Cross-Section Measurements for Proton- and Neutron-Induced Reactions Needed to Understand Cosmic-Ray Interactions on Earth and in Space

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Abstract. Primary cosmic rays interact directly with extraterrestrial bodies and cosmic-ray shower particles interact with the earth’s surface to produce small quantities of radionuclides and stable isotopes, which are measured routinely using appropriate techniques. Theoretical models are used to analyze these measurements to learn the history of the object or the cosmic rays that fell upon it. Cross sections for reactions producing these cosmogenic nuclides are essential input to these models. Most primary cosmic rays are protons, and good measurements of the cross sections for proton-induced reactions are essential. Most relevant cross sections are now well measured, but discrepancies still exist between the measurements and calculations. One explanation is that neutrons produced in primary cosmic-ray interactions also initiate spallation reactions contributing significantly to the cosmogenic nuclide inventory, but few of the relevant cross sections have been measured at energies >30 MeV. We have measured many of these needed cross sections for neutron-induced reactions using two different techniques. Cross sections at selected unique neutron energies >70 MeV are measured at iThemba LABS, South Africa (iTL) using quasi-monoenergetic neutron beams. Energy integrated (average) cross sections are measured at the Los Alamos Neutron Science Center (LANSCE), using ‘white’ neutron beams with an energy range of 0.1–750 MeV.

INTRODUCTION

In space, primary cosmic rays interact directly with extraterrestrial bodies to produce small quantities of short-lived (e.g., $^{22}$Na, $^{56}$Co, $^{54}$Mn), long-lived (e.g., $^{14}$C, $^{10}$Be, $^{26}$Al) radionuclides, and stable isotopes (e.g., Ne and Ar isotopes). These cosmogenic nuclides are measured routinely in small samples from meteorites and the lunar surface using a variety of techniques, which include non-destructive gamma-ray spectroscopy, Accelerator Mass Spectrometry (AMS), and Mass Spectrometry (MS) [1]. On earth, primary cosmic rays interact with the earth’s atmosphere to produce showers of cosmic-ray particles and these particles interact with the earth’s surface to produce cosmogenic nuclides [2]. Recent advances in the measurement techniques allow some nuclides to be measured routinely in samples from the earth’s surface.

Theoretical models have been developed to analyze these cosmogenic nuclide inventories. From these analyses, it is possible to learn the history of an object in space, for example, the exposure history and pre-atmospheric size of meteorites, e.g., [3]. From depth profiles of different cosmogenic nuclides measured in lunar samples, solar proton fluxes over the past million years can be estimated [4]. These estimates can be compared to the direct measurements of the solar proton flux, possible only over the past few solar cycles [5].

On earth, the field of terrestrial cosmogenic nuclides has evolved rapidly over the past few years and there are many earth-science applications such as surface exposure dating, paleoclimatology, geomorphology, tectonics, hydrology, and volcanology [6]. Most cosmogenic nuclides produced in the earth’s surface are due to neutron-induced reactions, although in some circumstances thermal neutron-induced and muon capture reactions are important.
The theoretical models developed by Reedy and Arnold demonstrate well the input data required to analyze the cosmogenic nuclide archives in meteorites or the lunar surface [7,8]. The basic expression is given in Eq. (1).

\[
P_x(d,R) = \sum_i N_i \sum_j \left[ \int_0^\infty \sigma_{ij}(E) \phi_j(E,d,R) dE \right],
\]

where \(N_i\) = the abundance of the \(i\)th element, \(\sigma_{ij}(E)\) = the cross section for producing nuclide \(x\) from element \(i\) with particle \(j\) of energy \(E\), and \(\phi_j(E,d,R)\) = flux of particle \(j\) at energy \(E\) and depth \(d\), in a body of nominal radius \(R\).

From Eq. (1), it is obvious that production rates must be measured in a sample with a well-known composition. The most abundant constituents of meteorites and the lunar surface include oxygen, silicon, calcium, magnesium, aluminum, iron, nickel, and potassium.

