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PREPARATION AND CHARACTERIZATION OF A $^{39}$Ar SAMPLE AND STUDY OF THE $^{39}$Ar(n$_{th}$,α)$^{36}$S REACTION

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Abstract

The $^{39}$Ar(n$_{th}$,α)$^{36}$S reaction has been studied for the first time. A sample containing $2.85 \times 10^{14}$ $^{39}$Ar atoms, was produced at the ISOLDE facility at CERN. The number of $^{39}$Ar atoms in the layer was determined by measuring the $^{39}$Ar β-activity using a primary standardization method. Subsequently, the sample was irradiated with thermal neutrons at the High Flux Reactor of the ILL. An upper limit of 0.29 b was obtained for the $^{39}$Ar(n$_{th}$,α)$^{36}$S reaction cross-section.

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1 INTRODUCTION

Investigation of thermal neutron-induced charged-particle reactions can deliver information on properties of highly excited compound nuclear states, such as spin and parity. Moreover, cross-section and precise Q-values can be obtained. In the case of $^{39}$Ar, the $(n,\alpha)$ reaction is energetically favoured.

From an astrophysical point of view, this reaction occurs in the nucleosynthesis of some rare (neutron rich) isotopes in the S to Ca region [1]. The branching of the weak s-process nucleosynthesis path at $^{39}$Ar plays a key role in the production of $^{36}$S, $^{40}$Ar and $^{40}$K. Once the origin of these species can be established, their calculated abundances will serve as constraints for stellar evolution scenarios ($^{36}$S, $^{40}$Ar) as well as for nuclear cosmochronology ($^{40}$K). A thorough investigation of the $^{39}$Ar branching requires the availability of accurate cross section data for all the reactions involved.

In the literature, no measurements on the $^{39}$Ar$(n,\alpha)^{36}$S reaction have been reported, the main reason certainly being the unavailability of suitable samples.

In this paper we report on the sample preparation and characterization as well as on the results obtained from neutron irradiation.

2 PREPARATION AND CHARACTERIZATION OF AN $^{39}$Ar SAMPLE

The preparation and characterisation of suitable samples is a prerequisite for reliable cross-section measurements. The production of an $^{39}$Ar sample is not straightforward; it is a radioactive gas with a half-life of 269 y [2] and commercially not available. Moreover, the sample had to be on a conductive substratum as it was also intended to be used in combination with an ionization chamber where it is incorporated in one of the electrodes. Therefore, an implantation in an Al foil seemed to be the most appropriate solution. This goal was achieved at the isotope separator ISOLDE at CERN in Geneva, Switzerland. Subsequently, the number of $^{39}$Ar atoms implanted in the Al foil was determined by measuring the radioactive decay of $^{39}$Ar which was performed at the Institute for Reference Materials and Measurements (IRMM) in Geel, Belgium.

2.1 Sample preparation

The CERN PS Booster provides protons of 1.0 and 1.4 GeV which are used to induce spallation, fragmentation and fission reactions in a variety of targets. For production of radio-isotopes close to stability (like $^{39}$Ar), the highest cross-sections are achieved via spallation of a close-by target nucleus. Calcium is the first element above argon forming stable compounds at higher temperatures which can serve as ISOL (isotope separation on-line) target. Calcium oxide targets are routinely used at ISOLDE for the production of neutron-deficient argon isotopes [3]. Thus, in a first test run, $8 \times 10^{12}$ $^{39}$Ar atoms were collected during a 24 h test implantation with a 5 g/cm$^2$ CaO target and an average proton current of 1.5 $\mu$A. This corresponds to a yield of roughly $6 \times 10^7$ atoms per $\mu$C of primary proton beam.

However, for a meaningful measurement of the $^{39}$Ar$(n,\alpha)$ cross-section a sample containing more atoms was required. To collect the latter within reasonable beamtime a more efficient target/ion source combination was needed.

2.1.1 Target

In the CaO target used, mainly $(p,3p(x+1)n)$ spallation reactions from $^{42+x}$Ca contribute to the production of $^{39}$Ar. The dominant $^{40}$Ca (97% natural abundance) does
practically not\(^1\) contribute to the production of \(^{39}\)Ar and thus the effective target thickness for \(^{39}\)Ar production was only about 0.15 g/cm\(^2\).

Therefore a target had to be found where the main component is an isotope with sufficient neutrons (\(N_{\text{target}} \geq 21\)). The use of gram amounts of CaO strongly enriched in the isotopes \(^{42-48}\)Ca is prohibitively expensive. The next target elements are scandium and titanium. The production cross section at 1.05 GeV of \(^{39}\)Ar from titanium is larger (16.9±1.6 mb \([4]\)) than from scandium (13.7±1.1 mb \([4]\)). Moreover, owing to the excessive cost of the rare element scandium only titanium was taken into consideration.

