Material Assay Campaign of the DarkSide-20k Experiment

Vicente Pesudo[a)] for the DarkSide-20k Collaboration

Centro de Investigaciones Energéticas Medioambientales y Tecnológicas, Av. Complutense 40, 28040 Madrid, Spain
Laboratorio Subterráneo de Canfranc, P. de los Ayerbe 22880 Canfranc-Estación, Spain

[a)] *Corresponding author: vicente.pesudo@ciemat.es*

Abstract. The DarkSide-20k experiment will search for dark matter in the form of WIMPs and has the potential to set the best limits for the spin-independent interaction of heavy WIMPs with nucleons (90% CL for $\sigma_{\mathrm{SI}} = 10^{-48}$ cm at WIMP mass $\mathrm{M}_\chi = 100\,\mathrm{GeV}/c^2$). The requirement for backgrounds of radiogenic origin is fewer than 0.1 events in 10 years of exposure. Such a stringent goal has not been achieved to date and imposes the establishment of rigorous requirements in terms of radiopurity of the detector materials. A thorough assay campaign has been running for five years to assess the radiopurity of candidate components, paying particular attention to the uranium and thorium decay chains. Different assay techniques are exploited: ICP-MS is sensitive to the chain precursors (^{232}Th and ^{238}U); HPGe to the gamma emitters through the chain; Po radiochemical separation to the often-ignored ^{210}Po content in the bulk of the materials. In such a way, it is possible to have accurate estimates of the background accounting for the breaking of the secular equilibrium of the uranium decay chain in all the materials. Beyond its common application, mass spectrometry is used for assessing the overall chemical composition of critical items, focusing on the quantification of elements with high (α,n) cross section, hence minimizing the uncertainty in the neutron yield. In this work, the organization of the assay campaign and its results to date are presented.

INTRODUCTION

DarkSide-20k is an experiment currently starting the construction phase at Laboratori Nazionali del Gran Sasso (LNGS) that has a program for ten years of data taking [1]. Within its rich scientific reach, its main goal is looking for dark matter in the form of weakly interacting massive particles (WIMPs). Direct WIMP searches are needle-in-a-haystack experiments, where a rare event is sought in a pool of events induced by known radiation.

This radiation has two origins: cosmogenic and radiogenic. Cosmogenically induced background is efficiently mitigated by locating experiments in deep underground laboratories [2] and using muon vetos. Radiogenic backgrounds are the consequence of all the materials on Earth having a certain amount of radioactivity. This activity is caused by a combination of astrophysics (formation of the planet in a particular location and moment), nuclear physics (processes actually producing such isotopes and letting them evolve during Earth's history), cosmic rays (the flux of in-falling high-energy radiation), and geology (dynamics of the material in the Earth's crust). The amount of parameters playing a role is thus large and translates into a variability in the activity that needs to be controlled. The procedure to do so is performing assays on the radioactivity of each candidate component to go into the detector.

DARKSIDE-20K

The main detector of DarkSide-20k is a dual-phase time projection chamber (TPC). The target will be 50 tonnes of liquid argon (LAr) from underground sources (underground argon, UAr), out of which the central 20 tonnes will be the fiducial volume (see [3] for an updated design). Dual-phase TPCs have led the direct search for WIMPs with mass above 10 GeV/c^2 in the last decade using xenon targets [4, 5, 6, 7, 8] thanks to large volumes, self-attenuation, passive and active shielding, position reconstruction, energy resolution, and particle discrimination.

The latter is mainly achieved via measuring the different rate between ionization and scintillation read out at a given electric field. This field induces the drift of the electrons to the anode on top at a fixed velocity. Once there, they are reaccelerated into the gas phase so that multiplication is induced and the associated electroluminescence flash can be observed. The drift time is proportional to the distance to the anode, i.e. the Z coordinate, so the time difference between the primary (S1) light pulse and this second (S2) light pulse gives mm precision in that coordinate. The S2 light pattern in the different light sensors of top readout plane provides XY resolution on the order of few cm.

LAr TPCs are especially effective in discriminating the electron recoils (ER) induced by the more abundant beta and gamma backgrounds from the expected nuclear recoils (NR) induced by the WIMPs interacting coherently with the Ar nucleus. This is thanks to the pulse-shape-discrimination capabilities of this target, with a singlet state preferentially populated by NRs and decaying in $\sim$ few nsec and a triplet state preferentially populated by ERs and decaying in $1.6\,\mu$s.

