



First experimental results of the $^{33}\text{S}(\alpha, p)^{36}\text{Cl}$ cross section for production in the Early Solar System

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ABSTRACT

Short-lived radionuclides (SLRs), with $t_{1/2} < \sim 5$ Ma, were extant in the Early Solar System (ESS). Their relatively short lifetimes and different chemical behavior make them useful for dating processes in the ESS. The existence of the SLR ^{36}Cl ($t_{1/2} = 3.01 \times 10^5$ a) has been determined from correlation between isotopic enrichment of its daughter isotope, ^{36}S , and Cl abundance in meteorites. The relatively high inferred initial Solar System $^{36}\text{Cl}/\text{Cl}$ ratio strongly suggests that ^{36}Cl was produced in the ESS by bombardment of solar energetic particles on gas and dust in the protoplanetary disc. A major uncertainty in the irradiation models is the absence of experimental data. Experimental data on key ^{36}Cl production reactions will help constrain the calculated yields. Of the reactions considered in the ESS models, the $^{33}\text{S}(\alpha, p)^{36}\text{Cl}$ reaction is one of the most important. Here we report first results of a measurement of the averaged reaction cross section in the energy range 1.84–2.04 MeV/A. Our result, 192 ± 33 mb (1σ), is significantly higher than results of previous calculations, 102 and 34 mb.

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1. Introduction

Short-lived radionuclides (SLRs), with $t_{1/2} < \sim 5$ million years (Ma), e.g., ^{10}Be , ^{26}Al , ^{36}Cl , ^{41}Ca , ^{53}Mn , ^{60}Fe , were extant in the Early Solar System (ESS). The evidence for their existence has been determined from correlations between their elemental abundances and isotopic enrichments of their daughter nuclides in meteorites. The short half-lives relative to the 4.568 Ga age of the Solar System (SS), makes them good chronometers for dating planetary processes in the ESS. In addition, inferring their abundances in the ESS contributes to our understanding of the nucleosynthesis processes which produced them. However, the origin of the SLRs is still a matter of debate. The large mass range of observed SLRs indicates that they were produced by different nucleosynthetic processes in different astronomical sites. Two sources were proposed:

- (1) Production in supernovae (SNe) or asymptotic giant branch (AGB) stars, and injection into the Solar Nebula [1–5]. In this case, the supernova shockwave [4] or strong stellar wind from the AGB star [5] could have triggered the collapse of the Solar Nebula to form the SS, while distributing freshly synthesized SLRs. The injection could have occurred as well later in the SS formation process, into the already formed protoplanetary disc [6].

- (2) Production in the ESS by bombardment of solar energetic particles (SEPs), from the young pre-main sequence Sun, on gas and dust in the protoplanetary disc. Light SEPs (p, α , ^3He) with energies in excess of 1 MeV/A, are thought to be associated with stellar X-ray flares. Intense X-ray luminosities are supported by measurements of young stellar objects (YSOs) in the Orion Nebular Cluster [7]. The X-wind model [8] attempts to explain production of SLRs in the protoplanetary disc during the T Tauri stage of the young Sun. This irradiation could have occurred as well during the late transition disk stage of the protoplanetary disc [9].

The existence of ^{36}Cl ($t_{1/2} = 0.301$ Ma) in the ESS was inferred from excesses of its daughter isotope, ^{36}S , in the chlorine-rich minerals (e.g., sodalite and wadalite) in meteorites, which is correlated with Cl abundances [10–13]. Chlorine-rich minerals are secondary products of alteration of primary minerals in the meteorites. Hence, ^{36}Cl is used to date late stage alteration processes in meteorites.

Available meteorite data on ^{36}Cl shows variations in the ESS abundances of ^{36}Cl . These inconsistencies are interpreted as due in part to uncertainties in the mechanism that introduced ^{36}Cl to the system as well as the time scale of the alteration process. However, ^{36}Cl abundances in the ESS are higher than production model calculations in both SNe and AGB stars [3]. This suggests that ^{36}Cl was produced by SEP irradiation from the proto-Sun. At the same time, little experimental cross section data are available for the relevant production reactions. Instead, model calculations use

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Hauser-Feshbach approximations, which are based on data for similar reactions on lower and higher mass targets [14–17]. The lack of experimental data accounts for a significant fraction of the overall uncertainty in the calculated yields [15].

