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Beam Interactions with Materials & Atoms

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Simulations of terrestrial in-situ cosmogenic-nuclide production

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Targets of silicon and silicon dioxide were irradiated with spallation neutrons to simulate the production of long-lived radionuclides in the surface of the Earth. Gamma-ray spectroscopy was used to measure ^{7}Be and ^{22}Na, and accelerator mass spectrometry was used to measure ^{10}Be, ^{14}C, and ^{26}Al. The measured ratios of these nuclides are compared with calculated ratios and with ratios from other simulations and agree well with ratios inferred from terrestrial samples.

1. Introduction

The interactions of galactic-cosmic-ray particles in the Earth's atmosphere produce a cascade of particles, some of which reach the Earth's surface and produce cosmogenic nuclides. Neutrons are the dominant producer of nuclides in the top meter of the Earth's surface, and muons become a major source of cosmogenic nuclides below a few meters. Long-lived cosmogenic radionuclides, such as 5730-year ^{14}C, 0.3-Ma ^{36}Cl, 0.7-Ma ^{26}Al, and 1.5-Ma ^{10}Be, and a few rare stable nuclides, such as ^{3}He and ^{21}Ne, made in-situ in certain materials can be used to study recent exposure histories [1]. The advances in the analyses of long-lived radionuclides using accelerator mass spectrometry (AMS) have revolutionized the use of these radionuclides, especially for in-situ terrestrial applications. At present, the use of these cosmogenic nuclides to study histories of targets or of cosmic radiation is often limited by inadequately known production rates.

Some production rates and ratios have been inferred from measurements of terrestrial samples with known irradiation conditions (e.g., refs. [2-6]). There are some uncertainties in the exposure ages and irradiation conditions of these samples, and only a few radionuclides (e.g., ^{10}Be and ^{26}Al) have been measured. A wide range of production rates have also been theoretical inferred (e.g., refs. [7,8]). These and other calculations (e.g., refs. [9,10]) for production of these nuclides by nucleons and muons could be improved with laboratory measurements of production cross sections and relative production ratios.

Laboratory simulations of these processes have many limitations, such as not reproducing the complex mix of particles and their energies, but do provide a controlled irradiation of well-characterized samples. A series of irradiations at the Los Alamos Clinton P. Anderson Meson Physics Facility (LAMPF) have simulated the production of long-lived radionuclides in surface rocks. Here we report on synthetic quartz and silicon that were exposed to neutrons. Preliminary results with some details not presented here were reported earlier [11-13]. Irradiations with muons were also done [11,12] and will be reported separately.

2. Experimental

To simulate the production rates and ratios due to the nucleon component (primarily neutrons) of cosmic rays, an irradiation was conducted using spallation neutrons produced in the beam stop of the ~ 1-mA 800-MeV proton beam at LAMPF. The beam stop produces a large flux of secondary particles, especially neutrons. Most charged secondary particles are stopped by ionization energy losses near the beam stop. Neutrons travel until they undergo nuclear interactions. Samples were exposed to these particles in the Los Alamos Spallation Radiation Effects Facility

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Table 1
Measured radionuclide concentrations (10^{10} atoms/g) in irradiations with spallation neutrons near the LAMPF beam stop (numbers in parentheses are the uncertainties of the last digits of the measurement)

Target	^{7}Be	^{10}Be	^{14}C	^{22}Na	^{26}Al [a]	^{26}Al [b]
Si	40.1 (0.4)	7.69 (0.46)	19.2 (4)	553 (5)	1320 (90)	1410 (110)
SiO_2	162 (1)	93.0 (4.7)	303 (3)	256 (5)	722 (50)	660 (52)
O [c]	269 (2)	168 (9)	552 (6)	–	–	–

[a] Measured in that sample only.
[b] From averages based on measurements of both Si-containing samples.
[c] Oxygen, as inferred from the SiO_2 and Si measurements.

