

Helium-3 Cross Sections and Implications for Early Solar System History

M.W. Caffee ^a, T. Faestermann, R. Hertenberg, G.F. Herzog ^b, G. Korschinek, I. Leya ^c,
R.C. Reedy ^d, and J.M. Sistierson ^e

^a Dept. Physics, Purdue University, W. Lafayette, IN 47907

^b Dept. Chemistry & Chem. Biol., Rutgers University, Piscataway, NJ 08854-8066

^c Physikalisches Institut, U. Bern, CH 3012, Bern, Switzerland

^d Earth Planetary Sciences, U. New Mexico, Albuquerque, NM 87131

^e Francis H. Burr Proton Therapy Center, Massachusetts General Hospital, Boston MA 02114

Early in the history of the solar system, an intense particle irradiation may have formed a portion of the ²⁶Al, ³⁶Cl, and ⁴¹Ca that left footprints in the isotope abundances of Mg, S, and Ca [1,2]. First attempts at modeling the irradiation considered only ¹H and ⁴He as nuclear-active particles [3,4]. Too little ²⁶Al resulted. Addition of ³He to the mix of nuclear-active particles increased ²⁶Al production, but gave too much ⁴¹Ca [3]. Lacking the measured ³He cross sections needed for these calculations, modelers relied on values generated by nuclear physics codes or adapted from studies of similar nuclear reactions [3,5]. To try to improve the input to the modeling calculations, we built on the work of [6] by measuring cross sections for the reactions of ³He with ^{nat}Mg and Al that produce ²⁶Al and the ones with Ca that produce ³⁶Cl. We then incorporated the cross sections into a model developed by [7].

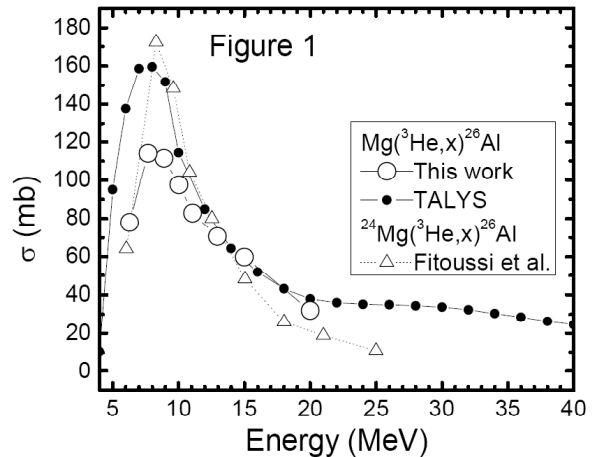
Targets: For the measurement of Mg cross sections, we used magnesium disks 10 mm in diameter, 10 μm thick, and with a purity of 99.9%. Our source of calcium was a sample of Cal-White, a finely ground limestone product manufactured as a paint pigment by Imerys. This material consists of 80.05 wt% CaCO₃, 15.6 wt% MgCO₃, and 4.2 wt% acid-insoluble residue. Semi quantitative XRF analysis of the residue gave 4.2 wt% Mg; 5.8 wt% Al; 34.1 wt% Si; 3.3 wt% K; 0.2 wt% Ti; and 0.4 wt% Fe. We fabricated Ca sandwich targets by suspending the limestone in xylenes, depositing ~6 mg of limestone on a 0.5 inch disk of Reynolds® Al foil, capping the deposited limestone with a second Al foil, and pressing the assembly hydraulically. The Al foils had a thickness of 4.55 ± 0.18 mg/cm² or 16.8 ± 0.7 μm assuming a density of 2.702 g/cm³. Reynolds Al foil typically contains 0.5 to 0.6 wt% silicon and from 0.6 to 0.9 wt% iron [8].

Irradiation: Each target was irradiated separately with ³He. ³He energies ranged from 15 to 36 MeV. To adjust the energy of the ³He beam downward from 15 MeV, we placed varying numbers of high-purity aluminum foils, each with a thickness of 12.7 μm in front of the targets. High-purity (>99.9 wt %) nickel foils with a thickness of 2 μm served as downstream catchers for all Mg targets and as upstream catchers for targets Mg4-Mg8. We did not include catcher foils, either up- or downstream, for the Ca sandwich targets. Fluences of ³He were measured with a Faraday cup placed 92 mm downstream from the target. The incident and exit energies were calculated by tracing 15,000 ³He particles through each target stack using TRIM [9]. From the particles' coordinates and direction cosines on exit, we calculated how many of them missed the Faraday cup. The correction factors are largest for the foils with the greatest number of degraders. Corrections were <6 % for 12 of the

16 targets, ~15 % in Ca7 and Mg8.

