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BEAM INTERACTIONS WITH MATERIALS AND ATOMS
Nuclear Instruments and Methods in Physics Research – Section B

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Measurements of proton-induced production cross sections for ^{36}Cl from Ca and K

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Abstract

Production cross sections for ^{36}Cl (half-life $= 3.01 \times 10^5$ y) have been measured for the nat.K(p, x), ^{39}K(p, x), nat.Ca(p, x) and ^{40}Ca(p, x) reactions up to 40 MeV. The results of nat.Ca(p, x) reaction are generally consistent with measurements performed at somewhat higher energies. With the completion of these measurements it is now possible to proceed with model calculations of the solar cosmic ray (SCR) flux over the last 400 ky based on measurements of lunar surface materials.

1. Introduction

Radionuclides produced in extraterrestrial surface materials by energetic solar flare particles provide clues to past solar activities, both particle intensities and energy spectra averaged over the radionuclide mean lives [1]. For example, recent ^{36}Cl (half-life $= 3.01 \times 10^5$ y) measurements by Nishiizumi et al. (1991, 1995) from lunar surface rock 74275 and 64455 indicate excess ^{36}Cl concentrations in the top several g/cm^2 [2,3]. This excess originates from solar cosmic rays (SCR) production of ^{36}Cl in the upper few cm of this particular rock. Ultimately, these lunar surface materials radionuclide measurements will yield the average SCR flux and spectrum over the past 400 ky. However, to derive the SCR parameters from a ^{36}Cl depth profile it is necessary to separate that ^{36}Cl produced by galactic cosmic rays (GCR) from that produced by SCR. In the top several cm of lunar surface material the dominant mechanism of ^{36}Cl production is via protons having several tens of MeV energy. In this regime SCR production dominates GCR production. Clearly an essential component in interpreting these data is precise production cross sections for charged particle reactions producing ^{36}Cl. As shielding increases secondary neutron reactions produced by the higher energy GCR dominate ^{36}Cl production. We report here experimental results for ^{36}Cl production cross sections from the reactions nat.K(p, x), ^{39}K(p, x), nat.Ca(p, x) and ^{40}Ca(p, x).

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2. Experimental

For the proton bombardments four targets were prepared: reagent grade KNO$_3$ and CaCO$_3$ of natural isotopic compositions and isotopically enriched ^{39}KNO$_3$ and ^{40}CaCO$_3$. The isotopic abundances of enriched ^{39}K and ^{40}Ca were 99.97% and 99.98%, respectively. The enriched isotopes were purchased from Oakridge National Laboratory. The CaCO$_3$ and KNO$_3$ targets were made into 15 mm diameter pellets of 30–50 mg/cm^2 thickness. The proton beam diameter (5 mm) is considerably smaller than the target diameter so the beam is always focused on the target. Each target pellet was placed between Al foils, which serve to prevent break-up of the target and as energy absorbers. Several alternating pellets and foils were placed as stacks inside an Al target holder. The stacked targets were bombarded by 35 and 40 MeV protons for about 10 min. The average beam current was 0.1 μA. This irradiation was performed at the AVF cyclotron at the Institute for Nuclear Study, University of Tokyo. The incident energy for each target pellet was calculated by the proton energy loss in passing through other targets and absorbers.

The proton beam flux was determined by ^{22}Na produced via the reaction ^{27}Al(p, α pn)^{22}Na in an Al monitor foil (200 μm in thickness) set in the middle of the stacked targets. For this flux determination the cross sections reported by Steyn et al. (1990) [4] were utilized. The ^{22}Na-derived γ-rays in the Al monitors were detected by a Ge detector measuring 511 keV annihilation γ-rays in coincidence with 1275 keV γ-rays. As an additional check the proton beam current was also measured by a Faraday cup.

The proton fluxes based on the Faraday-cup measurements agreed well with those based on the Al monitors (3% for 35 MeV bombardments and 5% for 40 MeV).

