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Depth profile of ^{41}Ca in an Apollo 15 drill core and the low-energy neutron flux in the Moon

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Abstract

Systematic measurements of the concentrations of cosmogenic ^{41}Ca (half-life = 1.04×10^5 yr) in the Apollo 15 long core 15001–15006 were performed by accelerator mass spectroscopy. Earlier measurements of cosmogenic ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, and ^{53}Mn in the same core have provided confirmation and improvement of theoretical models for predicting production profiles of nuclides by cosmic ray induced spallation in the Moon and large meteorites. Unlike these nuclides, ^{41}Ca in the lunar surface is produced mainly by thermal neutron capture reactions on ^{40}Ca. The maximum production of ^{41}Ca, about 1 dpm/g Ca, was observed at a depth in the Moon of about 150 g/cm^2. For depths below about 300 g/cm^2, ^{41}Ca production falls off exponentially with an e-folding length of 175 g/cm^2. Neutron production in the Moon was modeled with the Los Alamos High Energy Transport Code System, and yields of nuclei produced by low-energy thermal and epithermal neutrons were calculated with the Monte Carlo N-Particle code. The new theoretical calculations using these codes are in good agreement with our measured ^{41}Ca concentrations as well as with ^{60}Co and direct neutron fluence measurements in the Moon.

Keywords: cosmogenic nuclide; thermal neutron; production rate; lunar soil

1. Introduction

Reedy and Arnold developed a simple semi-empirical model for calculating the production rates of various cosmogenic nuclides made by high-energy spallation reactions as a function of depth in the Moon [1]. The model was later expanded and also used for studies of exposure histories of meteorites and other bodies [2]. Neutron-capture reactions in the Moon were modeled with the ANISN computer code by Lingenfelter et al. and Spergel et al. [3,4]. A new model is now being used for the calculation of cosmogenic-nuclide production rates in the Moon and meteorites by both spallation and neutron-capture reactions that is based on the Los Alamos High Energy Transport (LAHET) Code System [5,6].

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Unlike other cosmogenic nuclides, the majority of ^{41}Ca (half-life = 1.04 × 10^5 yr) in the lunar surface is produced by the capture of low-energy, thermal (~0.02 eV) and epithermal or resonance (~0.1–1000 eV), neutrons via the ^{40}Ca(n,γ)^{41}Ca reaction instead of high energy spallation reactions. The thermal neutron cross section for this reaction is 0.43 barns [7] and calcium captures ~10% of thermal neutrons in lunar material of average composition [8]. The measurement of ^{41}Ca in lunar cores allows us to explore the production versus depth relation for a cosmogenic radionuclide that is produced almost entirely by the capture of thermal neutrons. The only other information concerning reactions of thermal neutrons in well documented lunar samples come from the Apollo 17 Lunar Neutron Probe Experiment (LNPE) for the ^{235}U fission and ^{10}B(n,α)^{7}Li reactions [9] and ^{59}Co(n,γ)^{60}Co measurements [10]. The thermal neutron capture production rates or profiles in some meteorites were also measured for ^{60}Co ([11,12,14], Y. Nakamura, pers. commun., 1978) ^{59}Ni [15], ^{41}Ca [12,16–18], and ^{36}Cl ([12,19,20], Y. Nakamura, pers. commun., 1978). However, the determination of absolute depth scales in meteorite studies is much more complex and uncertain than that for lunar cores. A depth profile of ^{41}Ca was measured in the surface layers (top 16 g/cm^2) of lunar rock 74275 [21] to study the flux of solar protons from the spallation production of ^{41}Ca from titanium.

The depth profile of neutron-capture-produced ^{41}Ca mimics the acute sensitivity of the fluxes of low-energy (thermal and epithermal) neutrons to depth, composition, and effective radius of the irradiated body (e.g. [4]). Knowing the ^{41}Ca production rate versus depth profile will allow the use of ^{41}Ca in determining sample locations and meteoroid radii. A detailed knowledge of the depth profile for neutron-capture reactions is also very important for the determination of elemental composition of planetary surfaces using gamma rays made by neutron-capture reactions [8,22].