To further mitigate the backgrounds, around the TPC there will be an active veto based on the absorption of the neutron in a Gd-loaded acrylic wall and detection of the subsequent gammas in both the TPC and an additional external UAr layer. All the UAr is contained in a steel vessel and surrounded by a buffer of 650 tonnes of atmospheric Ar contained in a DUNE-like cryostat. This outer buffer will be instrumented for tagging muons and their byproducts.

Even with the discrimination power proven in the DEAP-3600 experiment (better than 10^{-9} in all the region of interest [9]) and the nested detector design, one needs to minimize the backgrounds coming from the materials in order to operate such a large detector and fulfill the scientific goals. The main endeavours in this direction are two. First, minimizing the ^{39}Ar content in the target is achieved with a specific program to extract [10], purify [11] and characterize [12] the UAr. Then, a worldwide effort is made on the selection of adequate materials to allow a trigger rate well below 200 Hz with fewer than 0.1 expected untagged NR backgrounds induced by materials in 10 years of exposure.

RADIOPURITY CONSTRAINTS FOR WIMP SEARCHES WITH LIQUID ARGON

The particular case of dual-phase TPCs using a LAr target, with enhanced particle-discrimination capabilities, shifts the importance from ER backgrounds (emanated Rn progeny is the main source of background in Xe TPCs as of now [7, 8]) to NR backgrounds, dominated by CEνNS, random coincidences, and neutrons from (α,n).

Notwithstanding that, the maximum drift time foreseen for DarkSide-20k is $\sim$5 ms, so a trigger rate of 200 Hz would induce a pile-up rate close to 1. Therefore, all the backgrounds, including those inducing ERs, need to be kept to the minimum so that the identification of S2 to its corresponding S1 can be done without ambiguity.

The γ radiation is the most critical ER background due to the relatively large range of the naturally occurring gammas above 500 keV. The main sources of such gammas are ^{40}K and the decay chains of ^{238}U, ^{232}Th, and ^{235}U. Other isotopes such as ^{60}Co and ^{137}Cs are also searched for, especially in materials where they are often found in non-negligible amounts (Co in iron and steel samples, for instance). High Purity Germanium (HPGe) counters in low-background environments [13, 14, 15, 16] are used in order to measure these activities. Then, the impact of these activities in the detector rates is quantified with Monte Carlo simulations using G4DS [17], where the full geometry of DarkSide-20k is implemented.

Regarding β decays, the range of the outgoing electron is short and only materials in the walls of the TPC or diluted in the target can induce detectable events. A source of triggers and a potential contribution to the background is the mentioned decay of ^{39}Ar in the target bulk, which might accidentally coincide with another event and together mimic a good WIMP-like event.

The main background leaking in the region of interest is a by-product of α decays. These tracks are even shorter than those of β, but the typical energies of the naturally occurring α-decays are enough to produce neutrons via (α,n) reactions in the nuclei of the surrounding material. Such neutrons, in the few-MeV range, can induce single recoils in the TPC that perfectly match the signal expected for a WIMP elastic recoil with a nucleus.

Having a proper estimate of such backgrounds is, hence, critical for the success of the experiment. Three ingredients are necessary for each component: measurement of its radiopurity, calculation of the neutron yield, and simulation of the neutron transport from the production location, with special attention paid to the energy (E) lost in the sensitive volumes (veto and TPC) that could allow the rejection of this background.

The neutron transport is also simulated with G4DS. These simulations show that the neutron veto will have an efficiency close to 90%, and the combined inefficiency in the analysis cuts – the proportion of neutrons leaking into the region of interest for WIMP searches per neutron produced – ranges from 10^{-4} in the vicinity of the TPC to 10^{-10} for the external cryostat.

The neutron yield via (α,n) is calculated using SaG4n [18, 19] and validated with neuCBOT [20]. SaG4n was developed within DarkSide and has the main advantage of being Geant4-based (easy to use) and being able to read ENDF-6 format libraries. As a consequence, the experimental information available in the JENDL libraries [21] can be used with this framework, instead of global theoretical approaches that do not take into account specific cross sections and resonances.

