter more volatile isotope i and the heavier less volatile isotope j, and N is the number of theoretical stages. Ideally, N represents the number of theoretical states of equilibrium along the column. The logarithm of the relative volatility, is given by:

$$\ln \alpha_{i-j} \cong \ln \frac{P_i}{P_j}$$

with P_i and P_j the vapour pressures of the two isotopes i and j. During the design phase, the number of theoretical stages (N) of 2870 has been conservatively obtained. Using the structured stainless-steel packing (Sulzer CY gauze) which filled the ARIA column, a total active height of about 287 m is expected. Given the packing installation requirements, a total column height of around 350 m has been set. For more details about the ARIA column see ref. [9].

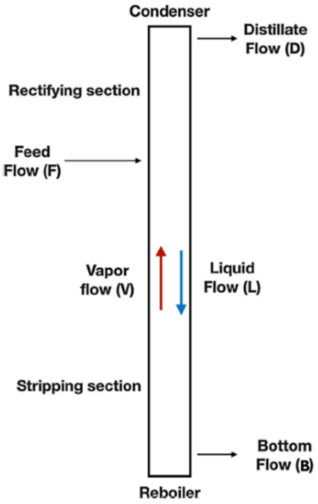

Figure 2. Basic operation principles of a continuous distillation column.

After years of research, it is going to be assumed that the calculated activity of ^{39}Ar in UAr, done by DarkSide-50, can be acceptable also for DarkSide-20k, therefore the ARIA column will be used only to enhance the chemical purity of the UAr treated by the Urania plant with no intention to deeply remove ^{39}Ar from UAr. Based on this hypothesis, the distillation process can speed up to $1\,\mathrm{t\,d^{-1}}$ because the calculation considers the boiling points of the residual gases remaining after the Urania purification (mainly N_2). The achieved purity after the ARIA plant will be 99.9999%.

However, since ^{39}Ar is mostly produced by cosmogenic activation, both transportation and storage times need to be carefully taken into consideration. Cosmogenic activation has to be kept under control by minimizing exposure on the surface and storing materials underground, avoiding flights, and even using appropriate shields [13, 14]. The time spent by the UAr at sea level or above sea level must be the shortest possible.

5 ARIA installation site

The ARIA column, which is 350 m high, presented significant challenges. A metal structure as tall as the Eiffel Tower would have been constructed to house the column above the ground level. It would have posed serious environmental and architectural arguments. Almost all of these problems have been solved by installing the column underground. For this reason, an agreement with the Carbosulcis mine was signed in 2015.

Carbosulcis is the last coal mine in Italy, and in December 2018 they stopped coal production due to European directive. However, the entrance to the mine is still guaranteed by four shafts (ranging from 350 to 500 meters deep) and one underground road. Below the ground level, the mine has a network of 30 km of tunnels. One of the shafts, Seruci, is going to host the ARIA column. It has a depth of around 350 m, a diameter of around 5 m, a brick shaft lining of around 40 cm and the bottom of the shaft is placed at −200 m above mean sea level. The Seruci shaft was built in the 1940s, and a lot of work has been done to make it suitable for the installation of the cryogenic column. The column is made of 28 identical central modules (3 t each). Every module has 3 support platforms, each platform is anchored to the ground by 4 concrete-embedded beams. Up to now, 23 platforms out of 87 have already been installed inside the shaft.

On March 15th, 2021, the first module placement test inside the shaft was fulfilled thanks to the effort of many people, both INFN and Carbosulcis. The test was a success. The module, which is 12 m high and has a diameter of 70 cm, was lifted down for some meters inside the shaft and then sheltered again in the warehouse at the end of the test.

6 ARIA protype plant — Seruci 0

While the installation of the 350 m column is proceeding, the Seruci 0 plant, a cryogenic distillation column 26 m tall, is fully installed and operative in a Carbosulcis surface building. It consists of three ARIA column parts: the reboiler, the condenser, and one central module (out of 28), together with all the auxiliary equipment of the final column. Additional details about this column can be found in [10].