The nuclear processes involved in the interaction of high-energy galactic cosmic ray (GCR) particles, which penetrate deep inside the irradiated body and produce a large number of secondary particles, were numerically simulated in detail by the LAHET Code System (LCS) [23], which is a system of 3D Monte Carlo particle production and transport codes. The transport of high-energy particles is done with the LAHET code, and neutrons with energies below a cut-off energy of 15 MeV are further transported to thermal energies (~0.02 eV) by the Monte Carlo N-Particle (MCNP) code [24]. LCS, together with its adaptations to planetary applications, are described in [25]. LCS has been used to calculate cosmogenic nuclide production rates both in meteorites and lunar samples and gave results that are almost always in good agreement with experimental data [5,26]. This model has also been used to calculate successfully the production rates of cosmogenic nuclides in the Earth's atmosphere and on the terrestrial surface [27].

Deep lunar drill cores are the best samples available for the study of the GCR production profile. Earlier measurements of ^{10}Be (half-life = 1.5 × 10^6 yr), ^{14}C (5730 yr), ^{26}Al (7.05 × 10^5 yr), ^{36}Cl (3.01 × 10^5 yr), and ^{53}Mn (3.7 × 10^6 yr) in the same Apollo 15 deep core [28–31] have provided confirmation and improvement of theoretical models [1,2] for production of these and other nuclides by cosmic ray-induced spallation in the Moon and in large meteorites. Lunar samples have been exposed at a known location in the solar system (1 A.U.) and have been exposed at accurately known shielding depths and geometries. Among the three Apollo deep drill cores, 15001–15006, 60001–60007, and 70001–70009, the Apollo 15 core is the best for cosmogenic nuclide studies because it is the least disturbed core. The other two cores are less suitable. The Apollo 16 long core had poor extraction during coring recovery of the core was incomplete [32] and the Apollo 17 long core had a recent large cratering event followed by a filling of surface material [33].

We report here a series of systematic measurements of ^{41}Ca concentrations in the Apollo 15 drill core. Theoretical calculations with LCS, especially MCNP, of depth profiles of low-energy neutrons and the rates for nuclear reactions, such as neutron capture, that they induce are presented and compared with available data sets.

2. Experiment and results

Nine soil samples were selected from the Apollo 15 drill core 15001–15006. The sample depth covered was from the surface to 218 cm (388 g/cm^2). Each soil was dissolved with HF/HNO_3 mixture and used for the previous ^{10}Be, ^{26}Al, and ^{36}Cl measurements [29,30]. After Be, Al, and Cl extraction, Ca was separated from each of the remaining solutions by anion and cation exchange column from the solution. After the Ca concentration was measured in the separated fraction, Ca carrier was added in order to provide a sufficient amount of Ca for preparation of the accelerator mass spectroscopy (AMS) target. Ca oxalate was precipitated from the solution by 4% ammonium oxalate. The Ca oxalate was ignited in a small quartz vial to form CaO. The CaO was converted to CaH_2 for use in the AMS ion source. The ^{41}Ca measurements were performed at the University of Pennsylvania AMS facility using established procedures [34]. The ^{41}Ca standard was prepared from the neutron irradiation of commercial CaH_2 and had a ^{41}Ca/^{40}Ca value of $(5.41 \pm 0.50) \times 10^{-12}$ calculated from neutron flux and thermal neutron cross section. The standard was typically measured by AMS to within 95% of the nominal ratio.

The measured ^{41}Ca/^{40}Ca ratios ranged from 1.5×10^{-13} to 1.9×10^{-12}. The chemical blank, prepared in an identical fashion starting from the Ca carrier solution, gave a ^{41}Ca/^{40}Ca ratio of 5×10^{-15}. The measured ratios were normalized to the ^{41}Ca standard. The results are shown in Table 1 along with the sample's depth and the Ca and Fe chemical composition [29]. Each quoted error represents the quadrature of the 1σ AMS measurement error (2–6%) and errors in the Ca atomic absorption spectrometry measurements (3%), but excludes the uncertainty of the absolute activity of the standard (9.2%).

The weights of samples shown in the table are different from the previous reports on ^{10}Be, ^{26}Al, and ^{36}Cl measurements because the Ca was extracted from a fraction of the remaining solution. Each weight was calculated to be equivalent to that of the original sample times the Ca fraction used here. The average chemical composition of the core sample is 0.238% K, 7.39% Ca, 1% Ti, and 12.0% Fe. The deepest sample, 15001,361, shows a quite different chemical composition from the average, containing relatively low K (0.154%) and Ca (6.17%) but high Fe (14.7%).

