

High Priority Request List cross-section measurements: ${}^7\text{Li}(\text{d},\text{x}){}^7\text{Be}/{}^3\text{H}$ and ${}^{39}\text{K}(\text{n},\text{p}){}^{39}\text{Ar}$

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Abstract. The Nuclear Energy Agency's High Priority Nuclear Data Request List is a compilation of the highest priority nuclear data requirements. The U-120M cyclotron of the Nuclear Physics Institute of the Czech Academy of Sciences is a suitable tool for studies of several reactions from this list. In this paper, we present the measurements of the ${}^7\text{Be}$ and ${}^3\text{H}$ production in the lithium-7 after the irradiation with deuterons and the validation measurement for the ${}^{39}\text{Ar}$ production in ${}^{39}\text{K}$ after irradiation with neutrons.

1 Introduction

The reactions from Nuclear Energy Agency's High Priority Nuclear Data Request List (HPRL) namely ${}^7\text{Li}(\text{d},\text{x}){}^7\text{Be}$ and ${}^7\text{Li}(\text{d},\text{x}){}^3\text{H}$ are important processes in the target system of the International Fusion Materials Irradiation Facility - Demo Oriented NEutron Source (IFMIF-DONES) [1]. They will produce 100% of the radioactive isotope ${}^7\text{Be}$ and 80% of tritium in the IFMIF-DONES lithium loop respectively. The accuracy of both cross sections will therefore impact the efficiency, design, and cost of the IFMIF-DONES radio-protection measures which should guarantee the safe accumulation of ${}^7\text{Be}$ and ${}^3\text{H}$ in the lithium loop heat exchanger and cold inventory traps [2–4].

Regarding tritium production in lithium by deuterons, only a few measurements using the activation tritium technique exist [5, 6]. The cross-section data is available up to 8 MeV, and has disagreements and large uncertainties. The ${}^7\text{Be}$ production cross section was measured from threshold to 13 MeV by activation technique in several experiments [7, 8]. At higher energies, there is only one measurement for natural lithium.

Cyclotron U-120M based at the Nuclear Physics Institute of the Czech Academy of Sciences (NPI CAS) can accelerate deuterons up to the energy of 20 MeV. Irradiations of lithium pellets with deuterons and subsequent measurements of ${}^7\text{Be}$ were performed on-site by HPGe detectors. Tritium production was measured at Karlsruhe Institute of Technology (KIT) following a method developed by Verzilov et al at JAERI [9, 10].

Preliminary results were obtained and track for future research was set.

The second reaction from the HPRL, ${}^{39}\text{K}(\text{n},\text{p}){}^{39}\text{Ar}$, produces ${}^{39}\text{Ar}$ with a decay half-life of 269 years and makes the dominant contribution to the long-lived radioactive inventories in the sodium–potassium alloy (NaK). This alloy is considered as coolant of specimens in the accelerator driven irradiation facilities that are designed for fusion material testing. Together with the competing reaction ${}^{39}\text{K}(\text{n},\text{np}){}^{38}\text{Ar}$ it also determines the total amount of argon gas which impacts on the thermal and mechanical properties of the sealed specimen containers [11]. The current poor knowledge of this reaction questions whether NaK could be used in the IFMIF-DONES designs. Additionally, since potassium is present in cement and concrete, the ${}^{39}\text{K}(\text{n},\text{p}){}^{39}\text{Ar}$ reaction impacts on the long-term radioprotection and shielding issues in the IFMIF-DONES testing vaults and future fusion power plants.

Potassium is a highly active material from a chemical point of view. The decay of the produced nucleus ${}^{39}\text{Ar}$ emits no gammas and it can be only observed by beta spectrometry. These facts limit the materials and experimental techniques that can be used. Our first attempt was to use the pressed K_2CO_3 powder as the target material. It appeared that the powder was hygroscopic, and the target became fragile after irradiation and subsequent manipulation. Mica foils (source Goodfellow) turned out to be the right alternative. The foils were irradiated using different neutron spectra (reactor, quasi monoenergetic, 14 MeV). The measurements of the beta spectrum electrons emitted during the decay of ${}^{39}\text{Ar}$ were used to determine the amount of produced isotope and its production cross section.