Also obvious from Eq. (1) is that the cross sections for all the relevant reactions in the sample producing the cosmogenic nuclide under study must be well known. Most cosmic rays are protons (~98% of solar (SCR) and ~87% of galactic (GCR)), so the primary need is for the cross sections of proton-induced reactions. Most SCR particles have energies <200 MeV, lose energy by ionization losses, and penetrate only the top few centimeters of an extraterrestrial sample. GCR particles can have energies up to \(\sim 10^{20}\) eV, penetrate many meters into an object, and nearly all lose energy by initiating spallation reactions producing many neutrons. These secondary neutrons also initiate a significant number of spallation reactions particularly at depth in an object, so the cross sections for neutron-induced reactions are also important. The remaining SCR and GCR particles are almost all alpha particles with only ~1% of GCR being higher-Z particles. The interactions of these particles do not contribute significantly to the total cosmogenic nuclide inventory [9].

If the calculated production rate and the measured production rate differed, the discrepancy was blamed on the lack of good cross-section measurements to use as input. Now most cross sections for the relevant proton-induced reactions are well measured, e.g., [4,10,11]. However, when using these cross-section measurements for proton-induced reactions in the calculations there can still be discrepancies between calculation and measurement for some cosmogenic nuclides, e.g., [12].

One possible explanation is that the contribution to the total cosmogenic nuclide inventory from secondary neutron-induced reactions should be included explicitly in the calculations. Unfortunately, almost no cross sections for relevant neutron-induced reactions have been reported at neutron energies \(\sim 30\) MeV [13,14]. If the cross section for the neutron-induced reaction is not known, often the cross section for the corresponding proton-induced reaction, or a cross section calculated from nuclear model calculations, or a cross section estimated from thick target bombardments made with high-energy proton beams is used. None of these are ideal solutions.

Over the past 10 years, we began by measuring cross sections for proton-induced reactions using thin target techniques, e.g., [4,11]. Once these were complete, we began to measure cross sections for neutron-induced reactions to use as input to the theoretical models. Two different techniques were used. The first used selected quasi-monoenergetic neutron energies >70 MeV at iThemba LABS, South Africa (iTL), and the second measured an energy-integrated (average) cross section using ‘white’ neutron beams with an energy range of 0.1–750 MeV at the Los Alamos Neutron Science Center (LANSCE), Los Alamos [14–17]. Measurements to be made in the future will focus on improving our understanding of cosmogenic nuclide production in the terrestrial environment. Cross sections for neutron-induced reactions will be measured at both quasi-monoenergetic energies and using ‘white’ neutron beams with different energy spectra to simulate different irradiation conditions on the earth’s surface.

**EXPERIMENTAL METHODS**

Our cross-section measurements at both iTL and LANSCE have several experimental parameters in common. At both iTL and LANSCE, the neutron beam was large enough to cover the target stack entirely. In nearly all irradiations, targets 50 x 50 mm were used. The thickness of the total target stack was designed to attenuate the neutron beam by <10% at almost all neutron energies. Small 15-mm-diameter monitor foils of C, Al, Ti, Fe, Ni, Cu, and Au were included at the back of the target stacks to search for suitable monitor reactions. Cu monitor foils were included so that we could compare our measurements to those of Kim et al. [18], one of the few sets of published cross sections at high neutron energies. Irradiation times were chosen to produce the optimum number of atoms required for AMS or MS determination of the nuclide under study. These irradiation conditions allowed good cross-section measurements for many of the reactions producing short-lived radionuclides.
The first technique used quasi-monoenergetic neutron beams generated by 80-, 120-, or 160-MeV protons on a Be target at iTL. The exact neutron energy incident on the target depended on the exact experimental set-up and thickness of the Be target used. At each energy, two identical target stacks were irradiated simultaneously: one stack in the beam at zero degrees and one in the beam at 16 degrees to the incident proton direction. The neutron energy spectrum at 16 degrees simulates that of the low-energy tail at zero degrees. Subtracting the normalized yield of the nuclide under study produced at 16 degrees from the yield produced at zero degrees allows the cross section to be calculated at an almost unique neutron energy. Typical irradiation times were ~40–50 hours [14–17].

The second technique measured an average cross section using a ‘white’ neutron beam with an energy range of 0.1–750 MeV at LANSCE. Neutrons with energies <0.1 MeV were removed from the beam by 2 inches of polyethylene placed well upstream of the target stack. A calibrated uranium fission chamber was used to measure the neutron fluence with energies above some instrumental low-energy cut-off through the target stack. Irradiation times ranged from half a day to several days. Corrections were made for all periods of no beam that lasted more than ~15 minutes; these corrections were greatest for the short-lived radionuclides produced [14–17].