The α radiation by itself, as in the case of the β radiation, is of short range and is only a problem in the surface of the TPC walls and in the bulk of scintillating materials. Surface contamination is most commonly due to exposure to ^{222}Rn in air. The decay products of Rn are not gases and plate out on the surfaces in contact with the air. These decays are fast up to ^{210}Pb, which has a half-life of 22 y and decays into ^{210}Bi (5 days) and then ^{210}Po (138 days). Even though the energy and position reconstruction capabilities of TPCs have a large rejection factor, these events

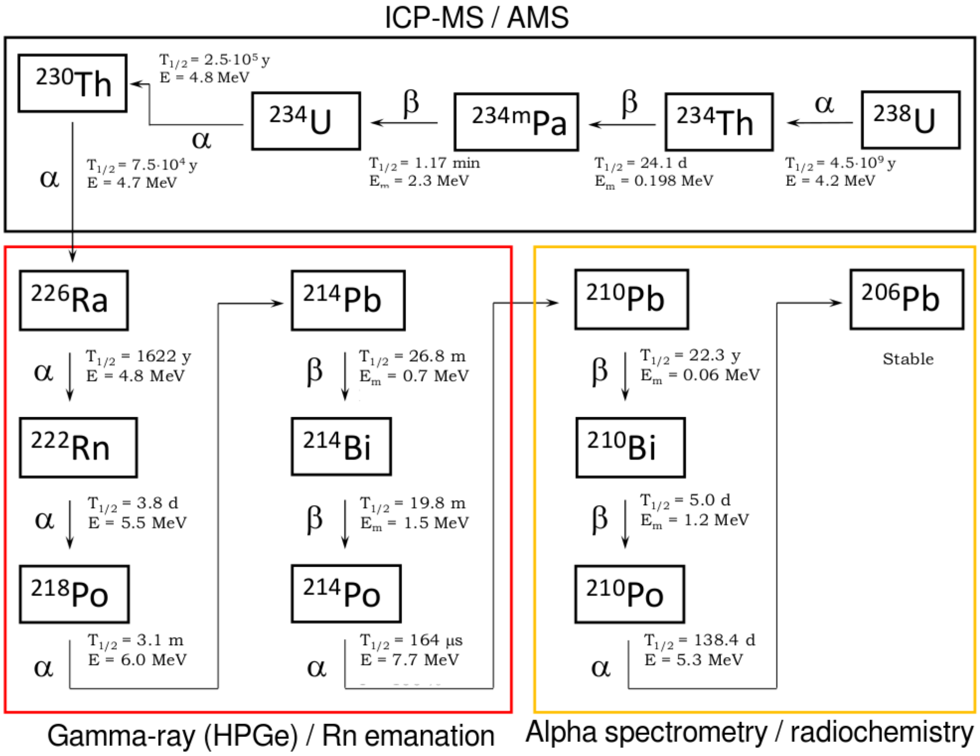

FIGURE 1. Full decay chain of ^{238}U showing the different sections after common breaking of secular equilibrium.

might leak into the region of interest if happening in coincidence with other events. A specific set of protocols to avoid exposure to Rn and to clean surfaces has been put in place.

THE ASSAY CAMPAIGN

The activity of a component depends on several factors, i.e. origin of raw materials and their history of exposure, production process and environment, storage, etc. As a consequence, an *a priori* selection of the materials suitable for building our experiments is not feasible, and a case-by-case study is necessary in order to understand the type of backgrounds induced by each component, quantify them, and guarantee that the chosen material allows for the scientific goals to be achieved.

Material assays using HPGe crystals in low-background facilities are common practice during the development of rare-event detectors. For the ^{238}U and ^{232}Th decay chains this technique is mainly sensitive to the central part of the series, where the E of the gamma rays is high and their intensity relatively large. The sensitivity to the top part of the ^{238}U chain is generally one order of magnitude lower, and the gamma line of ^{210}Pb requires specific detectors with thin windows that do not shield the 46-keV gamma line.

The state-of-the-art also includes ICP-MS, which has sensitivity to concentrations below 10^{-12} g/g and a shorter time needed to process the sample and get results. In practice, ICP-MS can measure only the long-lived isotopes at the top of the decay chains (^{238}U, ^{232}Th, and ^{235}U themselves) and is insensitive to the breaking of secular equilibrium. Within DarkSide, we are combining this kind of assay with X-ray fluorescence and chromatography to perform elemental analysis of samples in which the chemical composition is unknown or the presence of elements with large (α,n) cross sections is suspected. This information is fundamental in order to calculate the neutron yield in components with proprietary or undisclosed composition.