3. Discussion

Fig. 1 shows the measured depth profile of ^{41}Ca in the Apollo 15 drill core. Two facts are immediately apparent. First, the peak production occurs at a depth of ~ 150 g/cm^2, which corresponds to the peak low-energy neutron flux as observed at the Apollo 17 site by the LNPE [9]. This is much deeper than the peak production (~ 40 g/cm^2) of spallation nuclides like ^{26}Al [1,35,30] or ^{53}Mn [28] by galactic cosmic ray particles. Second, the absolute production at the peak, close to 80 dpm/kg, is far higher than expected for spallation reactions in samples of this composition [1]. The measured ^{41}Ca concentrations in small iron meteorite falls [36] and in the metal

Table 1
^{41}Ca results in the Apollo 15 deep drill core

Sample	Depth (cm)	Depth (g/cm^2)	Weight (mg)	Ca [a] (%)	Fe [a] (%)	^{41}Ca (dpm/kg)	^{41}Ca [b] (dpm/g Ca)
15006,267	2.6	4.2	33.3	7.30	11.8	22.9 ± 1.0	0.289 ± 0.014
15006,266	25.6	41.9	42.8	6.92	12.2	40.1 ± 1.5	0.557 ± 0.022
15005,463	41.3	68.1	40.5	7.17	11.7	61.4 ± 3.7	0.840 ± 0.052
15005,462	64.3	110.1	45.7	7.13	11.5	72.0 ± 3.8	0.998 ± 0.054
15004,214	93.0	162.2	71.6	7.52	12.3	79.7 ± 2.7	1.052 ± 0.035
15003,673	125.3	220.4	119.2	8.23	12.0	72.8 ± 2.7	0.880 ± 0.033
15002,582	165.0	290.8	197.4	7.23	12.1	50.6 ± 1.8	0.697 ± 0.025
15002,581	191.7	336.3	223.8	7.61	12.0	41.6 ± 2.3	0.545 ± 0.030
15001,361	217.7	387.8	296.7	6.17	14.7	25.0 ± 1.0	0.403 ± 0.017

[a] From [29].
[b] Low-energy neutron production by the ^{40}Ca(n,γ)^{41}Ca reaction.

phase of larger chondrites [16,37] indicate that ^{41}Ca production from spallation alone ranges from 24 ± 1 to 21 ± 2 dpm/kg Fe, respectively. Extrapolation to the lunar surface would reduce this amplitude by about a factor of 2, which would then correspond to a contribution of ~ 1.4 dpm/kg due to iron in the Apollo 15 core (12.0% Fe). The spallogenic ^{41}Ca in the core is calculated to decrease from about 1.8 dpm/kg at the surface to 0.1 dpm/kg using the Reedy–Arnold model [1]. Thus our measured peak activity of ^{41}Ca is ~ 50 times larger than expected by spallation reactions alone.

The low-energy neutron-capture production of ^{41}Ca is calculated by subtracting the spallation contribution from the measured activity. The low-energy neutron-capture ^{41}Ca concentration normalized to the Ca content, dpm ^{41}Ca/g Ca, of each sample is shown in the last column of Table 1 and in Fig. 2. The deepest sample, 15001,361, shows values for both ^{36}Cl [29] and ^{41}Ca that fall below a semi-log straight line when expressed as dpm/kg sample (Fig. 1). Using a unit of specific activity, dpm/g Ca, the value moves up to the straight semi-log relationship. The significantly lower Ca content of this sample than the mean value explains this result. The low ^{36}Cl in this sample was also explained by a lower Ca content [29].

The ^{60}Co (half-life = 5.27 yr) activities measured by Wahlen et al. [10] in the Apollo 15 long core (Fig. 4) are also consistent with our ^{41}Ca data, although complications due to strong ^{59}Co neutron-capture resonances at epithermal energies and variations in cosmic-ray fluxes over the 11 year solar cycle make this comparison less direct. The ^{41}Ca data show a peak position and curve shape in good agreement with the ^{235}U fission rate curve (Fig. 5) of Woolum and Burnett [9].