(LASREF) around the beam stop. The particle distributions at various locations in LASREF have been characterized [14,15] and are roughly similar to those in the Earth's surface. Targets of silicon, SiO_2, and several monitor foils were irradiated for about a day with these spallation neutrons.

The activities in the monitor foils and of the short-lived radionuclides, such as 2.6-a ^{22}Na and 53-d ^{7}Be, produced in the silicon and SiO_2 were determined by non-destructive high-resolution gamma-ray spectrometers at Los Alamos. At LAMPF, pieces of the silicon and SiO_2 were dissolved along with Al and Be carriers. The Al and Be were separated and taken to San Diego, where they were further purified [16,17]. The ^{26}Al/^{27}Al and ^{10}Be/^{9}Be ratios were measured on the University of Pennsylvania's tandem Van de Graaff accelerator [18,19]. The measured concentrations of these radionuclides are given in Table 1.

Measurements of ^{14}C were separately made for these beam-stop samples. Two different extractions were performed at Tucson on samples of a few mg of Si and SiO_2 and also on samples that had been physically diluted with quartz powder. Samples were precombusted to remove any organic contamination and then heated to melting [20]. Any CO was converted to CO_2. The CO_2 was measured volumetrically and reduced to graphite. The graphite was analyzed along with standards by AMS at the University of Arizona NSF Accelerator Facility for Radioisotope Analysis as described in ref. [21]. The results for the two different extractions agreed very well [13], and only the averages are given in Table 1.

3. Results

The concentrations of the radionuclides (Table 1) were high and easily measured. The production of ^{7}Be, ^{10}Be, and ^{14}C from pure oxygen in Table 1 was determined from the Si and SiO_2 measurements. The ^{22}Na in the SiO_2, which is made only from the silicon, is in good agreement (1%) with the ^{22}Na measurement in Si. However, the ^{26}Al measurements in Si (132×10^{11} atoms/g) and in the SiO_2 corrected to pure Si (154×10^{11} atoms/g) disagree by 17%, which is slightly greater than the sum of the $\approx 7\%$ errors for each measurement. Below, we use the average of these values, 141×10^{11} atoms/g for pure Si and 66×10^{11} atoms/g for SiO_2.

The ^{7}Be/^{10}Be ratio in the Si is 5.2, which is less than the ratio of ≈ 7.7 (5.39 mb/≈ 0.7 mb) measured in Si irradiated with 600-MeV protons [22,23]. The ^{7}Be/^{10}Be production ratio in oxygen is 1.6, which is much less than the proton-induced ratios in oxygen of 8.9 and 5.4 at 135 and 550 MeV, respectively (from ref. [24], using revised half-lives), and of 171 and 15.7 at 49 and 91 MeV, respectively [25]. As evident from the above ^{7}Be/^{10}Be ratios and as previously noted for ^{10}Be [26], neutrons and protons produce these two nuclides in relatively different yields and cross sections.

These results yield ^{26}Al/^{10}Be ratios of 183 in Si and 7.1 in SiO_2. The ^{10}Be and ^{26}Al contents of quartz from glacially-polished rock exposed to cosmic rays for ≈ 11 ka gave an ^{26}Al/^{10}Be ratio of 6.0 ± 0.4 [4]. Other measurements for natural samples gave similar production ratios, see Table 2. These ^{26}Al/^{10}Be production ratios agree well with our ratio of 7.1 ± 0.7 from spallation neutrons reacting with SiO_2.