A nitrogen isotope separation campaign was conducted with Seruci 0 in 2019 and then in 2021, another test was conducted with argon. In both cases, despite the short active height of Seruci 0, we observed isotopic separation. The main results of the first test are reported in [9]. Figure 3 shows the separation factor S_{28-29} for $^{29}N_2$-$^{28}N_2$, it grew after the start-up phase, and it reached approximately a value of 1.25 during the first run and 1.30 during the second, showing how the light isotope concentration increased at the top and decreased at the bottom and vice versa for the heavier isotope.

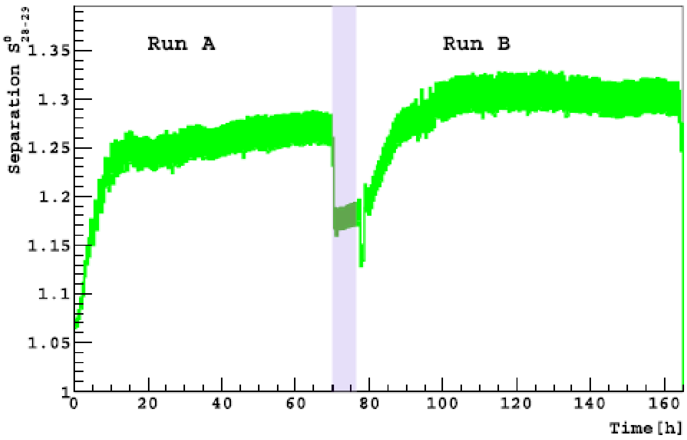

Figure 3. Separation factor S_{28-29} for $^{29}N_2$-$^{28}N_2$. The vertical grey band identifies a time slot where non-relevant data have been acquired (no stable conditions). Run A and Run B correspond to two different setups of the auxiliary system.

2024 JINST 19 C06019

In 2021, the second Seruci 0 test was performed with argon; for all the details see ref. [10]. Figure 4 shows the separation factor for ^{40}Ar-^{36}Ar (S_{36-40}), it reaches a value of around 1.5. Worst separation is reached for ^{40}Ar-^{38}Ar ($S_{38-40} = 1.2$) showing how the separation phenomenon gets harder as the relative volatility of the species tends to 1. These tests also confirmed the ARIA column flexibility, and the opportunity to use ARIA also for commercial applications, i.e. isotopic enrichment for medical applications.

The instrument used to measure the isotopic distillation performance is an MKS Instruments, Cirrus™ 3-XD quadrupole mass spectrometer. The mass spectrometer can clearly identify ^{36}Ar, ^{38}Ar, and ^{40}Ar peaks, but the concentration of ^{39}Ar is much lower than its detection limit (15 ppb). For this reason, radioactive measurements will be carried out with another experiment: DArT at LSC [11, 12]. This experiment is meant to verify the quality standards of the whole UAr procurement chain. It will analyse samples coming from Urania, ARIA, and DarkSide-20k TPC.

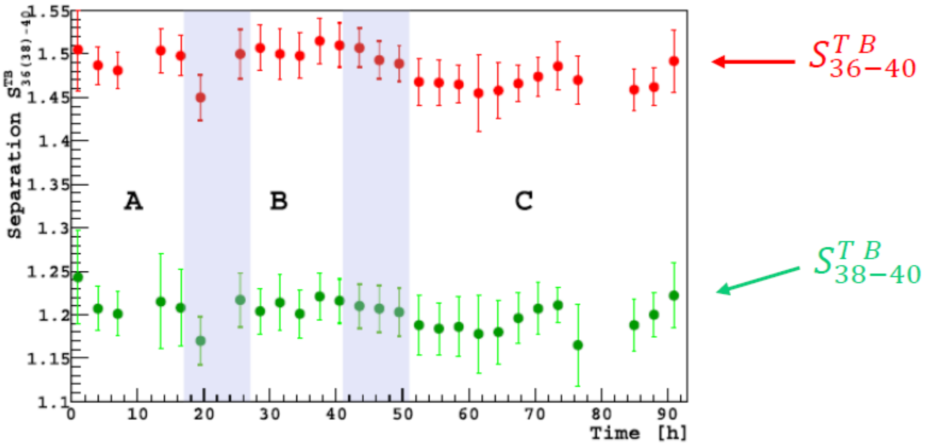

Figure 4. Separation factor S_{36-40} for ^{40}Ar –^{36}Ar and S_{38-40} for ^{40}Ar –^{38}Ar. The vertical grey bands identify two slots of time where non-relevant data have been acquired (no stable conditions). Run A, B, and C correspond to different pressure conditions inside the column.

