The ^{41}Ar(n,γ)^{42}Ar reaction

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Abstract. The cross-section of the thermal neutron capture ^{41}Ar(n,γ)^{42}Ar($t_{1/2}$=32.9 y) reaction was measured by irradiating a ^{40}Ar sample at the high-flux reactor of Institut Laue-Langevin (ILL) Grenoble, France. The signature of the two-neutron capture has been observed by measuring the growth curve and identifying the 1524.6 keV γ-lines of the shorter-lived ^{42}K(12.4 h) β^- daughter of ^{42}Ar. Our preliminary value of the ^{41}Ar(n,γ)^{42}Ar thermal cross section is 240(80) mb at 25.3 meV. For the first time, direct counting of ^{42}Ar was performed using the ultra-high sensitivity technique of noble gas accelerator mass spectrometry (NOGAMS) at Argonne National Laboratory, USA.

1 Introduction

Neutron capture reactions and their cross section are essential for basic and applied nuclear physics. It was recognized by Cameron [1] and Burbidge, Burbidge, Fowler and Hoyle [2] that they play a crucial role in stellar production of heavy elements. The quest for experimental determination of neutron capture cross sections has been intensely pursued for the study of the slow (s) process [3]. However, no experimental pathway exists to determine neutron capture rates on nuclei far from stability [4, 5] which are relevant to the rapid (r) process [6]. Various techniques have been proposed for providing indirect measurements of neutron-capture cross sections far from stability [7, 8]. Obtaining reliable data on neutron capture cross section for unstable isotopes remains a challenge and an essential task in contemporary research [9, 10].

Production of ^{42}Ar and its properties are not extensively studied. In the 1950's and 1960's, the half-life of ^{42}Ar was measured as 32.9±1.1 y and the cross section of the ^{41}Ar(n,γ)^{42}Ar reaction at thermal energy was determined as 0.5(1) b [11, 12]. ^{42}Ar(32.9 y) is thus known to undergo 100% β^- decay to shorter-lived ^{42}K(12.36 h), itself further β^- decaying to stable ^{42}Ca (Fig. 1). In an experiment approved at the National Ignition Facility (NIF) of Lawrence Livermore National Laboratory [13], we are considering ^{42}Ar as a candidate for the experimental observation of a rapid two-neutron capture reaction on ^{40}Ar. The extreme high-density plasma and high-density neutron environment of a laser-induced Inertial Confinement Fusion shot at NIF is the closest terrestrial analog of stellar explosive nucleosynthesis. The experiment will consist of a high-power laser shot on a DT filled capsule seeded with ^{40}Ar atoms, where ^{42}Ar could be produced by the two-neutron ^{40}Ar$(n,\gamma)^{41}$Ar$(n,\gamma)^{42}$Ar reaction within $\approx$ 100 ps.

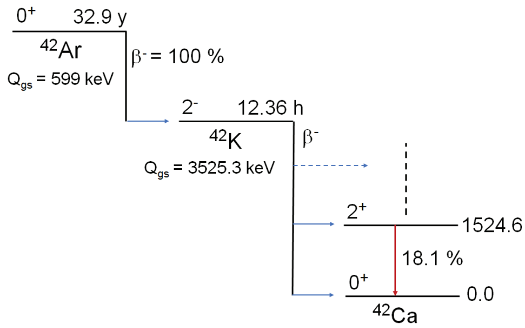

Figure 1. Simplified decay scheme of ^{42}Ar and ^{42}K.

The objectives of the present preparatory study, performed before the approved experiment at NIF, were twofold: (i) production of ^{42}Ar in a long irradiation of ^{40}Ar in a high flux of thermal neutrons and a new measurement of the ^{41}Ar$(n,\gamma)^{42}$Ar reaction cross, and (ii) first demonstration of direct detection of ^{42}Ar at ultra-high sensitivity, as required for the NIF experiment.

2 ^{40}Ar sample preparation and irradiation

A 0.768 cc high-purity quartz ampoule was filled with 99.992 % enriched ^{40}Ar gas [14] at 314(1) Torr and shipped to the high-flux reactor of Institut Laue-Langevin

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(ILL), Grenoble for irradiation. After a 8.17 days irradiation in the V4 beam tube with a thermal neutron fluence of $6.0(9)\times10^{20}$ cm^{-2} [15] and following decay of ^{41}Ar and short-lived activities co-produced in the quartz, the ampoule was shipped to Hebrew University. The ampoule was broken *in vacuo* in an *ad hoc* gas manifold (Fig. 2), quantitatively diluted with ^{nat}Ar ($N_{40} = 1.92(15)\times10^{21}$) to reach an atom ratio ^{42}Ar/^{40}Ar in the 10^{-12} range (see Section 4), and cryogenically transferred to a 11.2 cc stainless cylinder (sample ILL1) at a final pressure of 6.96(35) bar (25 °C). The measured Ar dilution factor between the original ampoule and sample ILL1 is 246(20).