After the non-destructive γ-ray counting (for ^{7}Be measurements), the KNO_3 target was dissolved in water and subsequently acidified with dilute HNO_3 along with ≈ 0.4 mg of Be (for ^{10}Be measurements) and ≈ 3 mg of Cl carrier. About 2 mg of Ca carrier was added to the proton irradiated KNO_3 targets for future ^{41}Ca measurements. The $CaCO_3$ targets were put in suspension in H_2O and subsequently dissolved with dilute HNO_3 along with the Be and Cl carriers. The Cl was precipitated as AgCl. In order to assess the recoil effects on the Al target holder which supported the KNO_3 and $CaCO_3$ targets, the surfaces of four Al holders (CS-1, 2, 7, and 8) were etched and the ^{36}Cl in the etchants were measured. Chlorine was also extracted from all four unirradiated target material types and used as process blanks. The ^{36}Cl concentrations were measured using AMS techniques at the Lawrence Livermore National Laboratory [5].

3. Results and discussion

The results are shown in Table 1. The measured ^{36}Cl/Cl ratios were normalized to NBS ^{36}Cl standards (diluted by K. Nishiizumi). The errors reflect the quadratic sum of the

$\pm 1\sigma$ AMS measurement errors and the integrated proton beam currents, and do not include the errors in the uncertainty of the absolute activities of the standards which are less than 1%. The uncertainties in proton intensities are mostly the result of uncertainties in the ^{27}Al$(n,\alpha pn)^{22}$Na cross sections by Steyn et al. (1990) [4] and include statistical errors (1–2%) in ^{22}Na γ-counting.

The CS-7 target was irradiated with 15.2 MeV protons, an energy below the production threshold for ^{36}Cl production from KNO_3. As expected, the measured ^{36}Cl/Cl ratios in the Al holder of CS-7 were identical to those of the Cl blanks, 7×10^{-15}. The absence of ^{36}Cl in the CS-7 target holder indicates the absence of contaminant ^{36}Cl throughout the chemical procedure. This is not always the case since ^{36}Cl was detected in the Al target holders for CS-1, 2, and 8. This most likely indicates that a small fraction of the ^{36}Cl was recoiled into the Al target holders. Fortunately, the recoiled fraction amounted to less than 0.5% and was neglected in the calculation of cross sections.

Figs. 1 and 2 show the proton-induced production cross sections for the Ca and K targets, respectively. The cross sections are presented as elemental cross sections for natural targets. Fig. 1 also displays the data recently reported by Schiekel et al. (1996) who measured ^{36}Cl production cross sections from natural Ca above 45 MeV [6]. Although there are no overlapping data between the

Table 1
Results on ^{36}Cl AMS measurements for the proton-irradiated targets

ID	Thickness (mg/cm^2)	Energy (MeV)	Incident proton (10^{14})	Cl/Cl (10^{-13})	Cross section (mb)
nat.KNO$_3$					
CS-1	49.3	39.7	4.21	248.3 ± 6.5	10.80 ± 0.78
CS-2	45.9	38.3	4.21	194.7 ± 3.7	8.89 ± 0.62
CS-3	27.3	35.3	4.04	64.4 ± 1.6	4.96 ± 0.42
CS-4	42.5	30.2	4.04	43.7 ± 1.5	2.12 ± 0.21
CS-5	42.4	25.3	4.04	16.8 ± 0.5	0.73 ± 0.14
CS-6	44.9	23.2	3.88	7.24 ± 0.16	0.15 ± 0.21
CS-7	46.9	15.2	3.88	4.24 ± 0.11	< 0.20
^{39}KNO$_3$					
CS-8	41.2	39.7	4.02	135.5 ± 2.8	7.17 ± 0.50
CS-9	42.2	38.4	4.02	88.4 ± 2.1	4.55 ± 0.33
CS-10	38.6	35.2	3.78	30.6 ± 0.5	1.83 ± 0.13
nat.CaCO$_3$					
CS-11	44.3	39.7	4.18	6.37 ± 0.17	0.296 ± 0.021
CS-12	42.6	38.4	4.18	5.19 ± 0.18	0.249 ± 0.019
CS-13	48.1	35.1	3.92	3.06 ± 0.18	0.138 ± 0.013
CS-14	48.0	29.8	3.92	0.89 ± 0.07	0.034 ± 0.006
CS-15	55.0	24.7	3.92	0.79 ± 0.14	0.023 ± 0.008
CS-16	48.1	21.9	3.95	0.63 ± 0.26	0.018 ± 0.014
CS-17	59.3	13.4	3.95	0.32 ± 0.07	< 0.014
^{40}CaCO$_3$					
CS-18	41.6	39.7	4.14	0.99 ± 0.18	0.046 ± 0.010
CS-19	44.3	38.4	4.14	0.59 ± 0.24	0.023 ± 0.012
CS-20	42.7	35.2	4.01	0.25 ± 0.06	0.008 ± 0.004

