

Distillation and separation of some rare isotopes and their applications

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received 21 April 2018

Summary. — We present two techniques related to the production of rare isotopes and their consequent application fields. The first is related to the Darkside Experiment that makes use of liquid argon as a sensible target for dark matter detection. In order to get an ultrapure and low radioactivity target, the argon will be purified in a 350 m tall distillation column placed in the Seruci mine in Sardinia (Aria project). Such a column will be also able to distillate rare and very valuable isotopes as ^{18}O , ^{15}N , ^{13}C used, for instance, in medical diagnosis. The second is about the $^3\text{He}/^4\text{He}$ separation via an inverse osmosis process that may take place at a temperature below 1.5 K. It provides the very rare and valuable ^3He isotope, used in various research fields as MRI lung screening, neutron detectors and in all the experiments in physics where a temperature below 1 K is required.

1. – The Darkside experiment

The Darkside experiment [1] searches for dark matter with a direct search method using liquid argon as target and using a powerful discrimination method against the background. It is located in the underground Laboratori Nazionali del Gran Sasso (LNGS) and is a worldwide new research program using liquid argon, as all the research groups have joined the new 20 ton scale experiment (DS-20k) aiming to a total exposure of 100 ton year and a further multi-100 ton scale experiment has been already anticipated. 68 research institutes and universities will take part to such a collaboration, more than 350 researchers, engineers and technicians, and 12 countries (Brazil, Canada, China, France, Greece, Italy, UK, Poland, Romania, Spain, Switzerland, USA). The Darkside experiment employs a double phase liquid argon Time Projection Chamber (TPC) for the WIMP (Weakly Interacting Massive Particles) search, where two signals are acquired contemporarily after each event: the scintillation signal, produced in the argon itself by the incoming particle, and the ionization signal obtained by accelerating the electrons (produced together with argon ions by the same particle) with an electric field of 200 V/cm towards the argon gas pocket on top of the chamber. Here the electrons are extracted and further accelerated with a higher field (3 kV/cm) and therefore produce electro-luminescence; the emitted light is collected by photosensors (*e.g.*, PMTs or

SiPM) and the resulting read-out signal is proportional to the ionization signal. The TPC is capable of 3-dimensional event reconstruction with cm-like precision on the xy plane (perpendicular to gravity) and mm-like precision on the z axis. The powerful background rejection is provided by the Pulse Shape Discrimination (PSD) of the scintillation signal (S1) and by the ratio between ionization and scintillation signals, $S2/S1$. The experiment operates with an active water Cherenkov muon veto and a liquid scintillator neutron veto surrounding the TPC. The goal is to reach, for the future experiments, a data taking campaign in instrumental background free mode, *i.e.* less than 0.1 background event in the total exposure. Liquid argon has several appealing properties as a dark matter target: it liquifies at 87 K simply using liquid nitrogen or cryogen free techniques; contaminants and impurities may be easily trapped (*e.g.*, radon); it may be scaled to larger masses; it has a sufficiently large atomic mass number A (the WIMP-nucleon cross section scales as A^2 for spin-independent interaction); it scintillates with high scintillation yield ($40 \text{ k}\gamma/\text{MeV}$) and is transparent to the emitted light; it has a high ionization signal (electroluminescence in argon gas); it provides an excellent discrimination power (PSD) through the different de-excitation times of the argon dimers produced by nuclear recoils (dimers in singlet state and decay time of about 6 ns) and by electron recoils (dimers in triplet state and decay time of about $1.6 \mu\text{s}$). The use of liquid argon has only one drawback: the cosmogenic production of the unstable isotope ^{39}Ar in the atmospheric argon (AAr) via the reaction $^{40}\text{Ar}(n, 2n)^{39}\text{Ar}$. The solution for this problem is the use of underground argon (UAr) from deep underground wells, located in Cortez, Colorado. The measured depletion factor of ^{39}Ar in UAr with respect to the AAr is $(1.4 \pm 0.2) \times 10^3$. Such a factor has been measured by the current Darkside experiment DS-50, which is presently running with UAr in background free mode. As already mentioned, the next step will be DS-20k, with a fiducial volume of 20 ton of liquid argon and next Argo (Argon Observatory) with a fiducial volume of 200 ton. The acquired data and subsequent Monte Carlo Simulations show that DS-20k will be able to run in background free mode for the total exposure of $100 \text{ ton} \times \text{year}$ with this depletion factor. As for the Argo experiment, with a total exposure of $1000 \text{ ton} \times \text{year}$ or more, a further depletion will be necessary.