Figure 2. Manifold used for the dilution of the irradiated Ar-gas with ^{nat}Ar. The ILL1 sample cylinder is at the right-hand end of the vacuum line.

3 ^{42}Ar detection using γ-ray spectrometry

Sample ILL1 was placed in contact with an efficiency-calibrated HPGe detector (Fig. 3) to follow the growth curve of shorter-lived β^- daughter ^{42}K via the 1524.6 keV γ-ray (18.1% intensity per decay) (Fig. 1). The γ spectra

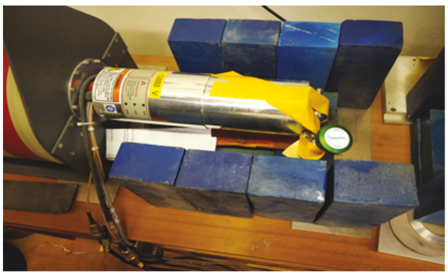

Figure 3. The ILL1 sample cylinder in contact with the end-cap of the HPGe detector at Hebrew University for the ^{42}K in-growth measurement.

of the cylinder were measured every 12.5 hours; Figure 4 shows the mean decay rate for each measurement interval, reaching $\approx$ 38 counts per hour at secular equilibrium in the given configuration. The cylinder was then placed at a 5 cm distance (Fig. 5) for the activity measurement. The γ-spectrum, accumulated for 196.9 hours, is shown in

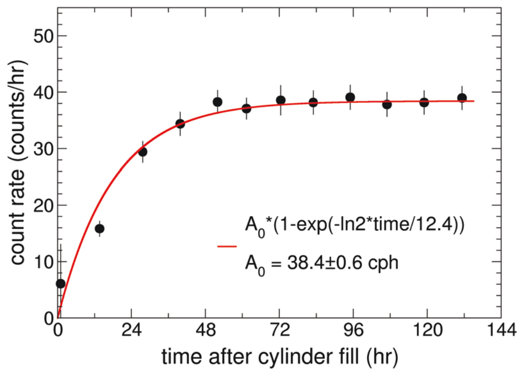

Figure 4. In-growth of ^{42}K (12.36 h) activity from decay of ^{42}Ar(32.9 y) produced by slow two-neutron capture ^{40}Ar(n,γ)^{41}Ar(n,γ)^{42}Ar. The solid line represents a fit to the data points with the expression $A(t) = A_{42}(1 - e^{-\lambda_{42K}t})$ where λ_{42K} is the ^{42}K decay constant, confirming production of ^{42}Ar.

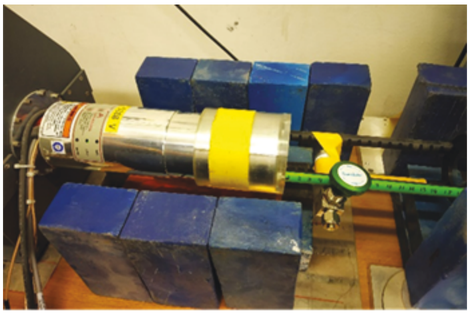

Figure 5. The ILL1 sample cylinder was placed at a 5 cm distance from the end-cap of the HPGe detector for activity measurement.

Fig. 6. A correction due to the extended geometry of sample ILL1 and attenuation in the cylinder walls, calculated by a Monte Carlo simulation, was applied. The measured ^{42}K activity at secular equilibrium (equal to that of ^{42}Ar) is $A_{42} = 4.0(3)$ Bq.