The combination of ICP-MS and HPGe counting allows a quantification of the breaking of secular equilibrium in ^{226}Ra (black to yellow boxes in Figure 1). Since Ra is soluble in water and is the last isotope in the ^{238}U chain with a half-life above 1000 y, secular equilibrium does not occur in most cases. This fact has been known and accounted for since the last generation of experiments, but the breaking of secular equilibrium in the bulk of the materials at the ^{210}Pb and ^{210}Po levels (yellow to green boxes in Figure 1) has not.

The assay campaign of DarkSide-20k is the first one that systematically performs radiochemical separation of ^{210}Po

[22] for all components. This effort has enabled us to characterize a general enhancement in the bulk activity of ^{210}Po with respect to ^{226}Ra. The ratio of ^{210}Po/^{226}Ra activity is very diverse, and DarkSide has measured values above 1000 – for instance in bismuth-rich solder pastes. This ratio is typically significantly larger than the ^{226}Ra/^{238}U ratio in the same chain, which rarely goes above 10. These results demonstrate the need of this comprehensive approach. The surface ^{210}Po activity is also quantified, using XIA spectrometers at Jagiellonian U. and SNOLAB, for samples with large area in the vicinity of the TPC.

Tools and Organization

The effort of the assay campaign is coordinated by the Materials Working group of the collaboration and distributed among different laboratories around the world. The main underground laboratories contributing are Laborari Nazionali del Gran Sasso (ICP-MS and HPGe), Laboratorio Subterráneo de Canfranc (LSC: ICP-MS and HPGe), SNOLAB (HPGe and surface counting), and Boulby (HPGe). Other institutions contributing to the assays are CIEMAT (ICP-MS, XRD and chromatography), Jagiellonian U. in Krakow (Po radiochemical separation and surface counting), Temple (HPGe), Aix-Marseille U. (Rn emanation and permeability), and Mendeleev U. (ICP-MS).

In order to organize this delocalized effort in a coherent way, a custom infrastructure has been put in place. The principal tools of this infrastructure are an online database and a radioactivity budget. The database centralizes the full assay process. When a new component is being considered, an assay request is uploaded, with the relevant information on available mass, format, urgency, manipulation instructions, supplier, responsible working group, and contact person. A decision on the kind of assay and its location is made according to this information and taking into account the queues in the corresponding labs. The database also stores the tracking information of the sample –shipping, onsite, on assay, waiting for report, or done. Eventually, a full report is uploaded together with a summary of the relevant measured values in a table format that allows for quick consultation and comparisons between samples.

The suitability of a component is evaluated in the radioactivity budget. This budget consists of a series of spreadsheets in which the radioactive contributions of all the components in the detector are evaluated and summed. This quantification is done using the results of the assays, the expected total mass (unit mass $\times$ # units) and the component's location in the experiment. The location is used in the simulations to evaluate the probability of gammas and neutrons to become actual events and optimize the setup accordingly. For the neutron estimate, the $(\alpha$,n) yield is calculated taking into account the precise chemical composition.

Current Status

As of now, more than 200 assays have been performed, out of which 52 different components are included in the current radioactivity budget. The effort specifically focused on DarkSide-20k has been going on since 2017, and all the information is stored in a database that includes the former experience and data from DarkSide-50.

The current estimate of NRs leaking into the region of interest after cuts is 0.1. This is a best estimate biased to the conservative side, since it combines central values, conservative estimates, and upper limits of the assay results. As a consequence, error bars cannot be given at this stage. The main contributions to the NR background are due to the photoelectronics and cryostat, with the substrate of the PCBs as the single element with largest contribution. Globally, the top 10 contributors represent 85% of the budget.

A clear evidence of the importance of measuring the bulk ^{210}Po activity is that even though it only contributes one α decay (compared to the eight in the full uranium series and six in the thorium series) and this alpha has relatively low energy, it induces up to 26% of the expected neutron production due to the strong breaking of secular equilibrium at the ^{210}Pb level.

THE UNDERGROUND ARGON PROGRAM

DarkSide-20k would not be possible without the use of UAr. The ^{39}Ar activity of 1 Bq/kg in natural argon combined with the 50 tonnes contained in the TPC and the 5 ms of maximum drift time would make the detector unfeasible to operate. An activity of 0.7 mBq/kg was measured by DarkSide-50 using UAr, and lower activities are expected if contamination after extraction is prevented. Measuring such a low β activity is a challenge, and none of the techniques mentioned in this text are suitable. Instead, an ad hoc experiment is being located at the Canfranc Underground Laboratory to assay the activity of each UAr batch after its extraction in Urania [10] and its purification in Aria [11]. This is the DArTinArDM (or DArT) experiment [12, 23]. It exploits the former ArDM experiment as an active veto, with 1.5 kg of UAr in a separated volume in its center. It has sensitivity to measure activities of 0.07 mBq/kg with an uncertainty of 40% in a week of data-taking.