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2 $^7\text{Li}(\text{d},\text{x})^7\text{Be}$ and ^3H production measurements

2.1 Experimental setup

The Li_2CO_3 powder pressed into the pellets was chosen as the target material. The diameter of the press form was 13 mm. The deuteron beam size was smaller than the diameter of the pellets, hence only the thickness and the mass of each pellet were the essential parameters. They were 1 ± 0.01 mm and 0.207 ± 0.001 gram. The irradiation was performed with 20 ± 0.5 MeV deuterons. The energy degraders of metallic foils (titanium, copper, aluminum) were used to irradiate at lower deuteron energies. The chamber setups for three irradiations are shown in the Fig. 1. The metallic foils were also used as monitors. The energy loss was estimated on the basis of the SRIM code calculation [12] and checked with data from monitors. After the irradiation, it turned out that one of the Li_2CO_3 pellets was damaged during the irradiation (first scheme in Fig. 1). The deuteron charge deposited in this pellet could not be determined and the results were excluded from further analysis. The cross section data were measured only for three average deuteron's energies namely 16.0, 12.5, and 9.8 MeV. After irradiation activity of different isotopes accumulated in pellets and monitors was measured at the NPI CAS, using a high-purity Ge detector [13]. The activated samples were placed at a distance of 20 cm or 30 cm from the end-cap of the detector in order to have the dead time less than 3%.

Beta spectrometry was used to determine the amount of produced tritium. The pellets were sent to KIT, where the necessary equipment for this procedure is available. The Li_2CO_3 pellets were dissolved in a mixture of two acids, nitric (grade 61%) and acetic acid (grade 100%), and then incorporated into liquid scintillator (Clearsol-II, Nacalai Japan). All pellet processing was done in standard 20 ml glass vials for beta counting. Beta counting was done with a commercial liquid scintillation beta spectrometer Triathler (Hidex). The beta activity in each sample was determined from the count rates of the sample, a background sample without tritium, and a traceable tritium standard sample with known tritium activity including uncertainty.

2.2 Results and discussion

The measured numbers of activated nuclei in the monitor foils were used to determine the energy and the flux of the deuterons. The IAEA recommended cross sections were used [14].

The preliminary results for the cross section of the ^7Be production are shown in Table 1, the comparison with already existing data illustrated in Fig. 2 together with the other experimental data [8, 15–17], databases TENDL-2009 [18] and ENDF/B-VIII.0 [19]. The present data point at the lower-energy region below 10 MeV is consistent with the data reported by Guzhovskij et al. [17]. However, the present data are considerably lower in magnitude than the data from TENDL-2009 database and data reported by