7 Conclusions

The DarkSide-20k experiment aims to search evidence for dark matter. To improve the sensitivity for WIMP research, a TPC with 50-ton fiducial volume of depleted UAr is being designed. The UAr procurement chain begins in Colorado where UAr extracted from the Doe Canyon Deep Unit has an activity of ^{39}Ar 1400 times lower than in AAr. 120t of UAr will be treated by the Urania plant reaching a purity of 99.99%. Afterwards, UAr will be sent to ARIA to be chemically purified by cryogenic distillation. The ultra-pure UAr (at least 99.9999%) is finally sent to LNGS where will fill the DarkSide-20k TPC. To date, since ARIA will be run only with the intention to remove the content of chemical pollutants, such as N_2, and not to reduce the content of ^{39}Ar, great attention must be paid to transportation and storage times in order to minimise cosmogenic activation of purified UAr.

However, the tests performed with the Seruci 0 plant demonstrate the capability of doing isotopic distillation even with a very short column. Scaling it up to 350 m will allow to improve this capability, reaching high grades of purity also for chemical species having α very close to one.

References

[1] M. Schumann, *Direct Detection of WIMP Dark Matter: Concepts and Status*, *J. Phys. G* **46** (2019) 103003 [arXiv:1903.03026].

[2] DarkSide collaboration, *Results From the First Use of Low Radioactivity Argon in a Dark Matter Search*, *Phys. Rev. D* **93** (2016) 081101 [*Addendum ibid.* **95** (2017) 069901] [arXiv:1510.00702].

[3] R. Saldanha et al., *Cosmogenic production of ^{39}Ar and ^{37}Ar in argon*, *Phys. Rev. C* **100** (2019) 024608 [arXiv:1902.09072].

[4] J.K. Böhlke, *Variation in the terrestrial isotopic composition and atomic weight of argon (IUPAC technical report)*, *Pure Appl. Chem.* **86** (2014) 1421.

[5] J. Xu et al., *A Study of the Residual ^{39}Ar Content in Argon from Underground Sources*, *Astropart. Phys.* **66** (2015) 53 [arXiv:1204.6011].

[6] S.M.V. Gilfillan et al., *The noble gas geochemistry of natural CO2 gas reservoirs from the Colorado Plateau and Rocky Mountain provinces, USA*, *Geochim. Cosmochim. Acta* **72** (2008) 1174.

[7] DarkSide collaboration, *DarkSide-50 532-day Dark Matter Search with Low-Radioactivity Argon*, *Phys. Rev. D* **98** (2018) 102006 [arXiv:1802.07198].

[8] DarkSide collaboration, *Low-Mass Dark Matter Search with the DarkSide-50 Experiment*, *Phys. Rev. Lett.* **121** (2018) 081307 [arXiv:1802.06994].

[9] DarkSide-20k collaboration, *Separating ^{39}Ar from ^{40}Ar by cryogenic distillation with Aria for dark-matter searches*, *Eur. Phys. J. C* **81** (2021) 359 [arXiv:2101.08686].

[10] DarkSide-20k collaboration, *Measurement of isotopic separation of argon with the prototype of the cryogenic distillation plant Aria for dark matter searches*, *Eur. Phys. J. C* **83** (2023) 453 [arXiv:2301.09639].

[11] DarkSide-20k collaboration, *Design and Construction of a New Detector to Measure Ultra-Low Radioactive-Isotope Contamination of Argon*, 2020 *JINST* **15** P02024 [arXiv:2001.08106].

[12] D.D. Mairena, *Monitoring ^{39}Ar Background for DarkSide 20 k with DArTinArDM*, in the proceedings of the *Light Detection In Noble Elements (LIDINE 2023)*, Madrid, Spain, 20–22 September 2023.

[13] DarkSide-20k collaboration, *Study of cosmogenic activation above ground for the DarkSide-20k experiment*, *Astropart. Phys.* **152** (2023) 102878 [arXiv:2301.12970].

[14] S. Cebrian, *Study of cosmogenic activation above ground of Ar for DarkSide-20k*, 2024 *JINST* **19** C02011 [arXiv:2312.02183].

2024 JINST 19 C06019