CONCLUSIONS AND OUTLOOK

The DarkSide-20k collaboration aims at having fewer than 0.1 background events after cuts in the region of interest due to nuclear recoils in 10 years of exposure, and a total rate well below 200 Hz in the TPC to avoid large pile-up rates. In order to fulfill this challenging goal, a worldwide effort has been going on for more than 5 years focused on a thorough assay campaign, performing at least three measurements (ICP-MS, HPGe counting and Po separation) of most components that will go into the detector. To coordinate this effort, an infrastructure based on an online database and a series of online spreadsheets has been put in place.

This infrastructure has been a cornerstone for the development of DarkSide-20k, informing the design and the choice of components. As a result, currently the experiment fulfills the specifications of 0.1 NR backgrounds in the region of interest after analysis cuts and a total rate in the TPC below 100 Hz.

As a byproduct, this effort has produced several relevant contributions for the low-background community: new procedures for neutron yield estimates, standardization of assays (beyond HPGe and ICP-MS), novel ways for bookkeeping and storing results, and a vast new series of measurements that will be published soon.

Besides the tasks developed within the Materials Working Group, there is a fundamental role played by underground facilities (LSC, LNGS, Boulby and SNOLAB) and other facilities (Temple, CIEMAT, Mendeleev U., Jagiellonian U., Aix-Marseille U.) to fully characterize the radioactivity with no secular-equilibrium assumption.

ACKNOWLEDGMENTS

The DarkSide Collaboration would like to thank LNGS, LSC, SNOLAB, Boulby, and their staff for invaluable technical and logistical support. We acknowledge the financial support by the Spanish Ministry of Science and Innovation PID2019-109374GB-I00 (MICINN), “Unidad de Excelencia María de Maeztu: CIEMAT - Física de partículas” (Grant MDM2015- 0509), the U. S. National Science Foundation (NSF) (Grants No. PHY- 0919363, No. PHY-1004054, No. PHY-1004072, No. PHY-1242585, No. PHY-1314483, No. PHY-1314507, associated collaborative grants, No. PHY-1211308, No. PHY-1314501, No. PHY-1455351 and No. PHY-1606912, as well as Major Research Instrumentation Grant No. MRI-1429544), the Italian Istituto Nazionale di Fisica Nucleare (Grants from Italian Ministero dell’Istruzione, Università, e Ricerca Progetto Premiale 2013 and Commissione Scientific Nazionale II), the Natural Sciences and Engineering Research Council of Canada, SNOLAB, and the Arthur B. McDonald Canadian Astroparticle Physics Research Institute, LabEx UnivEarthS (ANR-10-LABX-0023 and ANR-18-IDEX-0001), the São Paulo Research Foundation (Grant FAPESP-2017/26238-4), and the Russian Science Foundation Grant No. 16-12-10369, the Polish National Science Centre (Grant No. UMO-2019/33/B/ST2/02884), the Foundation for Polish Science (Grant No. TEAM/2016-2/17), the International Research Agenda Programme AstroCeNT (Grant No. MAB/2018/7) funded by the Foundation for Polish Science from the European Regional Development Fund, the Science and Technology Facilities Council, part of the United Kingdom Research and Innovation, and The Royal Society (United Kingdom). I.F.M.A is supported in part by Conselho Nacional de Desen- volvimento Científico e Tecnológico (CNPq). We also wish to acknowledge the support from Pacific Northwest National Laboratory, which is operated by Battelle for the U.S. Department of Energy under Contract No. DE-AC05-76RL01830.

REFERENCES

1. C. Aalseth el al., “DarkSide-20k: A 20 tonne two-phase LAr TPC for direct dark matter detection at LNGS,” Eur. Phys. J. Plus **133**, 131 (2018).
2. M. Ambrosio et al., “Vertical muon intensity measured with MACRO at the Gran Sasso laboratory,” Phys. Rev. D **52**, 3793 (1995).
3. T. Thorpe *et al*, “The DarkSide-20k TPC and underground argon cryogenic system,” arXiv:2210.00322 (2022).