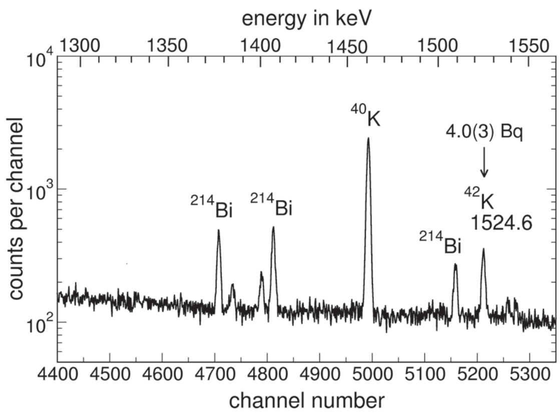

Figure 6. γ spectrum of sample ILL1 at secular equilibrium (8.21 days counting at 5 cm distance from the HPGe detector). The ^{42}K (^{42}Ar) activity is 4.0(3) Bq. Room background lines are identified.

4 Determination of the thermal ^{41}Ar(n,γ)^{42}Ar cross-section

The number of ^{42}Ar atoms in sample ILL1 calculated from the ^{42}Ar activity is $N_{^{42}Ar} = A_{42}/\lambda_{^{42}Ar} = 6.0(5)\times10^9$, where $\lambda_{^{42}Ar}$ is ^{42}Ar(32.9 y) decay constant. We extract now an atom ratio ^{42}Ar/^{40}Ar=3.12(35) $\times\ 10^{-12}$ using the number N_{40} of ^{40}Ar atoms in sample ILL1 (see Section 2). The atom ratio in the original irradiated ampoule is obtained using the measured dilution ratio (246(5), see Section 2) as ^{42}Ar/^{40}Ar= 7.7(9) $\times\ 10^{-10}$.

The expression for the ratio R =^{42}Ar/^{40}Ar produced by the slow two-neutron capture ^{40}Ar(n,γ)^{41}Ar(n,γ)^{42}Ar is given by:

$$R(t_i) = \frac{{\Phi_n}^2 \sigma_{^{40}Ar(n,\gamma)} \sigma_{^{41}Ar(n,\gamma)}}{\lambda_{41}} \left(t_i - \frac{1 - e^{-\lambda_{41} t_i}}{\lambda_{41}} \right), \quad (1)$$

where Φ_n and t_i represent the mean thermal neutron flux and irradiation time, respectively, $\sigma_{^{40}Ar(n,\gamma)}$ and $\sigma_{^{41}Ar(n,\gamma)}$ the cross section of the respective reactions at thermal neutron energy, and λ_{41}, the decay constant of ^{41}Ar($t_{1/2}$ = 109.61(4) min).The $\sigma^{2200}_{^{40}Ar(n,\gamma)}$ cross section (25.3 meV) is taken as 0.673(65) b [16]; decay of ^{42}Ar($t_{1/2}$ = 32.9 y) is neglected.

Substitution of all values results in a preliminary value of $\sigma^{2200}_{^{41}Ar(n,\gamma)}$ = 240(80) mb for the thermal neutron capture ^{41}Ar$(n,\gamma)^{42}$Ar reaction cross section, significantly smaller than the value of 0.5(1) b reported in [12]. A second ^{40}Ar irradiation is planned at ILL for improved neutron fluence monitoring.

5 Detection of ^{42}Ar by Noble-Gas Accelerator Mass Spectrometry

Accelerator mass spectrometry (AMS) is an ultra-sensitive technique for detection of rare long-lived radionuclides. Single ions are counted after acceleration and a succession of magnetic and electrostatic analyses and identification, usually by nuclear detection methods; see recent reviews in [17, 18]. While conventional AMS facilities are based on negative-ion production and injection, AMS analysis of noble gases must resort to positive-ion production due to the instability of their negative ions. The positive-ion Noble-Gas Accelerator Mass Spectrometry (NOGAMS) technique, developed at Argonne National Laboratory (ANL) is described in detail in [19]. It was used to detect long-lived ^{39}Ar(268 y) [20, 21] at isotopic abundance sensitivity, ^{39}Ar/Ar, in the range 10^{-12} to below 10^{-16}. In this work, we developed the NOGAMS method for detection of ^{42}Ar to be used in the analysis of Ar samples from a NIF shot (see Section 1).