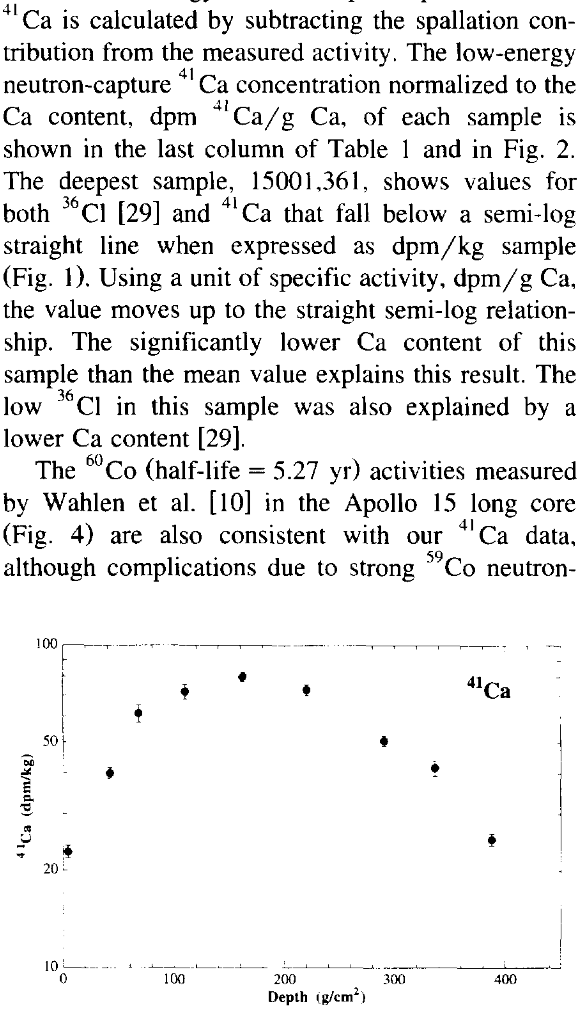

Fig. 1. Measured ^{41}Ca activities in the Apollo 15 drill core 15001–15006. The error bars include $\pm 1\sigma$ AMS measurements and chemical analysis of Ca and exclude error of absolute activity of the ^{41}Ca standard (±9.2%).

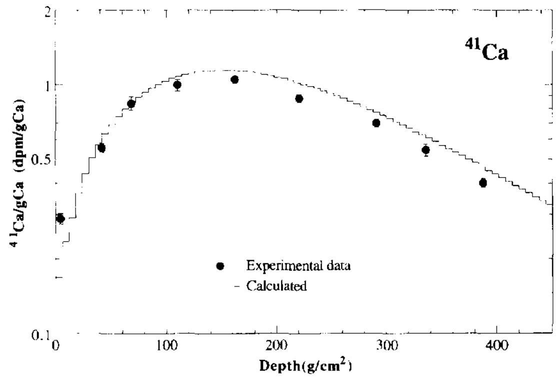

Fig. 2. Low-energy neutron produced ^{41}Ca activities in the Apollo 15 drill core. The experimental data have been normalized to Ca concentrations after subtraction of small amounts of spallation contribution (see text). The solid line is the calculated production rate using the LAHET Code System, assuming ^{41}Ca from ^{40}Ca (n,γ) reaction.

Except possibly for the near-surface (4.2 g/cm^2) sample, we believe that the ^{41}Ca results presented here are unaffected by 'gardening' of the lunar regolith on this time scale [38]. Although it is known that the top ~ 3 g/cm^2 or more were lost during or subsequent to sampling, based on solar cosmic ray (SCR)-produced ^{22}Na profiles [39], the near-surface sample, 15006,267, depth 4.2 g/cm^2, also contains SCR-produced ^{26}Al [30]. ^{41}Ca is also produced via ^{41}K(p,n), ^{42}Ca(p,pn), and Ti(p,x) reactions by SCR particles. In a study of lunar basalt 74275 (7.6% Ti content), the SCR production of ^{41}Ca was shown [21] to be no larger than 10 dpm/kg on the surface and is exhausted at depths greater than 5–8 g/cm^2. Moreover, the SCR-produced ^{41}Ca in sample 15006,267 is low because of the low isotopic abundance of ^{41}K (6.73%) and ^{42}Ca (0.647%) and the low elemental abundance of K (0.225%) and Ti (1%). Some of the ^{41}Ca observed in 15006,267 was produced by high-energy GCR particles. As indicated above, the ^{41}Ca production rate by spallation is calculated to be about 1.8 atom/kg · min at the depth of 4 g/cm^2, much lower than the observed activity of 22.9 ± 1.0 dpm/kg. Hence, we conclude that the majority of ^{41}Ca in near surface samples is produced by low-energy neutron capture. It is, however, possible that the near-surface sample 15006,267 is somewhat affected by mixing with deeper material, resulting in an enhanced ^{41}Ca content.