Titanium foil targets were already used at ISOLDE for the production of neutron-deficient calcium radio-isotopes \([5]\). With a high packing fraction (nearly 30% of bulk density) they provide a good target thickness of about 25 g/cm\(^2\). However, because of the low melting temperature of titanium (1668 °C) and a local overheating during impact of the intense proton beam pulses (up to 3.4 \(\times 10^{13}\) protons in a 2.4 \(\mu\)s beam pulse), the titanium foil targets showed rapid sintering, causing strongly hampered release characteristics.

As an alternative to pure elements, carbides or oxides are often used as compounds with relatively high temperature stability \([6]\). Different oxides showed good release characteristics for noble gases \([3]\). Thus a titanium oxide target was considered. However, certain oxides (e.g. thoria) also tend to sinter rapidly when used in powder form. Here it is of advantage to use instead thin fibres, which have fewer mutual contact points and provide a better mechanical stability even when sintering starts locally at the contact points. Felts and wool from zirconium (and other oxide) fibres are commercially available and have already been successfully used at ISOLDE \([5]\). However, titanium fibres were not available and had to be produced in-house via the fossilization process: a rayon felt is impregnated with a titanium chloride solution and subsequently dried and oxidized at 600 °C under standard air atmosphere. This titanium felt was filled into a rhenium boat and outgassed at 1000 °C under vacuum. The rhenium boat with 34 g of titanium fibres was then introduced into a standard ISOLDE target container (tantalum tube with 20 mm inner diameter and 200 mm length), giving an effective target thickness of about 8.3 ± 1.0 g/cm\(^2\) titanium\(^2\).

\(2.1.2\) Ion source

To obtain ‘pure’ argon beams, a water-cooled transfer line is used to ‘freeze out’ less volatile isobars before they can reach the ion source. The latter was an ISOLDE type FEBIAD ion source (MK7). By adding stable argon with a calibrated leak to the ion source, an ionization efficiency of 3.5% was measured. This is consistent with the total efficiency of about 4% for \(^{39}\)Ar release and ionisation which was calculated from the known production cross-section and the measured \(^{39}\)Ar intensity. For the collection of the long-lived \(^{39}\)Ar, the target was run conservatively at temperatures below 900 °C.

For the isotope separation, the low mass side (GLM) of the General Purpose Separator (GPS) \([7]\) was used. The GPS consists of a double-focusing H-magnet with a bending angle of 70° and has a mass resolving power of \(M/\Delta M = 2400\). After magnetic separation the \(^{39}\)Ar ions (\(1^+\)) were implanted at 60 keV in a 12 \(\mu\)m Al foil. The yield was about

\(\)\(^1\) Minor contributions could occur from (n,2p) reactions by secondary neutrons created in the target and by (p,2p\(\pi^+\)) reactions.

\(\)\(^2\) The uncertainty on the thickness arises from the stochiometric ratio between Ti and O which was not explicitly determined.
$5 \times 10^8 \, ^{39}\text{Ar}$ ions per $\mu$C. These ions were collected during 3 days with up to $4.1 \, \mu$A average current of 1.0 GeV protons.

### 2.2 Sample characterization

Since $^{39}\text{Ar}$ is a $\beta$-decaying radionuclide ($Q_{\beta^-} = 0.6 \, \text{MeV}$), the number of $^{39}\text{Ar}$ atoms implanted in the aluminium foil could be determined by means of activity measurements. These measurements were performed with a $4\pi$ gas-flow pressurized proportional counter (PPC) [8], used at the IRMM for primary standardization work. The detector consists of a cylindrical gas chamber with an inner diameter of 80 mm and a central planar cathode dividing it into two D-shaped counters with an anode wire each. The stainless steel anode wires are 150 mm long and have a diameter of 21 $\mu$m. The $^{39}\text{Ar}$ ion-implanted aluminium foil was mounted in the central hole of the cathode and the emitted $\beta$-particles were detected and counted with close to 100% efficiency. Nevertheless, several measures had to be taken to obtain the right activity value, i.e. correcting for count loss due to absorption in the foil, electronic noise and system dead time.