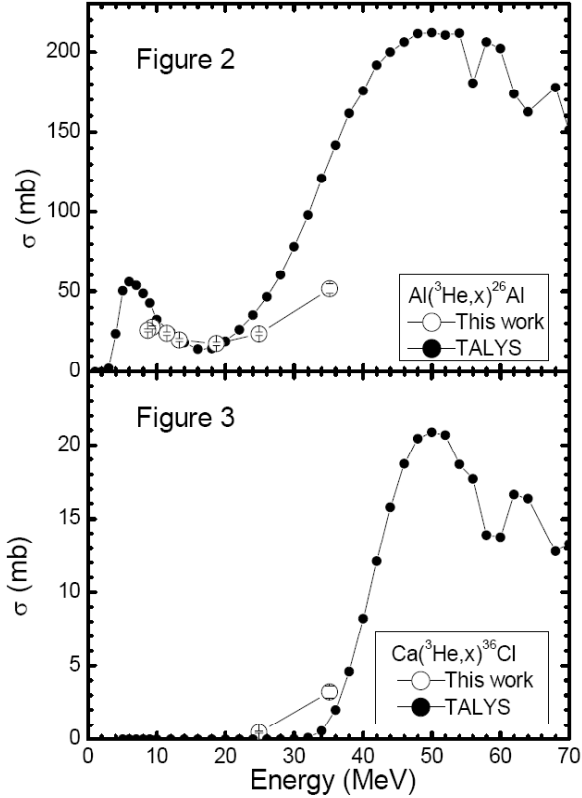
Chemical separation of ²⁶Al and ³⁶Cl: The weighed Mg and Ni foils were dissolved in 1 mL of a carrier solution containing 10 mgf Al in 5% HNO₃. Ni was separated by precipitation of Ni(OH)₂ with 2.5 M NaOH. After evaporation of the supernatant solution and re-dissolution in 1 M HCl, Al was precipitated at pH ~8 in NH₃-NH₄Cl buffer. The resulting precipitate of Al₂(OH)₃ was heated at 950°C to form the oxide. The aluminum facing foils of targets Ca1 and Ca2 were dissolved in 2.5 M NaOH after the addition of Cl⁻ and Ca²⁺ carrier. All but the insoluble fraction of the limestone then was dissolved in a few mL of 3 M HNO₃. Chloride precipitated as AgCl on the addition of Ag⁺(aq). The AgCl was purified as in [10]. After removal with HCl of excess Ag⁺ from the supernatant solution, the new supernatant solution contained some iron from the aluminum foil. We removed the iron by extraction into isopropyl ether from 8 M HCl. Remaining aluminum was converted to Al₂O₃ as described above. The supernatant contained calcium, which was reserved for the measurement of ⁴¹Ca.

Accelerator mass spectrometry: We measured the ²⁶Al/²⁷Al and ³⁶Cl/Cl ratios (10⁻¹²) of the samples at Purdue University. For blanks we found ²⁶Al/²⁷Al = 0.02 and ³⁶Cl/Cl = 0.06. In targets Ca1 to Ca8, the ²⁶Al/²⁷Al ratios ranged from 30 to 110 and in targets Mg1 to Mg8 from 3 to 20. Before blank corrections, the measured ³⁶Cl/Cl ratios in targets Ca4 and Ca5 were 2.78 and 0.30, respectively.



Results: Cross sections were calculated from the standard relation $\sigma = N_{product}[atoms] / (N_{target}[atom/cm^2] \times N_{helium-3})$. Preliminary results are shown in Figures 1-3. Uncertainties in σ are estimated to be 10-15%. The main ones are in the fluence, 5-10% and the target thicknesses, ~2% for Mg and ~10% for Ca.

On average, our cross sections for $\text{Mg}({}^3\text{He},x){}^{26}\text{Al}$ reactions are 81% of the values predicted by the TALYS code (Figure 1). We can also compare our results to those of [6] who irradiated ${}^{24}\text{Mg}$ foils. In the energy range above 10 MeV, from the TALYS calculations, we would have expected our cross sections to be larger. In fact we find smaller cross sections from about 6.5 to 13.6 MeV. Such a difference might arise if TALYS [11] overestimates the cross sections for the reactions with ${}^{25}\text{Mg}$ and ${}^{26}\text{Mg}$.



We have no experimental cross sections for comparison with our results for either $\text{Al}({}^3\text{He},x){}^{26}\text{Al}$ (Figure 2) or $\text{Ca}({}^3\text{He},x){}^{36}\text{Cl}$ (Figure 3). The agreement with the results of the TALYS modeling is good. Our $\text{Al}({}^3\text{He},x){}^{26}\text{Al}$ cross section at the lowest energy (not shown) has a large uncertainty because of the fluence measurement. Excluding this point, the average ratio of $\sigma_{\text{experimental}}/\sigma_{\text{TALYS}}$

is 0.77 ± 0.04 . Our two cross sections for ${}^{36}\text{Cl}$ appear consistent with the expected reaction threshold near 30 MeV. The cross section of ~ 3 mb at 35 compares favorably with the TALYS result of about 2 mb at 36 MeV.

Implications for an early irradiation: Models of early solar system irradiation have relied on cross section predicted by nuclear codes of uncertain accuracy. Our new data indicate that one such code, TALYS, predicts cross sections for ${}^3\text{He}$ -induced reactions fairly well - mostly within a factor of 2 - and better than competing nuclear models tried to date. With 1) our newly measured cross sections, 2) increased confidence in TALYS cross sections, and 3) some previously overlooked data [12], we updated the model calculations of [7]. For spectral parameters as in [7], the calculated, initial ${}^{41}\text{Ca}/{}^{26}\text{Al}$ ratio varies between 78 and about 1000, values far higher than any determined for an early solar system condensate. The calculated range for ${}^{10}\text{Be}/{}^{26}\text{Al}$ is 30 for gradual events to 800 for impulsive events. Conceivably, 0.5-1.0 My could have passed without further irradiation before CAIs formed. Radioactive decay then would have brought the calculated ratios into agreement with canonical early solar system values. However such a long time gap seems unreasonable because it is hard to imagine that the temperatures stayed so high for so long outside the region of irradiation. We conclude that a one-stage, uniform irradiation of early solar system matter by protons and alpha particles, either with or without ${}^3\text{He}$, cannot explain meteoritic observations. If other results (${}^7\text{Be}$; [1,2]) require local irradiation, appropriate scenarios will have to be constructed ad hoc.

References

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