The electron cyclotron resonance ion source ECR-III [22] was fed (at a partial pressure in the low 10^{-7} Torr range) with the ^{42}Ar gas sample ILL1 (Section 2). Highly-charged $^{42}Ar^{8+}$ ions were accelerated to 5.5 MeV/u in the ATLAS superconducting linear accelerator at ANL. The ions were then analyzed in a split-pole Enge spectrograph in gas-filled mode [23] to separate isobaric ^{42}Ca

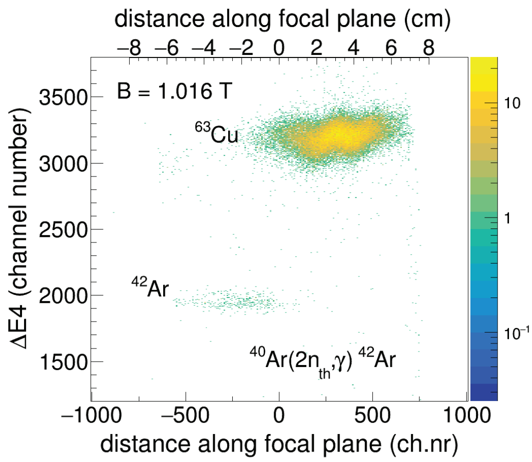

Figure 7. First NOGAMS identification spectrum of ^{42}Ar from sample ILL1: energy loss in anode ΔE4 of the Monica detector vs focal plane position. See text.

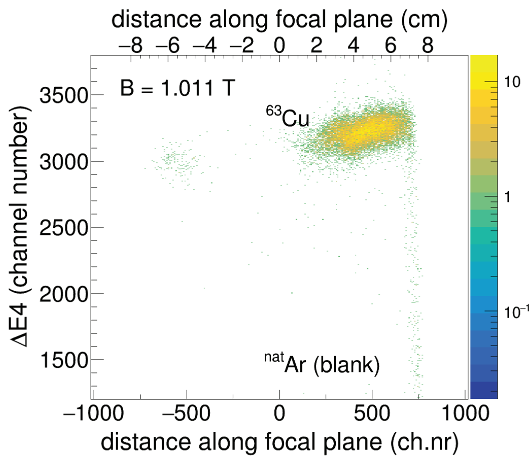

Figure 8. Spectrum taken under the same conditions of ATLAS as in Fig-7 for a nonirradiated (blank) ^{nat}Ar sample for the background subtraction.

and other beam contaminants resulting from ion source impurities. The position-sensitive focal-plane ionization chamber, nicknamed Monica [24], was used to identify and count incoming ions by measuring their energy loss and positions. Fig. 7 shows the identified ^{42}Ar group. A ^{63}Cu group, well separated from ^{42}Ar, is observed originating from $^{63}Cu^{12+}$ ions, which are degenerate in their mass-to-charge ratio with $^{42}Ar^{8+}$ ions, and transported identically through ATLAS. The ^{63}Cu ions were likely produced from materials present in the microwave injection system. Isobaric $^{42}Ca^{8+}$ contaminant ions were observed but are totally deflected out of the detector acceptance by the gas-filled spectrograph at the magnetic field setting used. The ^{42}Ar count rate for the gas sample used (^{42}Ar/^{40}Ar=3.1$\times10^{-12}$) was 6.8 counts per hour (cph). For comparison, Fig. 8 shows a spectrum obtained for a ^{nat}Ar gas sample under similar conditions as ILL1. No counts are observed in the ^{42}Ar region (< 0.02 cph) demonstrating an abundance sensitivity in the 10^{-15} range. Further quantitative analysis of the isotopic ratio ^{42}Ar/Ar obtained from the NOGAMS data will allow us to confirm or correct the ^{41}Ar(n,γ)^{42}Ar cross-section value and the ^{42}Ar half-life value.

6 Summary

A ^{40}Ar sample was irradiated at the high-flux nuclear reactor at ILL, Grenoble. Two successive neutron captures by ^{40}Ar produced ^{42}Ar through the ^{40}Ar(n, γ)^{41}Ar(n, γ)^{42}Ar reactions. ^{42}Ar was identified by observation of the growth curve of its β^- daughter ^{42}K and of the subsequent 1524.6 keV γ-transition. The ^{42}Ar sample activity was measured as 4.0(3) Bq. From the corresponding ^{42}Ar/^{40}Ar atom ratio in the irradiated sample, a preliminary value of 240(80) mb is determined for the cross-section of the ^{41}Ar(n, γ)^{42}Ar reaction at thermal energy. In addition, for the first time ^{42}Ar, with an isotopic abundance in the 10^{-12} range, was directly identified and counted by noble-gas accelerator mass spectrometry. An abundance sensitivity in the 10^{-15} range is demonstrated. We plan to apply the technique to a search for ^{42}Ar produced by a rapid two-neutron capture in a high-power laser shot on a DT filled capsule seeded with ^{40}Ar atoms at NIF.

Acknowledgements

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