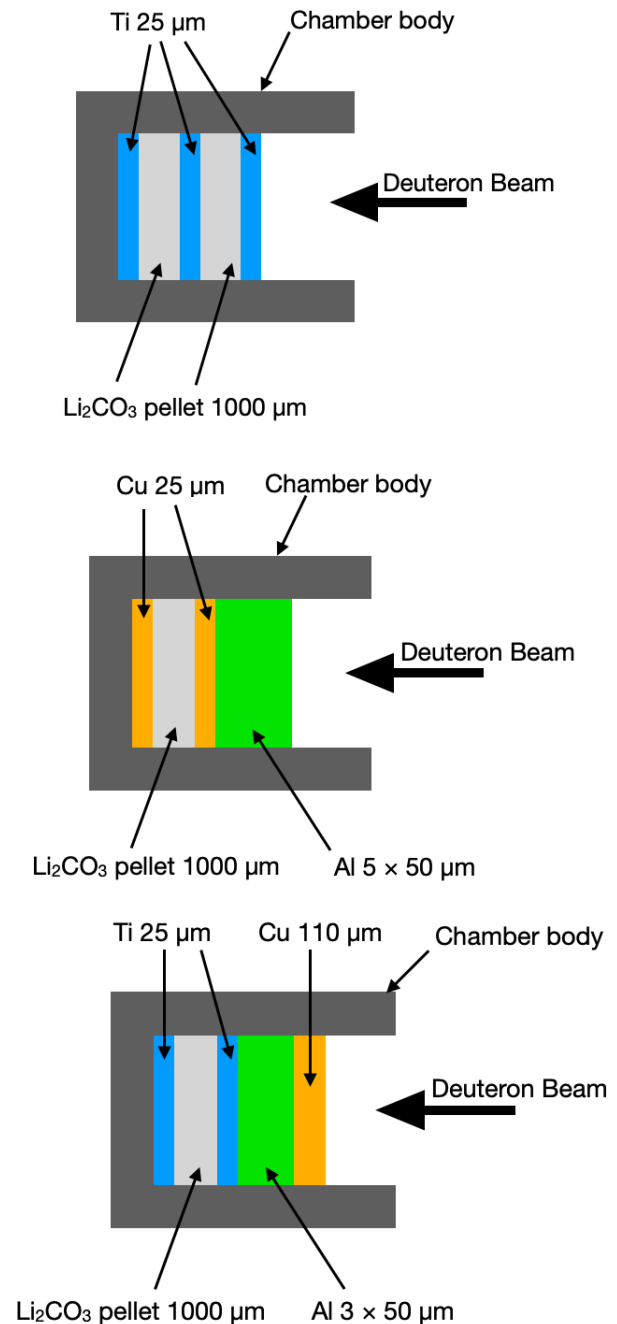


Figure 1. Targets schemes for different average deuteron energy: 16.0 (top), 12.5 (middle), 9.8 (bottom) MeV

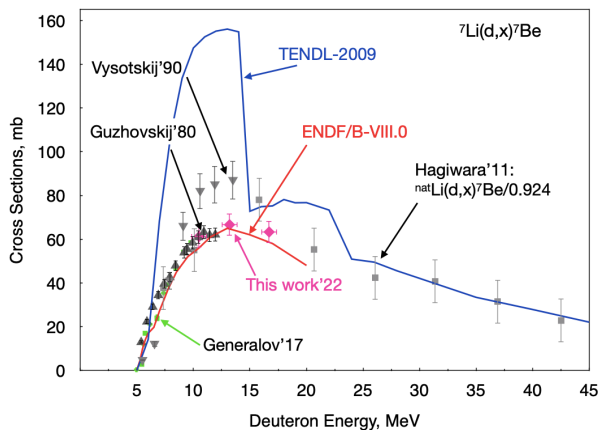
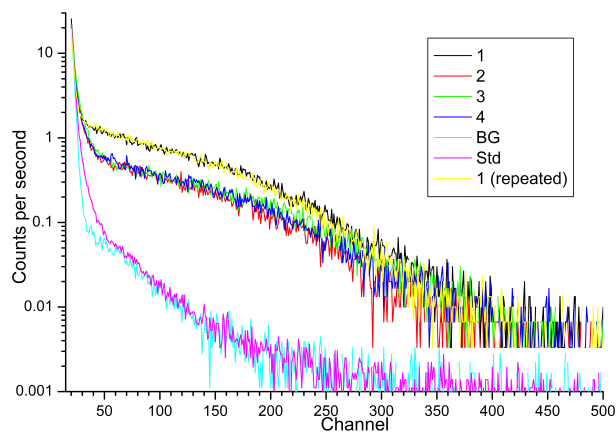
Vysotskij et al. [15]. On the other hand, there is a good agreement with ENDF/B-VIII.0 database and the present data points are following the trend in terms of shape.

Regarding the tritium production by the deuteron irradiation of the pellets, one can find an illustration of the recorded beta spectra in Fig. 3. Contribution from the tritium lies in the range of 30-120 channels. The response was also measured for the standard tritium source (std in Fig. 3) and both responses were compared.

The tritium activity in the samples could not be determined with good accuracy. This was to a large extent due to an additional beta activity with higher beta

Table 1. Preliminary results for ^7Be production cross section

Average energy, MeV	CS, mb
16.0 ± 0.7	63.3 ± 5
12.5 ± 1.0	66.6 ± 5
9.8 ± 1.2	61.2 ± 5

**Figure 2.** Cross section of $^7\text{Li}(d,x)^7\text{Be}$ reaction compared with other experimental data and libraries**Figure 3.** Measured beta spectra from samples irradiated with different average deuteron energy: 16.0 (1), broken sample (2), 12.5 (3), 9.8 (4) MeV, standard source (Std) and the background measurement (BG).

energies and a comparably high activity that dominated the collected counts in lower spectrum channels where also the betas from tritium decay are registered. That difference we considered to be a contribution of scattered gammas from ^7Be decay. Due to these facts, it was not possible to extract cross section but we formulate a list of demands for the future experiment.

