Stellar $^{78,80,84,86}$Kr(n,γ) Reactions Studied by Activation at SARAF-LiLiT, Atom Trap Trace Analysis and Decay Counting

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Received xxxxxx
Accepted for publication xxxxxx
Published xxxxxx

Abstract

We report on (n,γ) neutron capture experiments performed with the Liquid-Lithium Target (LiLiT) and the mA-proton beam at 1.92 MeV (2-3 kW) from the Soreq Applied Research Accelerator Facility (SARAF). The setup yields high-intensity 30-keV quasi-Maxwellian neutrons (3-5×10$^{10}$ n/s) closely reproducing the conditions of s-process stellar nucleosynthesis. The $^{78,80,84,86}$Kr(n,γ) reactions at the border between weak- and strong- s-process were studied. A Ti sphere filled with 107.7 mg of natural Kr gas was placed in an irradiation chamber downstream of LiLiT with a gold foil used as a neutron fluence monitor during the activation. The activities of the short-lived Kr isotopes ($^{79,85m,87}$Kr) were measured by γ decay counting with a HPGe detector. The long-lived Kr isotopes ($^{81,85g}$Kr) were measured by atom counting via Atom Trap Trace Analysis (ATTA) at Argonne and Low-Level Counting (LLC) at Bern.

Keywords: s-process, $^{84,86}$Kr(n,g), $^7$Li(p,n), liquid-lithium target, high-intensity neutron source, Maxwellian Averaged Cross Section (MACS), Atom Trap Trace Analysis (ATTA), Low Level Counting (LLC)

Neutron-induced reactions remain at the forefront of experimental investigations for the understanding of stellar nucleosynthesis and chemical evolution of the Galaxy. We report on recent experiments performed with the mA-proton beam at 1.92 MeV (2-3 kW) from the Soreq Applied Research Accelerator Facility (SARAF) and the Liquid-Lithium Target (LiLiT), yielding high-intensity (3-5×10$^{10}$ n/s) quasi-Maxwellian neutrons at ~30 keV [1-4], close to the thermal conditions of the stellar slow (s-) neutron capture process. The SARAF accelerator is a superconducting linear RF accelerator designed to produce a beam of light ions ($m/q \leq 2$) with a continuous-wave (CW) beam intensity up to 5 mA. The project, initiated in the early 2000s with state-of-the-art technology, was divided into a Phase I (as used so far) with an energy range up to 4 (5)MeV for protons (deuterons) to be followed by a Phase II (presently under construction) with a maximum energy of 35 (40) MeV. The layout of Phase I is shown in fig. 1. Light-ion H’ or D’ are extracted at 20 keV/u from an Electron Cyclotron Resonance Ion Source (ECRIS), mass-analyzed and injected via a Low-Energy Beam Transport (LEBT) line to a 176MHz four-rod Radio-Frequency Quadrupole (RFQ) with a final energy of 1.5MeV/u. A short Medium-Energy Beam Transport (MEBT) line transports the
beam to a Prototype Superconducting Module (PSM) housing six Half-Wave Resonators (HWR) followed by the High Energy Beam (HEBT) line towards the LiLiT target at Station 1 (fig. 1) via two 45° bending magnets.

Fig. 1: Schematic diagram of the SARAF Phase I high-intensity superconducting linear accelerator and the LiLiT beam line. See text for component acronyms.

Fig. 2: (left) Schematic illustration of the physical principle of the LiLiT system (see text). For clarity, one side wall of the nozzle region was removed in the drawing. The incident proton beam impinges directly on the liquid lithium flow (windowless) and within the first μs produces a flux of neutrons, kinematically forward-focused if the incident energy is close to the neutron threshold. The protons are stopped within the lithium film and the beam power (of the order of MW/cm³, see [2-4]) is conveyed by the flow to a heat exchanger. (right) View of the free-surface liquid lithium flow photographed from the target chamber viewpoint.

The Liquid Lithium Target (LiLiT) was specifically designed and built to dissipate the high power of the CW proton beam from SARAF (up to ≈ 5kW) and produce quasi-Maxwellian neutrons to be used for nuclear astrophysics experiments [2]. Figure 2 illustrates the principle of the target. Liquid lithium is forced-flown from the top into a 15mm wide, 1.8mm thick film supported by a thin stainless steel wall (concave as viewed in the proton beam direction). The proton beam impinges directly (windowless) onto the lithium surface emitting neutrons mainly in the forward direction (if the incident energy is close to the neutron threshold). The liquid lithium acts both as a (thick) target producing neutrons near the surface (∼ 4 μm deep) and as a beam dump where the high power (∼ 0.7MW/cm³/mA at the Bragg peak depth, see [2-4]) is mechanically transported by the fast flow (2–5m/s) to a reservoir and heat exchanger (see [2-4] for details).

First experiments were dedicated to benchmark the experimental system by measuring the Maxwellian Averaged Cross Section (MACS) of several targets. The MACS of $^{94}$Zr and $^{96}$Zr, important isotopes for understanding the s-process evolution, were determined as 28.0 ± 0.6 mb and 12.4 ± 0.5 mb respectively [5], based on activation measurements and detailed analysis, in good agreement with previous measurements, with lower uncertainties. Using α-, β-, γ-spectrometry and accelerator mass spectrometry, we extended our experimental studies of neutron capture reactions to several targets of astrophysical interest [6-10].