IX. PRODUCTION RATES/CROSS SECTIONS

two experiments, the agreement is good with the exception of the measurement at 45 MeV by Schiekel et al. (1996). A possible explanation for this seemingly anomalous measurement is the additional production of ^{36}Cl caused by secondary neutron reactions on a Ca target located at the end of stacked targets. Knock-on collision processes involving high-energy protons produce sufficient high energy neutrons to account for the excess ^{36}Cl via ^{40}Ca(n, x)^{36}Cl reactions. This particular neutron-induced reaction has a relatively large cross section reaching a peak of 80 mb at around 25 MeV according to our recent measurements. A similar effect may account for our results for natural KNO$_3$. The CS-7 sample bombarded well below the reaction threshold for ^{36}Cl nevertheless contained a small but detectable amount of ^{36}Cl. The apparent cross section was 0.2 mb. Secondary neutrons are most likely responsible for this excess via the exothermic nuclear reaction of ^{39}K(n, α)^{36}Cl, the cross section of which has a maximum of 160 mb at 6–8 MeV [7]. In this particular reaction the neutrons are most likely derived from an evaporation process occurring in the stacked target assembly and are more or less isotropic for low energy incident protons. The excess cross section observed in CS-7 is explained by neutron (> 3 MeV, forward) to proton (incident) ratio of ≈ 0.0016. In the neutron production experiments reported for 30 MeV protons on thick targets by Nakamura et al. (1983), the n/p ratio was 0.0005 for C, 0.0021 for Fe and 0.0027 for Cu [8]. Assuming that secondary neutrons are isotropic and produced in proportion to the amount of target, a correction in
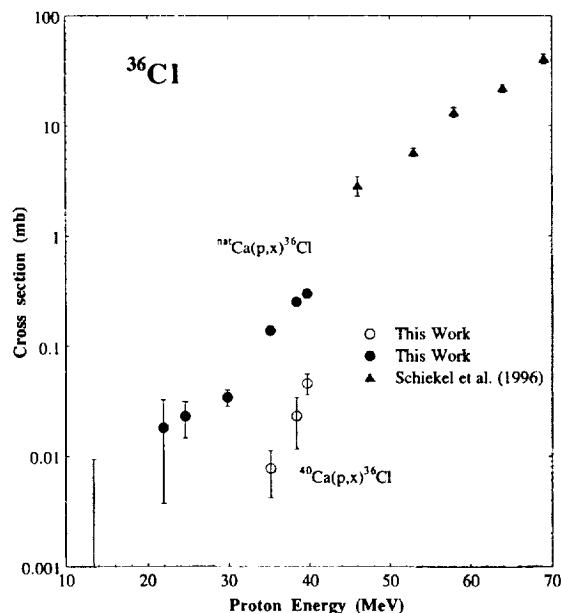

Fig. 2. Cross section in mb for ^{36}Cl production from natural K and ^{39}K as a function of proton energy.

all KNO$_3$ samples for this contribution was made with an attached error of 100%.