The signal processing was performed by two parallel branches, a multi channel analyser (MCA) and a single channel analyser (SCA). The SCA branch consists of an adjustable threshold discriminator succeeded by a non-extending re-triggerable dead-time generator of zero recovery time and a live time clock gate, which processes the system clock pulses and establishes the live time of the measurement. By using an accurate and SI-traceable frequency generator and imposing a non-extending dead-time of known duration, the SCA branch is well suited for unbiased live time counting. The MCA branch was used for visual inspection of the energy spectrum and for the assessment of the lost fraction of counts below the SCA threshold discriminator. This is done by extrapolating down to zero energy, as shown in Fig. 1. The lower-level discriminator was set at 1 keV. By using a high electronic gain, and limiting the maximum output voltage, the extrapolated part of the spectrum was typically 1%.

![Theoretical and measured $^{39}\text{Ar} \, \beta$-spectrum](image)

**Figure 1:** Theoretical and measured $^{39}\text{Ar} \, \beta$-spectrum. The measured spectrum is shifted towards lower energies as the dimensions of the chamber are smaller than the range of the $\beta$-particles. The insert illustrates the extrapolation to zero energy (cut-off 1 keV) of the $^{39}\text{Ar} \, \beta$-spectrum measured at high gain.
The PPC was operated with a gas mixture of 90% argon and 10% methane at 0.1 MPa. As the output signal depends on the gas density inside the PPC, it is important to work under stable and reproducible conditions, independent of atmospheric conditions in the laboratory. For this purpose a specially designed active gas-flow control system was used to maintain the gas density within 0.01% of the reference value \[9\]. These conditions could be reproduced throughout the measurement campaign, also after opening the PPC for source manipulation.

The main source of uncertainty on the measured activity resides in the unknown quantity of absorbed electrons in the aluminium foil. This problem was tackled by performing additional measurements, gradually increasing the effective source thickness by adding identical Al foils on both sides of the original one. As shown in Fig. 2, the count rate turned out to be linearly dependent on the total foil thickness and the absolute activity was obtained by extrapolation to zero thickness. The linearity as well as the amount of the count loss was confirmed by EGS4 \[10\] Monte Carlo simulations (see Fig. 2).

![Figure 2: Measured and simulated \(^{39}\text{Ar}\) \(\beta\)-spectrum as a function of the foil thickness. The derived activity corresponds to the extrapolated value at zero foil thickness.](image)

Finally, an \(^{39}\text{Ar}\) activity of 23.3 ± 0.3 kBq was deduced, corresponding to \(2.85 \times 10^{14}\) atoms. The overall uncertainty on the number of atoms is 2%, taking into account the contributions from absorption, low-energy tail extrapolation and the half-life \[2\].

The implantation depth was calculated using the FORTRAN code TRIM (TRansport of Ions in Matter \[11\]). An average range of 0.06 \(\mu\)m was obtained for 60 keV \(^{39}\text{Ar}^{1+}\) ions in Al.

3 MEASUREMENTS WITH THERMAL NEUTRONS

From the level scheme drawn in Fig. 3 it is clear that after thermal neutron capture only \(\alpha\)-emission from the \(^{40}\text{Ar}\) compound nucleus to the ground state of \(^{36}\text{S}\) is possible. Only 3\(^-\) levels of \(^{40}\text{Ar}\) will be involved since \(\alpha\)-emission from a 4\(^-\) state is parity forbidden. In principle, also \((n_{\text{th}},\gamma\alpha)\) transitions can take place. For the \((n_{\text{th}},\alpha_0)\) transition a Q-value of 3.07 MeV corresponding to an \(\alpha\)-energy of 2.76 MeV is obtained using the masses from \[12\]. With an energy of 2.76 MeV for the particle to be detected, one has to be very cautious for the possible presence of 2.72 MeV background tritons produced by
(n,α) reactions in ⁶Li impurities in the sample backing and the detector environment, the ⁶Li(nₜh, α)t cross-section being 940 b.

![Diagram](image)

Figure 3: Level scheme illustrating the possible transitions after thermal neutron capture by ³⁹Ar.