The ^{10}Be/^{7}Be and ^{26}Al/^{10}Be ratios that we obtained from our irradiations can also be compared with preliminary results [11,12] from the irradiation of SiO_2

Table 2
^{26}Al/^{10}Be ratios measured from these simulations or some terrestrial samples and several calculated ratios

Sample(s)	Measured ratio(s)		Calculated production ratio	
Early predictions			4.2 [8]–20.7 [7]	
Libyan desert glass	$\lesssim 7$	[2]		
In-situ quartz	2.5–6.7	[3]	$\Rightarrow \approx 6$	[3]
Sierra quartz	6.0 ± 0.4	[4]	≈ 8 [a]	
Antarctic rocks	~ 6.2	[5]	≈ 8 [a]	
Antarctic rocks	6.5 ± 1.3	[6]	≈ 8 [a]	
LAMPF, neutrons	7.1 ± 0.7 [a]		≈ 8 [a]	

[a] This work.

with stopping negative muons (μ^-). The $^{10}Be/^{7}Be$ ratios vary widely (e.g., ~ 23 for the stopped μ^-), even greater than the variations noted above for cross-section ratios at various proton energies. The $^{26}Al/^{10}Be$ ratios for stopped muons (≈ 7.0) and neutrons are similar.

Our ratios for $^{14}C/^{10}Be$ in Si, SiO_2, and oxygen are 2.50, 3.26, and 3.29, respectively. Using the proton cross sections of refs. [22,23,25] for ^{10}Be and of ref. [27] for ^{14}C, we can compare our ratios for neutrons with proton-induced ratios. These ratios for protons reacting with Si and O increase with decreasing proton energy, with ratios near unity for ~ 500 MeV and ~ 10 for ~ 50–70 MeV protons, but scatter about our measured ratios.

Using the cross sections for ^{10}Be and ^{26}Al from ref. [26] and ref. [10], respectively, $^{26}Al/^{10}Be$ ratios were calculated for both the LAMPF irradiations and for natural irradiations. Although the exact spectral shapes for the energetic particles in these irradiations are not well known, we can get some ideas of relative trends and whether the cross sections are reasonably consistent with the measurements. Our calculated $^{26}Al/^{10}Be$ ratios in Table 2 are in good agreement with the ratios from our simulation and for natural quartz. Using the cross sections for ^{22}Na from Si in ref. [9], we calculated a $^{26}Al/^{22}Na$ ratio similar to the measured ratio. Our results and the cross sections for ^{10}Be production in ref. [26] suggest that the neutron-induced cross sections for ^{7}Be from oxygen are ~ 0.7 of those measured for protons. For ^{14}C, we found that we needed to increase the assumed cross sections of ref. [9] for production of ^{14}C from oxygen by 10% and more at the lowest energies to get better agreement with the measured ratio.

Our cross sections for ^{10}Be, ^{14}C, and ^{26}Al gave good agreement between calculated production rates and activities measured in the Knyahinya meteorite [28]. To get production rates for terrestrial samples, we plan to use the Monte Carlo particle transport/production codes used by ref. [28] and our cross sections.

4. Conclusions

Spallation neutrons near the LAMPF beam stop were used to study the production of ^{7}Be, ^{10}Be, ^{14}C, ^{22}Na, and ^{26}Al in silicon and SiO_2. These irradiations gave $^{26}Al/^{10}Be$ ratios similar to those measured with documented natural samples, indicating that other ratios from our irradiations could be applied to natural samples. Production ratios varied with the target and with the energy and the nature of the incident particles, illustrating the complex nature of predicting such nuclear interactions and their ratios.

Several excitation functions for the production of these radionuclides were tested. Some sets of cross sections (^{10}Be, ^{22}Na, and ^{26}Al) were found to be good. Other cross-section sets for production by neutrons had to be modified (^{14}C) or were shown to be poor (^{7}Be). These good or modified cross sections are being used for calculations of cosmogenic-nuclide production rates in extraterrestrial materials and could be used for terrestrial applications.

While simulations at accelerators, such as those reported here, have limitations, they are useful in determining and checking relative production rates in terrestrial samples. The controlled nature of such irradiations is an advantage for many problems, such as determining production from elements that are hard to study directly in natural samples, such as sodium. They also can give production ratios for radioactive nuclides relative to stable nuclides, e.g. $^{26}Al/^{21}Ne$.

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