The e-folding attenuation length for the production of ^{41}Ca from thermal neutron capture is 175 g/cm^2 for depths larger than about 300 g/cm^2. A similar result is observed in the Apollo 17 LNPE data for the fission of ^{235}U (shown in Fig. 5). In both cases, the LCS-calculated lengths, discussed below for pure exponential attenuation for depths beyond 300 g/cm^2, is also 175 g/cm^2. This e-folding length, which characterizes the reduction in ^{41}Ca production from low-energy neutron reactions and, consequently, the attenuation in the lunar thermal neutron flux, is similar to the e-folding lengths of 173–177 g/cm^2 measured in the Apollo 15 core for spallogenic production of ^{10}Be, ^{26}Al, and ^{53}Mn [28–30] but is slightly shorter than the e-folding length of 190 g/cm^2 observed for ^{36}Cl [29].

3.1. Theoretical calculations

We compared our and similar measurements for products of neutron-capture reactions with two different sets of theoretical calculations, those of Lingenfelter et al. [3] (often used for such reactions) and the ones that we did using LCS. LCS uses a different sets of computer codes that have been developed since the work of [3] and has worked well with cosmogenic nuclides made by spallation reactions.

Lingenfelter et al. [3] modeled the lunar neutron fluxes, spectra, and capture rates as a function of the macroscopic cross section. An effective total macroscopic capture cross section, Σ_{eff}, for the Apollo 15 drill core is calculated to be 0.0080 cm^2/g using the average chemical composition of the core, including 14 ppm Sm and 19 ppm Gd. Lingenfelter et al. [3] calculated the peak (~ 150 g/cm^2) neutron capture rate for a 100 barn 1/v cross section as a function of the effective total macroscopic cross section (their fig. 6). Using the value of 0.0080 for Σ_{eff} in Apollo 15 core and a 1/v thermal cross section for Ca of 0.43 barn [7], we calculate from [3] the peak steady-state decay rate (or production rate) of ^{41}Ca to be 0.88 dpm/g Ca, which is ~ 15% lower than the observed value.

For the LCS calculations, the target body was simulated as a sphere with the radius of the Moon, 1738 km. Irradiation of the lunar surface was simulated with an isotropic GCR omnidirectional particle flux of 4.56 nucleons/s · cm^2, with an energy distribution corresponding to the GCR primary particle flux averaged over a solar cycle [26]. The only normalization made to the LCS calculations was the incident flux, which was obtained by fitting cosmogenic radionuclides in the Apollo 15 deep drill core [26]. As the particle fluxes are strongly depth dependent, the outer (surface) part of the sphere was divided into concentric shells with a thickness of 6 g/cm^2. In each shell proton and neutron fluxes were calculated. Statistical errors of the calculated fluxes, running 100,000 primary GCR particles, were less than 3%.

We started our calculations with the study of depth and energy dependencies of neutron yields. The total neutron yield calculated by LAHET is (29.3 ± 0.15) neutrons per primary GCR nucleon, and (15 ± 0.1) of them are produced or slowed below the 15 MeV energy cut-off for neutron transport by LAHET. Similar neutron yields were reported by [40]. The depth distributions of neutrons produced with energies above and below 15 MeV and their sum are presented in Fig. 3. The depth distribution of the source neutrons with energies below 15 MeV exhibits a shallow maximum at a depth of ~ 40 g/cm^2 and decreases exponentially at greater depths, with an e-folding length of 165 g/cm^2. This neutron source profile is similar to those used in earlier calculations [3,4]. However, this source e-folding length is not that for the lunar thermal-neutron flux because source neutrons (~ 0.1–15 MeV) undergo much scattering in the volatile-poor Moon before they are thermalized (~ 0.02 eV).

Running MCNP with the above source neutron spatial and energy distribution, differential fluxes of thermal and epithermal neutrons were calculated. Having calculated these final (equilibrium) neutron fluxes, the rates of the reactions investigated were calculated by integrating over energy the product of these fluxes with corresponding cross sections taken from the ENDF/B-VI library [41], which is coupled with MCNP. Several calculated profiles are presented in Figs. 2, 4 and 5. The good agreements

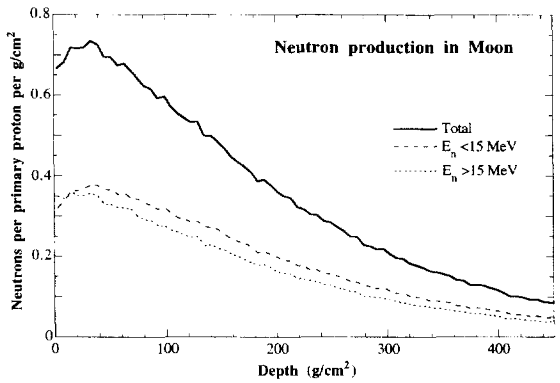

Fig. 3. Neutron production by GCR particles vs. depth in the Moon. The depth distributions of neutrons produced with energies above and below 15 MeV and their sum are presented.