The production rate P_{ij} by bombardment of SEP j on target nuclide i is calculated by the general equation

$$P_{ij} = \sum_i N_i \sum_i \int_0^\infty F_j(E) \sigma_{ij}(E) dE,$$

where N_i is the number of target nuclei i , $F_j(E)$ is the flux of SEP j as a function of energy and $\sigma_{ij}(E)$ is the energy dependent reaction cross section. The projectile flux is generally represented by the power law, $F_j(E) = E^{-p}$, where the value of p usually varies between 2 and 5. Lower values of p and near solar He/H and ${}^3\text{He}/{}^4\text{He}$ ratios correspond to gradual events. Impulsive events have a sharper energy spectrum (higher values of p) and are enriched in ${}^3\text{He}$ and ${}^4\text{He}$. The relative contributions of gradual and impulsive events are usually left as an adjustable parameter. A number of reactions are considered in the irradiation models. These include ${}^{33}\text{S}(\alpha, p)$, ${}^{34}\text{S}(\alpha, pn)$, ${}^{36}\text{S}(p, n)$, ${}^{34}\text{S}({}^3\text{He}, p)$, and ${}^{35}\text{Cl}({}^3\text{He}, 2p)$ [14,16]. According to the models, ${}^{33}\text{S}(\alpha, p)$ ${}^{36}\text{Cl}$ was a major contributor to the production of ${}^{36}\text{Cl}$ in the ESS, which is why we decided to measure the cross section of this reaction.

We have developed a procedure to measure ${}^{36}\text{Cl}$ by accelerator mass spectrometry (AMS) at the Nuclear Science Laboratory (NSL) at University of Notre Dame using a gas-filled magnet (see Section 2.3). We present here the results of the first cross section measurement of an energy range of 1.84–2.04 MeV/A to prove the feasibility of the technique for future measurements.

2. Experimental outline

The measurement of the ${}^{33}\text{S}(\alpha, p){}^{36}\text{Cl}$ reaction cross section is separated into three steps: (1) Activation of a target and collection of produced ${}^{36}\text{Cl}$; (2) chemical separation of the ${}^{36}\text{Cl}$ and precipitation as a usable cathode material, AgCl; (3) AMS counting of the ${}^{36}\text{Cl}$. Performing the measurement with AMS has distinct advantages. ${}^{36}\text{Cl}$ has no measurable γ -lines and its half-life is too long for β -counting at low abundances, making AMS a desirable technique to study this reaction. ${}^{36}\text{Cl}$ has been measured in rock and ground water samples with AMS in many laboratories. The difficulties and methods associated with measuring ${}^{36}\text{Cl}$ with AMS are well understood which helped in the development of the technique for our measurement.

2.1. Activation

The experiment was performed at the Nuclear Science Lab at the University of Notre Dame. The activation was done in inverse kinematics with a ${}^{33}\text{S}$ beam incident upon a He gas cell at the end of the beamline. This allowed us to have a pure ${}^4\text{He}$ target and ${}^{33}\text{S}$ beam without the use of material enriched in ${}^{33}\text{S}$. The ${}^{33}\text{S}$ was extracted out of a FeS cathode from a MC-SNICS (Multi-Cathode Source of Negative Ions via Cesium Sputtering) source. The beam was then injected into the FN tandem accelerator at 9 MV. Selecting the 9^+ charge state after the analyzing magnet gave us a beam energy of 90 MeV. The gas cell was previously constructed in the measurement of the ${}^{40}\text{Ca}(\alpha, \gamma){}^{44}\text{Ti}$ reaction [18]. A 2.5 μm thick Ni foil (99.95%) separated the gas cell from the beamline. The Ni foil was continuously rotated to avoid overheating. High-purity helium gas filled the gas cell at the pressure of 5 Torr. The pressure and flow was controlled by a gas handling system which held the gas cell at constant pressure. A 0.25 mm thick Al foil (99.999%) was placed 24 cm from the entrance foil to collect the ${}^{36}\text{Cl}$ produced. Both the gas cell and catcher were isolated to

allow charge current integration throughout the activation run. The sample was activated for about 77 h with an average beam intensity of 37 nA on target. To account for any contaminations in the beam or from the beam interacting with the entrance or catcher foils, a 42 h run with the gas cell filled with argon gas was performed. The gas pressure was set so the ${}^{33}\text{S}$ ions would have the same energy loss as they did in the helium [19].