3 $^{39}\text{K}(n,p)^{39}\text{Ar}$ measurements

3.1 Experimental setup

1x1 cm mica i.e. potassium aluminum silicate foils of 200 μm thickness (K-60-SH-000160 from Goodfellow) were cut and weighed. These foils were irradiated at different facilities in order to understand the impact of different neutron spectra.

Prior and after the irradiation, the mica foils were measured using the Si(Li) detector [20]. A special holder fixing the position of the mica foil and the Pb shielding was used. The measurements after the irradiation were performed in regular time intervals, the electron spectrum was recorded every 48 hours to follow the decay of parasitic activities.

Before irradiation, there is a known amount of ^{40}K in mica foil which can be determined by weighing the foil and using the natural abundance precisely measured many times by the AMS method [21, 22]. The decay of ^{40}K and ^{39}Ar produce a beta electron spectrum. The beta spectrum is continuous and was calculated using the BetaShape code [23, 24] code. We assume that the electrons in the energy range 200-400 keV produced by ^{40}K and those produced by ^{39}Ar (in irradiated mica) have the same probability to be registered (if the geometry of the measurement is the same) no matter from which radionuclide they originate.

For beta spectrum measurements we used Si(Li) detector with lead shielding. With such a setup three measurements are essential: background without the sample, the mica foil before irradiation, and the time series of measurements of the irradiated mica foil. All the measurements were performed in exactly the same geometry. The continuous electron spectrum in the energy range 200-400 keV was compared. The excess of the betas in this energy range tells us the number of ^{39}Ar related to the number of ^{40}K .

The background has to be measured since the excess of the betas from the ^{40}K in the mica is not very significant even in the closest position (the closest distance to Si(Li) detector to which the foil can be placed is 5 mm). The lead shielding improves the situation. The Signal To Background ratio was 10% without the shielding and 25% with the shielding.

The code FISPACT-II [25] was used to predict the produced electron emitters at different cooling times after the irradiation with neutrons. Their beta spectra were calculated with BetaShape. It was found that the range 200-400 MeV is free from the influence of other known produced radionuclides.

3.2 Results and discussion

One mica foil was irradiated in H1 channel of NPI's reactor (LVR-15) [26]. Fluence of the fast neutrons was $1.32 \times 10^{15} \text{ n/cm}^2$. The response measured with the Si(Li) is shown in the Fig. 4.

The recorded response spectrum was entirely dominated by the radioactivity from impurities activated by

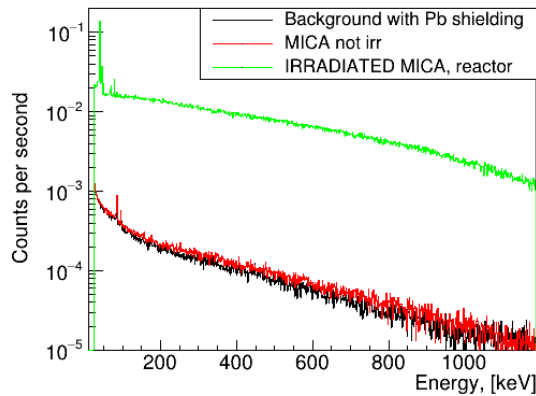


Figure 4. Measured response from the sample irradiated in the reactor channel. The cooling time is 20 days.