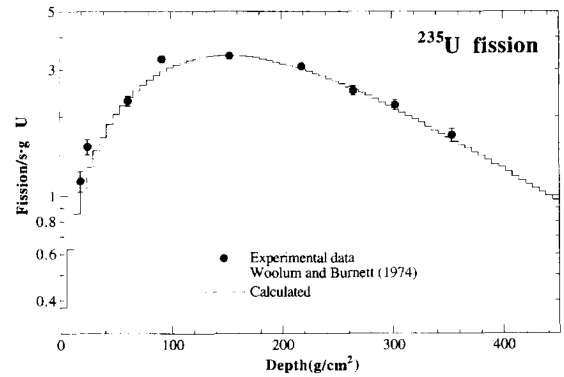

Fig. 5. Calculated fission rate for ^{235}U (solid line) vs. depth compared with ^{235}U fission rates measured in the Lunar Neutron Probe Experiment [9].

between the calculated depth profiles and the measured data $^{40}Ca(n,\gamma)^{41}Ca$ and $^{59}Co(n,\gamma)^{60}Co$ [10] are presented in Figs. 2 and 4. We do not know why the deeper measured ^{41}Ca points are to the left of the calculated curve in Fig. 2 while the ^{235}U fission measurements and calculations in Fig. 5 show no such displacement. As noted above, LCS reproduces well the observed attenuation lengths of these two reactions. The capture rate for $^{35}Cl(n,\gamma)^{36}Cl$ was also calculated, and the ^{36}Cl depth profile is identical with that of $^{40}Ca(n,\gamma)^{41}Ca$ but is (atom/min · g · Cl) 88 times larger, due to the larger thermal cross section of ^{35}Cl. For the ^{235}U fission rate in Fig. 5, the agreement between the Lunar Neutron Probe Experiment data [9] and calculations is very good.

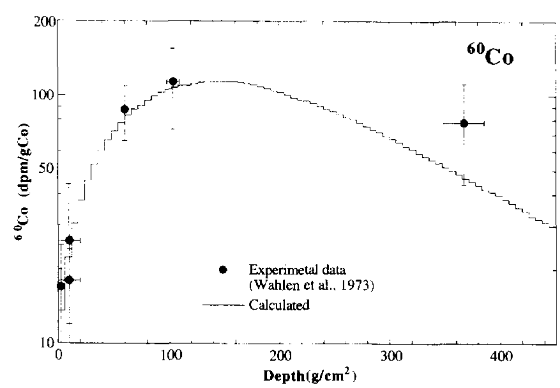

Fig. 4. Calculated neutron capture rates of ^{60}Co (solid line) vs. depth compared with the measured activity of ^{60}Co in the Moon [10].

There is a similarly good agreement of LCS calculations [22] with the results of the LNPE [13] for the $^{10}B(n,\alpha)^7Li$ reaction.

4. Conclusions

Measurements of ^{41}Ca in nine samples from the Apollo 15 deep drill core from very near the lunar surface (4 g/cm^2) to a depth of 388 g/cm^2 have been performed using AMS. The dominant production mode for ^{41}Ca is via thermal neutron capture on ^{40}Ca. Following minor corrections for the spallogenic production channel, the resultant ^{41}Ca depth profile in the Apollo 15 core maps the development and intensity of the thermal neutron flux in the Moon. A peak ^{41}Ca production of 80 dpm/kg (or 1 dpm/g Ca) occurs at a depth of 150 g/cm^2 (about 90 cm), which precedes an exponential fall-off in production with an e-folding length of 175 g/cm^2. This is in contrast to the behavior in the production of spallogenic nuclides in the Moon, where production peaks at ~ 40 g/cm^2. Most spallogenic nuclides also attenuate with e-folding lengths of ~ 175 g/cm^2.

Our ^{41}Ca profile shows good agreement with calculations based on the LAHET Code System. These calculations were extended to other lunar thermal neutron data sets for ^{60}Co and the LNPE data for the ^{235}U fission rate, with both also exhibiting good agreement. The successes observed in modeling these low-energy and thermal neutron reactions augers well for the application of LCS to other neutron-capture reactions, such as for meteorite shielding and size studies, planetary elemental mapping studies using gamma rays following neutron capture, and estimating neutron corrections for spallogenic nuclides such as ^{36}Cl.

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