### 3.1 Experimental procedure

The neutron irradiations were performed at the High Flux Reactor of the Institut Laue-Langevin (ILL) in Grenoble, France. A first lengthy measurement, comprising 1609 hours of neutron irradiation, took place at the end of the 87 m curved thermal neutron guide H22, where a flux of about 3.5 × 10⁸ n/cm² s is available. A schematic top-view of the experimental set-up is depicted in Fig. 4. The neutron beam entered the vacuum chamber through a thin Al window. The sample was mounted at 30° with respect to the neutron beam axis. A suited surface barrier detector was mounted parallel with the sample out of the beam and slightly collimated in order to avoid detection of particles under small incident angles. Using a 200 µm detector and doing measurements with the ³⁹Ar sample consecutively in and out of the neutron beam and with a dummy sample, made clear that, indeed, 2.72 MeV tritons were produced in ⁶Li impurities in the sample backing material and in the detector environment. In order to avoid this hindrance a 25 µm detector was used. Even under the smallest incident angles permitted by the collimation, the 2.72 MeV tritons are not completely stopped. In this way a clean detection of the 2.76 MeV α-particles was possible. The detector energy calibration was done by means of the ¹⁰B(nₜh,α)⁷Li and ⁶Li(nₜh,α)t reactions. The thermal neutron flux was regularly determined by means of the ²³⁵U(nₜh,f) reaction for which a cross-section value of 584.25 ± 1.10 b was adopted according to the ENDFB-VI data file. Therefore, the ³⁹Ar sample was replaced by a well-calibrated ²³⁵U sample strictly maintaining the geometric configuration. Nevertheless, in spite of the extremely long irradiation, no clear indication of an α-peak at 2.76 MeV nor a sign of (n,γα) transitions was observed.
Consequently, in an attempt to improve counting statistics, a modified experimental set-up was installed at the PF1 cold neutron guide, which delivers a much more intense neutron flux of $5 \times 10^9 \text{n/cm}^2\text{s}$. In addition to the higher neutron flux, a double Frisch-gridded gas flow ionization chamber, enabling a $2\pi$ detection geometry, was used (see Fig. 4). A gas mixture of 90% argon and 10% methane kept at a constant pressure of 1.1 bar was used as a detection gas. Also here, the detection chain was calibrated by means of the $^{10}\text{B}(n_{\text{th}},\alpha)^7\text{Li}$ and $^6\text{Li}(n_{\text{th}},\alpha)t$ reactions. The flux was constantly monitored by means of the $^{10}\text{B}(n_{\text{th}},\alpha)$ reaction for which a cross-section value of 3839 b was taken from the ENDFB-VI data file. In this set-up, however, the 2.72 MeV background tritons produced in the sample backing material are stopped in the effective area of the detector thus hampering a clean detection of the 2.76 MeV $\alpha$-particles. Doing comparative measurements with the sample turned over $180^\circ$ and with a dummy sample made clear that there was no $\alpha$-enhancement in the expected energy region. The effective side of the sample could be irradiated only for 20 hours due to the limited beamtime.

3.2 Results and discussion

For the transformation of the measured count rates into cross-sections the following formula was used:

$$\sigma_\alpha = \left( \frac{N_f}{N_{\text{Ar}}} \right) \left( \frac{C_\alpha}{C_f} \right) \sigma_f$$

where $N_f$ and $N_{\text{Ar}}$ are the number of atoms/cm$^2$ of the ‘flux’ (i.e. $^{235}\text{U}$ or $^{10}\text{B}$) and the $^{39}\text{Ar}$ samples, respectively, $C_f$ and $C_\alpha$ the count rates of the ‘flux’ (i.e. $^{235}\text{U}(n,f)$ or $^{10}\text{B}(n,\alpha)$) and $^{39}\text{Ar}(n_{\text{th}},\alpha)^{36}\text{S}$ reactions, respectively. Using the data from the flux calibration measurements and solving Eq. (1) for $C_\alpha$ a count rate of 1.14 counts per hour per barn was calculated for the first measurement at H22 and 452 counts per hour per barn for the measurement at PF1. The observed particle energy spectra were then used to derive an upper limit for the $^{39}\text{Ar}(n_{\text{th}},\alpha)^{36}\text{S}$ cross-section. Adopting a detection level of 3 times the standard deviation of the average background level and $\alpha$-peak width estimations from the different detector calibration measurements results in respective upper limits of 0.41 (H22 measurement) and 131 counts per hour (PF1 measurement) for the $^{39}\text{Ar}(n_{\text{th}},\alpha)^{36}\text{S}$
count rate. These values correspond to cross-section upper limits of respectively 0.36 b and 0.29 b, the H22 measurement and the one done at PF1 thus being in perfect agreement.

4 CONCLUSION

In the present work a first experimental investigation of the \( ^{39}\text{Ar}(n_{\text{th}},\alpha)^{36}\text{S} \) reaction cross-section has been performed. A sample containing \( 2.85 \times 10^{14} \) \(^{39}\text{Ar} \) atoms was produced at ISOLDE: \(^{39}\text{Ar} \) atoms were produced through spallation of a titanium oxide target and subsequently ionized and implanted at 60 keV into an Al foil. The number of sample atoms was determined through \( \beta \)-activity measurements at the IRMM. Thermal neutron irradiation of the sample at the High Flux Reactor of the ILL enabled the determination of an upper limit of 0.29 b for the \(^{39}\text{Ar}(n_{\text{th}},\alpha)^{36}\text{S} \) cross-section.

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