2.2. Chemistry

The samples were left for a few months to allow for any short-lived activities to decay. The chemistry was then performed at Purdue University's PRIME lab. Three samples were prepared. The "Blank" sample was an unused aluminum foil identical to the ones used in the experiment. The "Beam Blank" sample was the catcher used for the run with the gas cell filled with argon. The "Activated" sample was the foil used in the reaction run. All samples were prepared with 50 g stable chlorine carrier (chlorine concentration of 1.101 mg/g) to fix the ${}^{36}\text{Cl}/\text{Cl}$ ratio and producing 55 mg of chlorine. Each foil was then cut into 8 pieces and added to the carrier. Roughly 25 mL of HNO_3 were added to each sample. Since Al oxidizes in nitric acid, which inhibits the dissolving of the aluminum, 2–3 mL of HF were added to each sample to prevent oxidation. After 3 h, 25 mL more of HNO_3 were added to the aliquots and left to dissolve overnight. The next morning, 45 mL of the samples were decanted into new vials. 2–3 mL of AgNO_3 were added to the aliquot forming a AgCl supernatant. The vials were centrifuged and the excess solution was decanted away. The decanting and centrifuging was repeated until all the samples were collected. It was assumed that ${}^{36}\text{Cl}$ and the added Cl carrier were recovered with the same efficiency. Samples were finally baked at 70 °C for three days to remove any remaining moisture.

2.3. AMS measurement

The samples, now in the form of AgCl, were packed into aluminum cathodes. To reduce any possible sulfur contamination, the cathodes and loading tools went through a chemical cleaning procedure that included HNO_3 and NaOH. To further reduce possible sulfur contamination, each cathode base was packed with AgBr.

The AMS measurement was performed with the Magnet for Astrophysical Nucleosynthesis studies Through Isobar Separation (MANTIS) AMS system at the Nuclear Science Lab [20,21]. The original tune was done with a 74.7 MeV ${}^{35}\text{Cl}^{8+}$ beam at a terminal voltage of 8.3 MV. This beam was tuned through the beamline to a Faraday cup in the scattering chamber located before the spectrograph (FC5 in Fig. 1). All magnetic elements were then scaled to a ${}^{37}\text{Cl}$ beam based on the frequency of the NMR probe in the analyzing magnet. To avoid hysteresis in the SNICS magnet, its magnetic field was left unchanged. To inject the different mass, the platform voltage (called bias voltage from here after) of the ion source was changed. Since ${}^{37}\text{Cl}$ has a slightly different injection energy due to the smaller bias voltage, the terminal voltage was adjusted to maximize beam output.

The MANTIS system uses a Browne-Buechner spectrograph in which the ions are bent in a 1 m radius, single dipole magnet equipped with two detectors. The first is a Parallel-Grid Avalanche Counter (PGAC) that gives the magnetic rigidity of the beam as it exits the dipole magnet. A 4-way split anode Ionization Chamber (IC) sits atop the PGAC. The PGAC and IC were filled with isobutane at 3 Torr and 8.5 Torr, respectively, controlled by separate gas handling systems. The two detectors were separated from each other and from the spectrograph by 350 $\mu\text{g}/\text{cm}^2$ Mylar windows.

Since the ${}^{36}\text{Cl}$ mass is almost identical to that of its isobar, ${}^{36}\text{S}$, we used the Gas Filled Magnet (GFM) approach, with 2.3 Torr of N_2 gas, to separate the isobars [22]. The spectrograph was isolated

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