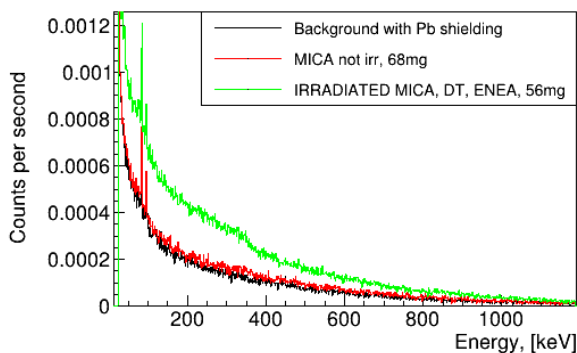


Figure 5. Measured response from the sample irradiated at FNG. The cooling time is 10 days.

thermal neutrons. Some responsible isotopes were identified by a separate gamma spectrometry measurement, to name a few: ^{60}Co , ^{46}Sc , ^{59}Fe , ^{134}Cs , and etc.

Another irradiation took place at the Frascati Neutron Generator (FNG) [27] which provides 14 MeV neutrons through $\text{T(d,n)}\alpha$ reaction. The FNG target was at least four meters away from the walls and roof and the sample was located very close to the FNG target (from 1 to 5 mm, depending on the irradiation). The contribution of scattered neutrons was estimated to be at least five orders of magnitude weaker than the 14 MeV neutron fluence.

The impurities in the mica foil could therefore not be activated by intermediate and thermal neutrons at such scale than during the LVR-15 irradiation. Some contribution from other unknown sources was however measured using the Si(Li) detector, see Fig. 5.

The ^{39}Ar nuclide emits the electrons through β^- decay (the endpoint is 565 keV). The surplus of the pulses above the 565 keV endpoint can be attributed to the decay of unknown activated nuclides. After cooling times long enough that the radioactivity of other nuclides decreases,

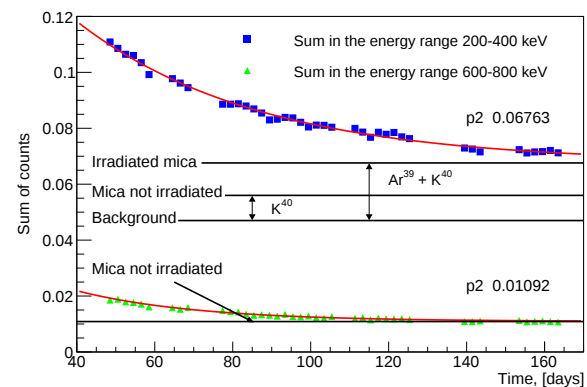


Figure 6. The analysis of the contribution in different energy ranges.

only the surplus of the electrons from ^{39}Ar decay is measured.

The pulses in the two energy ranges were measured in regular time intervals (48 hours). The results of such monitoring are shown in Fig. 6. The electrons from the decay of ^{39}Ar and ^{40}K contributed the pulses to the energy range of 200-400 keV. The surplus in the energy range of 600-800 keV was monitored to determine the cooling time when the radioactivity from the unknown sources decreases below the measurable limit. The curve could however be fitted with the exponential function and obtain the level to which it converges which coincides with one from not irradiated mica foil. It can be seen that the necessary cooling time is >160 days. In the same manner, could be obtained the level to which the sum in the energy range 200-400 keV converges. After background subtraction from the results of the measurements of irradiated and not irradiated mica foil one can calculate the ratio of the pulses produced by ^{40}K alone and together by $^{39}\text{Ar}+^{40}\text{K}$.

The resulting ratio was used to calculate the ratio of the number of nuclei of these two isotopes. Because the number of nuclei of ^{40}K is known (determined from the mass of the foil and its chemical composition), the number of produced nuclei of ^{39}Ar can be calculated. The cross section was calculated by dividing this number with the integral neutron flux (Nb monitor foils were used), and the obtained number was: 110.1 mbarn. This number is in good agreement with the experimental data from earlier authors that used AMS [28] and is ca 3x smaller than the data from [29]. It confirms the validity of JEFF3.3 [30] evaluation.

4 Conclusion

The efforts to measure the cross section for two reactions of high importance for the experimental setup IFMIF-DONES that will be used for fusion experiments are described in this paper.

The experimental cross sections for the reaction $^7\text{Li(d,x)}^7\text{Be}$ were obtained, while the tritium production

by deuterons did not yield the experimental result, but has set the track for future studies.

Regarding the ^{39}Ar production by 14 MeV neutrons in potassium, the methodics for such measurements was worked out and promising preliminary results were obtained.

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