The neutron capture cross sections of the Kr isotopes are of special interest for s-process nucleosynthesis [11,12]. Krypton has isotopes, $^{78,80,82}$Kr, the abundances of which are solely due to the s-process nucleosynthesis due to the fact that they are shielded from the r-process by $^{78,80,82}$Se (Fig. 3).
As it is impossible to determine the solar abundances of Kr via the analysis of primitive meteorites (carbonaceous chondrites CI) [13], the calculation of these abundances via s-process nucleosynthesis is an important and reliable alternative. Kr is a prominent example for an isotopic anomalous component in meteorites of pure s-process pattern [14, 15]. The test that the isotopic pattern is really an s-process pattern depends strongly on the reliability of the neutron capture cross sections. The importance of the s-process branching at 85Kr has been widely discussed [16-18]. This branching point is the only one from which the neutron density of the weak s-process can practically be derived as the laboratory half-life of 85Kr is unchanged under stellar conditions. Moreover, the existence of a short-lived isomeric state 85mKr (4.5 h) introduces two independent 84Kr(n,γ) neutron capture paths. The isomeric state is populated preferentially to the ground state 85gKr (10.8 y), due in part to the higher spin of the latter. Short-lived 85mKr does not thermally equilibrate before decaying preferentially (79%) by β to 85Rb than by internal transition to the longer-lived ground state, feeding then 85Rb.

We describe and give here preliminary results of the natKr(n,γ) experiments at stellar energies. The short lived Kr isotopes (79,85m,87Kr) were measured by γ decay counting with a HPGe detector. The long-lived Kr isotopes (81,85gKr) were measured by atom counting via Atom Trap Trace Analysis (ATTA) at Argonne and Low-Level Counting (LLC) at Bern.

natKr gas, encapsulated in a small sphere (Fig. 4) made of titanium (diameter = 10 mm, shell thickness 0.2 mm [19]), was irradiated at SARAF-LiLiT.
The activities of the short-lived Kr isotopes ($^{79,85m,87}$Kr) were measured with a High-Purity Ge (HPGe) detector. An example of one of the spectra is presented in Fig. 5. The $^{85m}$Kr ($t_{1/2} = 10.8$ y) nuclide was detected by low-level $\beta$ counting [20] and by $\gamma$ spectrometry. The long-lived Kr isotopes $^{81}$Kr ($t_{1/2} = 2.3 \times 10^5$ y) and $^{85}$Kr (10.8 y) were measured by Atom Trap Trace Analysis (ATTA) [21] at the ATTA-3 laboratory at ANL [22]. A schematic diagram of the ATTA setup at ANL is presented in fig. 6: an atom of a particular isotope is selectively captured and repeatedly excited by resonant laser light in a magneto-optical trap (MOT) and detected by observing its fluorescence. The isotopic ratios are measured by trapping successively the rare isotope and a reference isotope by shifting the laser frequency accordingly for an activated sample and comparing the measured ratios to those of a Kr sample considered as standard. Low-level-counting measurements are preferably performed in a low-background environment. One such state-of-the-art laboratory is located 35 meters (70 m. w.e) below the surface in an underground laboratory at the University of Bern in Switzerland. 99.6% of $^{85}$Kr decays by $\beta$-radiation with a maximum energy of 0.69 MeV without $\gamma$-emission. Gas proportional counting is applied for the identification of $^{85}$Kr decay. Because of the relatively high $\beta$-energy, the energy spectrum above the 3 keV electronic cut-off is taken into account. The deposition of the $\beta$-energy within the gas volume increases with pressure. In addition, a smaller volume of the counters reduces the background because the surface (and hence the influence) of self activity of the counter material (mostly high purity copper) are smaller. Gas proportional counters of different volumes (10-100 mL), pressures (10-23 bar) and methane/argon admixtures (p5-p40) are used at Bern. Depending on the configuration, counting time for $^{85}$Kr ranges from 3 days to 6 days [23].
Fig. 6: Diagram of the cooling and trapping stages on the ATTA beamline at ANL. The total length of the atomic beamline is approximately 2 m. Lasers and optics are located on an adjacent laser table of a similar length.

The preliminary results of the number of nuclei activated at the end of the irradiation, \( N_{\text{act}} \), and the \(^{84}\text{Kr}/\text{Kr} \) ratio are presented in Table 1.

Table 1: Summary of the preliminary \(^{84}\text{Kr}/\text{Kr} \) nuclei ratio and the number of nuclei activated via the \(^{84}\text{Kr}(n,\gamma) \) reactions at the end of the irradiation, \( N_{\text{act}} \).

| Nuclei    | \(^{84}\text{Kr}/\text{Kr} \) | \( N_{\text{act}} \)   | detection method          |
|-----------|-------------------------------|------------------------|---------------------------|
| \(^{79}\text{Kr} \) | \( 3.6(3) \times 10^{-13} \) | \( 2.8(2) \times 10^8 \) | \( \gamma \)-spectroscopy |
| \(^{81}\text{Kr} \) | under analysis                |                        | ATTA                      |
| \(^{85m}\text{Kr} \) | \( 3.2(1) \times 10^{-12} \) | \( 2.5(1) \times 10^9 \) | \( \gamma \)-spectroscopy |
| \(^{85}\text{Kr} \) | under analysis                |                        | \( \gamma/\text{ATTA/LLC} \) |
| \(^{87}\text{Kr} \) | \( 2.00(5) \times 10^{-13} \) | \( 1.55(4) \times 10^8 \) | \( \gamma \)-spectroscopy |

Acknowledgements

We would like to thank the SARAF and LiLiT (Soreq NRC) operation staffs for their dedicated help during the experiments. This work was supported in part by Pazy Foundation (Israel) and by Israel Science Foundation (Grant Nr. 1387/15). M.T. gratefully acknowledges the support of the Eshkol Foundation of Israel Ministry of Science and Technology.

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