We made a similar correction for Ca(p, x) reactions. CS-17 has a small cross section of ca. 0.01 mb. One possible explanation is a contribution from the ^{43}Ca(p,2 α) reaction, however reactions emitting two alpha particles are highly suppressed by Coulomb barriers for low excitation energies. Alternatively, the ratio of secondary neutrons to incident protons for higher energy group neutrons (> 15 MeV) is an order of magnitude lower than for low energy evaporation neutrons (> 3 MeV) [8] and could yield an apparent cross section of the same order as the observed value for CS-17. Although there is no conclusive evidence it is nevertheless assumed that neutrons of > 15 MeV were emitted in the forward direction and a correction (100% uncertainty) was applied to all samples. These corrections were small and negligible for most of the Ca samples.

The excitation function of the nat.Ca(p, x)^{36}Cl reaction has a rather complex structure. Since natural calcium is a composite of several isotopes, it includes reactions from several target nuclei. The relevant reactions include the ^{40}Ca(p,^{3}He2p), ^{40}Ca(p,4pn), ^{42}Ca(p,α2pn), ^{43}Ca(p,2 α), ^{44}Ca(p,2 αn) reactions. Their Q-values are −27.5, −35.3, −26.8, −6.4 and −17.6 MeV, respectively. For the nat.K(p, x)^{36}Cl reaction, it is clear from Fig. 2 that the ^{41}K(p,αpn) reaction is the dominant means of ^{36}Cl production below 35 MeV. The Q-value for this reaction is −16.5 MeV and is much larger than that of the ^{39}K(p,3pn) reaction, −26.9 MeV.

Fig. 1. Cross section in mb for ^{36}Cl production from natural Ca and ^{40}Ca as a function of proton energy. The data by Schiekel et al. (1996) [6] for natural Ca above 46 MeV are also plotted.
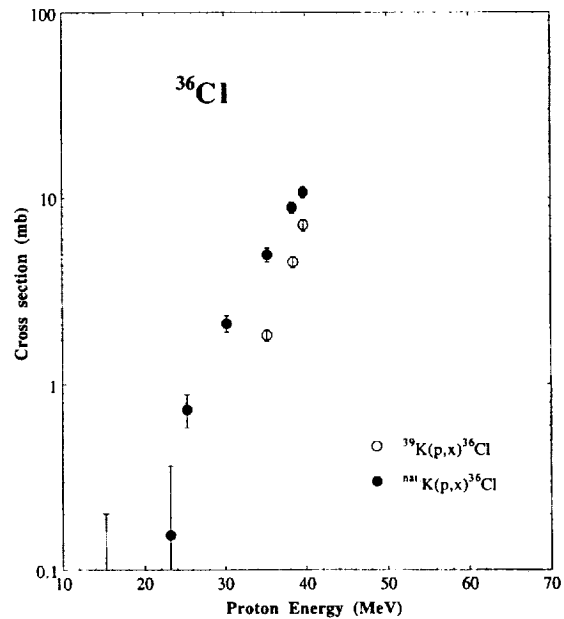
4. Conclusion

We have measured ^{36}Cl production cross sections for the nat.K(p, x) and nat.Ca(p, x) reactions as well as the ^{39}K(p, x) and ^{40}Ca(p, x) reactions below 40 MeV. With the incorporation of this low-energy data into the excitation functions for these reactions we can now proceed with the refinement of the model calculation of the SCR intensity during the last 400 ky based on measurements from lunar surface samples. In the future we plan to measure the cross sections in the medium proton energy region where some discrepancies are suspected when one compares our low energy data and the high energy data by Schiekel et al [6]. The neutron-induced cross section measurements on ^{36}Cl production are in progress and will appear elsewhere.

Acknowledgements

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