After irradiation at either iTL or LANSCE, the short-lived radionuclides produced in both the targets and the monitor foils were determined using non-destructive gamma-ray spectroscopy. The cross sections for many of reactions producing short-lived radionuclides have been measured. After completion of these measurements, the targets are sent for AMS or MS analysis. The necessary chemical preparation before AMS determination is complete for many of the irradiated targets. At a later date, the yields of \(^{10}\)Be, \(^{14}\)C, \(^{20,21,22}\)Ne, \(^{26}\)Al, \(^{36}\)Cl, and \(^{41}\)Ca will be measured.

**RESULTS AND DISCUSSION**

The status of the cross-section measurements for the production of short-lived radionuclides at iTL and LANSCE is summarized in Table 1. These measurements took place over several years, so there are replicate independent measurements for many cross sections. At iTL, the energy of the incident neutron beam depended on the exact experimental conditions and so varied a little from year to year. The cross-section measurements for neutron-induced reactions producing long-lived radionuclides and stable isotopes that are still in progress are given in Table 2.

Before we had the final cross section values for the reactions listed in Table 1, we used our preliminary values to recalculate the \(^{22}\)Na production rate in lunar rocks and were able to demonstrate that using measured cross sections for neutron-induced reactions explicitly in the model calculation led to calculated production rates in much better agreement with the measurements [15,17].

The average cross-section measurements for neutron-induced reactions are proving to be very useful tools. Not only are they good information in themselves, but their real value may be to help define the behavior of the cross section as a function of energy for a particular reaction. From the calibration of the uranium fission chamber used to monitor the neutron fluence at LANSCE, we know the incremental neutron fluences over finite sized energy bins from 0.1 to 750 MeV. These incremental fluences, the cross sections measured at iTL at unique neutron energies, plus any other reported cross-section measurements, can be used to calculate a theoretical average cross section. If this theoretical value agrees with the value measured at LANSCE, then the cross sections used as input to the calculation are probably correct [14–17].

**CONCLUSIONS**

We are in the process of measuring relevant cross sections for neutron-induced reactions that are needed to improve our understanding of cosmic ray interactions with extraterrestrial materials. Good measurements of cross sections for many neutron-induced reactions producing short-lived radionuclides are complete. From these measurements, we have shown that production rates calculated using theoretical models using our preliminary values of these cross sections as input, are in better agreement with measured production rates. This indicates that secondary neutron-induced reactions do indeed contribute significantly to the total cosmogenic nuclide inventory in extraterrestrial materials.
TABLE 1. Completed cross-section measurements for neutron-induced reactions.

<table>
<thead>
<tr>
<th>Product</th>
<th>70.7 – 73.5 MeV</th>
<th>110.8 – 112.2 MeV</th>
<th>151.6 MeV</th>
<th>Average 0.1–750 MeV</th>
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<tr>
<td>7Be</td>
<td>SiO₂</td>
<td>SiO₂</td>
<td>SiO₂, Al</td>
<td>SiO₂, Si, Al, Mg</td>
</tr>
<tr>
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<td>SiO₂, Al</td>
<td>SiO₂, Al</td>
<td>SiO₂, Al</td>
<td>SiO₂, Si, Al, Mg</td>
</tr>
<tr>
<td>24Na</td>
<td>SiO₂, Al</td>
<td>SiO₂, Al</td>
<td>SiO₂, Al</td>
<td>SiO₂, Si, Al, Mg</td>
</tr>
<tr>
<td>42Sc</td>
<td>Ti, Fe</td>
<td>Ti, Fe</td>
<td>Fe</td>
<td>Ti, Fe, Ni, Cu</td>
</tr>
<tr>
<td>42Sc</td>
<td>Ti, Fe</td>
<td>Ti, Fe</td>
<td>Fe</td>
<td>Ti, Fe, Ni, Cu</td>
</tr>
<tr>
<td>51Cr</td>
<td>Fe</td>
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</table>

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REFERENCES