2. – Aria project: Ar process and isotopes production

The need of chemically purifying the underground argon for DS-20k and to further deplete it for Argo are the motivations for the construction of the Aria project, a very tall distillation column capable of separating the two argon isotopes ^{39}Ar and ^{40}Ar . The underground argon will be extracted from the CO_2 wells of the Doe Canyon, located at Cortez, Colorado and shipped to Seruci, in Sardinia, where the Aria project will be constructed. The distillation column has an operating temperature of about 87 K and will exploit the small volatility difference of the two isotopes for their separation. Beyond the capability of separating argon isotopes, the Aria distillation column will provide a great separation power also for some rare stable isotopes with important applications in nuclear medicine and in some industrial fields (as oil industry and nuclear power plants). As examples of medicine application we may consider: ^{13}C labeled urea used in breath tests to detect helicobacter pylori infection; ^{13}C for studying metabolic changes in the brain by MRI to diagnose neuropsychiatric disorders; study of metabolic transformations of drugs in pharmaceutical industry using ^{13}C , ^{15}N , ^{17}O , ^{18}O . Such isotopes will be produced by Aria, entering a market now constrained by supply and where the costs are dominated by the energy required for their separation.

3. – Application fields of helium-3 isotope and the SOPHIE project

Before describing the application fields of the ^3He isotope there are some relevant aspects we should first consider. ^3He is present in traces (between 0.5 and 10 ppm) in natural helium, mostly made of ^4He . It is presently obtained from the ^3H β -decay which is produced for the nuclear weapon programs. Therefore its availability is very limited and its cost is very high. We may say that there is presently a lack of a suitable nuclear-independent technology capable of satisfying the market demand.

Given its very large cross section for neutron capture, ^3He -filled proportional counters are the best performing detectors for neutrons. They are very effective for monitoring ports and terminals to prevent possible rogue smuggling of nuclear materials. Their increase in use may prevent possible nuclear terror threats, this being one of the major concerns on National Security nowadays, as reported by the former US president Obama at the 2016 International Nuclear Security Summit.

^3He can be hyper-polarized and, upon inhalation by patients, its distribution in lungs can be detected through an advanced Magnetic Resonance Imaging (MRI) scanner to produce extremely detailed anatomical and functional imaging of lung ventilation, characterized by their unprecedented precision. This tool for medical diagnostics was developed twenty years ago and has since been held back from large scale deployment due to the lack of availability of the ^3He isotope. It provides early and specific detection of metastatic cancer cells and other lung diseases.

Dilution refrigerators (DR) and ^3He refrigerators allow to reach temperatures below 1 K and down to few mK. DR are the only systems able to reach very low temperatures (2 mK) for an indefinite period of time. The high cost of ^3He makes all these refrigerators even more expensive. They are widely used in physics in several fields as semiconductors (Quantum Hall effect, quantum dots, Single Electron Tunneling), superconductors (Quantum computing, Josephson Junctions, Flux vortices), solid state physics (heavy fermions systems, metal insulator transition, spin glass, mesoscopic systems, giant magnetoresistance, nanoelectronic primary thermometry). Another very interesting application field is astroparticle physics, where cryogenic detectors using transition edge sensors [2, 3] are widely used for dark matter experiments as CRESST, Edelweiss and SuperCDMS [4] and magnetic microcalorimeters are deployed for the neutrino direct mass measurement, as in ECHO [5].

Future applications include also the use of ^3He as fuel for 2nd and 3rd generation nuclear fusion plants using the ^3He - ^3He (D- ^3He) cycle that does not (almost) generate neutrons, whereas the D-T fusion wastes about 80% of the energy in neutrons, which are also difficult to contain.

The SOPHIE project aims at the separation of ^3He from natural helium through an inverse osmosis process provided by thermo-mechanical pumps at a temperature below 2 K. With a working temperature of 1.35 K it is possible to separate ^3He and ^4He in a continuous mode, at a fast rate and with much lower costs than the present ones, and get a 95% purity ^3He with 5% of ^4He and less than 0.01% of impurities. For all the above mentioned applications, such composition is perfectly suitable. It is still possible with the same system to produce ^3He at a 99% purity level (1% of ^4He) if required by other particular applications.

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