4. D. S. Akerib *et al.* (LUX Collaboration), “Results from a search for dark matter in the complete LUX exposure,” Phys. Rev. Lett. **118**, 021303 (2017).
5. E. Aprile *et al* (XENON Collaboration), “Dark matter search results from a one ton-year exposure of XENON1T,” Phys. Rev. Lett. **121**, 111302 (2018).
6. X. Cui *et al* (PandaX-II Collaboration), “Dark matter results from 54-ton-day exposure of PandaX-II experiment,” Phys. Rev. Lett. **119**, 181302 (2017).
7. D. S. Akerib *et al.* (The LUX-ZEPLIN Collaboration), “First dark matter search results from the LUX-ZEPLIN (LZ) experiment,” arXiv:2207.03764 (2022).
8. E. Aprile *et al*, “Projected WIMP sensitivity of the XENONnT dark matter experiment,” JCAP **2020**, 031–031 (2020).
9. P. Adhikari *et al.* (DEAP-3600), “Pulse-shape discrimination against low-energy Ar-39 beta decays in liquid argon with 4.5 tonne-years of DEAP-3600 data,” Eur. Phys. J. C **81**, 823 (2021).
10. W. M. Bonivento *et al.* (The DarkSide-20k Collaboration), “Low radioactivity argon for dark matter and rare event searches,” in *16th International Conference on Topics in Astroparticle and Underground Physics (TAUP 2019) 9-13 September 2019, Toyama, Japan*, J. Phys.: Conf. Ser., Vol. 1468 (2020) p. 012234.
11. P. Agnes *et al.* (The DarkSide-20k Collaboration), “Separating 39Ar from by 40Ar cryogenic distillation with Aria for dark-matter searches,” European Physical Journal C **81**, 359 (2021).
12. C. E. Aalseth *et al* (The DarkSide-20k Collaboration), “Design and construction of a new detector to measure ultra-low radioactive-isotope contamination of argon,” JINST **15**, P02024 (2020).
13. M. Laubenstein, “Screening of materials with high purity germanium detectors at the Laboratori Nazionali del Gran Sasso,” International Journal of Modern Physics A **32**, 1743002 (2017).
14. I. Bandac, S. Borjabad, A. Ianni, R. Nuñez-Lagos, C. Perez, S. Rodríguez, and J. Villar, “Ultra-low background and environmental measurements at Laboratorio Subterráneo de Canfranc (LSC),” Applied Radiation and Isotopes **126** (2017), 10.1016/j.apradiso.2017.02.046.
15. I. Lawson and B. Cleveland, “Low background counting techniques at SNOLAB,” in *Workshop on Low Radioactivity Techniques: LRT2013*, American Institute of Physics Conference Series, Vol. 1549, edited by L. Miramonti and L. Pandola (2013) p. 16.
16. P. R. Scovell *et al*, “Low-background gamma spectroscopy at the Boulby Underground Laboratory,” Astroparticle Physics **97**, 160–173 (2018).
17. P. Agnes *et al* (The DarkSide-50 Collaboration), “Simulation of argon response and light detection in the DarkSide-50 dual phase TPC,” JINST **12**, P10015 (2017).
18. E. Mendoza, D. Cano-Ott, P. Romojaro, V. Alcayne, P. García Abia, V. Pesudo, L. Romero, and R. Santorelli, “Neutron production induced by α-decay with Geant4,” NIM A **960**, 163659 (2020).
19. V. Pesudo *et al.*, “SaG4n: Calculation of (α,n) yields for low background experiments using Geant4,” in *16th International Conference on Topics in Astroparticle and Underground Physics (TAUP 2019) 9-13 September 2019, Toyama, Japan*, J. Phys.: Conf. Ser., Vol. 1468 (2020) p. 012059.
20. S. Westerdale and P. Meyers, “Radiogenic neutron yield calculations for low-background experiments,” NIM A **875**, 57–64 (2017).
21. K. Shibata *et al*, “JENDL-4.0: A New Library for Nuclear Science and Engineering,” J. Nucl. Sci. Technol. , 1–30 (2011).
22. T. Mroz *et al*, “Studies of bulk 210Po/210Pb contamination in high purity copper for low background detectors,” `https://indico.ific.uv.es/event/6178/contributions/15941/` (2021), tAUP2021 oral presentation.
23. V. Pesudo *et al.* (The DarkSide-20k Collaboration), “Measurement of the underground argon radiopurity for dark matter direct searches,” in *17th International Conference on Topics in Astroparticle and Underground Physics (TAUP 2021), 26 August-3 September 2021, Valencia*, J. Phys.: Conf. Ser., Vol. 2156 (2021) p. 012043.