THERMAL IONIZATION MASS SPECTROMETRY STUDIES OF SM AND GD ISOTOPIC SHIFTS IN LUNAR METEORITES DUE TO NEUTRON CAPTURE: A PROGRESS REPORT. K. C. Welten[1], T. L. Owens[2], D. J. DePaolo[2], [1]Space Sciences Laboratory, University of California, Berkeley, CA 94720-7450 (e-mail: kcwelten@berkeley.edu); [2]Center for Isotope Geochemistry, Department of Earth and Planetary Science, UC Berkeley & Lawrence Berkeley National Laboratory.

Introduction. Among meteorites, the lunar meteorites have unique cosmic-ray exposure (CRE) histories with long exposure times on the Moon, followed by very short transfer times from the Moon to Earth, typically 10^3-10^6 yr. Due to these short transfer times of lunar meteorites in space, the inventories of most cosmogenic radionuclides with half-lives of 10^5-10^7 yr contain contributions from exposure on the Moon. The radionuclide results indicate that about half of the lunar meteorites were irradiated at depths of <1000 g/cm^2 (<5 m) during their recent cosmic-ray exposure on the Moon [1]. While the cosmogenic radionuclides only constrain the irradiation conditions during the last few Myr, the concentrations of stable cosmogenic noble gases in lunar meteorites suggest exposure times of ~100 Myr to a few Gyr [2]. However, the details of this long exposure on the Moon are not well constrained, because the cosmogenic noble gas concentrations and their ratios do not provide accurate information on the average irradiation depth. Consequently, it is not clear how long the lunar meteorites were exposed near the surface of the Moon and if they were exposed at a constant depth, or if their exposure conditions in the lunar regolith changed with time as a function of lunar impact events.

Method. To fill this gap in our knowledge of the evolutionary history of meteorites from the lunar surface, we will measure the stable isotope composition of Samarium (Sm) and Gadolinium (Gd). Several isotopes of these elements, specifically ^{149}Sm and ^{157}Gd, have exceptionally large neutron-capture cross sections. If these nuclides are subjected to bombardment by low-energy neutrons, the ^{150}Sm/^{149}Sm and ^{158}Gd/^{157}Gd ratios are shifted away from their average solar system abundance. The degree of this shift is a function of the integrated low-energy neutron dose, which is much higher for 2π exposure on the lunar surface than for 4π exposure as a small object in space. In our quest to understand the conditions and duration of cosmic-ray exposure of lunar meteorites on the surface of the Moon (before their ejection into space as small objects) we will measure Sm and Gd isotopic compositions in lunar meteorites that were ejected from the top few meters of the lunar surface. Based on measured cosmogenic noble gas concentrations in lunar meteorites, we expect isotopic shifts in ^{150}Sm/^{149}Sm and ^{158}Gd/^{157}Gd of 0.1-5‰ (1-50 epsilon units), depending on the average depth at which the meteorites were irradiated on the lunar surface. The depth dependence of the lunar neutron flux is well understood based on previous measurements of the lunar neutron flux [3] and of neutron-capture produced ^{41}Ca in the Apollo 15 drill core [4].

Chemical procedures. We dissolved aliquots of 100-150 mg of terrestrial rock standards in HF/HNO_3. The standards include BCR-2 (USGS) and several standards from the Geological Survey of Japan, including two basalts (JB-2, JB-3) that were previously measured by others [5]. After complete dissolution of the samples, we separated the REE's from the major elements using a 100 ml Dowex AG50W X8 cation column and then separated Sm and Gd from the other REE's on a Dowex AG50W X4 column by elution with 0.2M 2-methyllactic acid. We typically loaded 300-600 ng of the Sm and Gd fractions on a Re double filament. For the Sm and Gd standards, we loaded 100 ng of Sm and 500 ng of Gd, both prepared from Ames high purity metal.

TIMS measurements. To measure the isotopic composition of Sm and Gd, we used UC Berkeley's Thermo Finnigan Triton multi-collector system, which is equipped with one fixed Faraday cup in the center and 4 movable Faraday cups on each side.

For $^{152}Sm^+$ we obtain typical beam intensities of 1-5 V. We simultaneously measured the seven isotopes of Sm as well as interfering species of ^{143}Nd on the low-mass side and ^{155}Gd on the high-mass side. A typical measurement consists of 100-200 cycles of 10 seconds each. We corrected for instrumental mass fractionation effects using a ^{147}Sm/^{152}Sm ratio of 0.56083 [6]. We corrected the ^{144}Sm, ^{148}Sm and ^{150}Sm signals for Nd interferences, based on the measured ^{143}Nd/^{152}Sm ratios of ~3×10^{-5} for the Sm standard and ~2×10^{-4} for the terrestrial basalt (Table 1). Corrections for Gd interferences at mass 152 and 154 are negligible for the Sm standard, while the correction for ^{154}Gd interferences in JB-2 is small but significant (~0.01%).

For the Gd standard, we obtained $^{158}Gd^+$ beam intensities of 1-5V at moderate settings and a maximum of ~15V at high ionization settings. For terrestrial samples, we obtained much lower and less stable beam intensities (<1V), but we anticipate to achieve stable beam intensities of >1 V in the near future. For the Gd standard, we simultaneously measured the

seven isotopes as well as possible interferences of ^{150}Sm and ^{162}Dy. Corrections for Sm and Dy interferences were negligible for the Ames Gd standard. We corrected for instrumental mass fractionation effects based on a ^{156}Gd/^{160}Gd ratio of 0.9361 [6]. This assumption may not always be correct for lunar samples, since the ^{156}Gd/^{160}Gd ratio may be elevated due to neutron capture on ^{155}Gd. For lunar meteorites, we will thus make corrections for mass fractionation using a constant (^{155}Gd+^{156}Gd)/^{160}Gd ratio of 1.6129, assuming that the depletion of ^{155}Gd due to neutron capture is equal to the enrichment of ^{156}Gd [5,7].

Results. Preliminary results for Sm and Gd are shown in Tables 1 and 2, respectively. The measured Sm isotopic composition of two aliquots of the Ames metal standard shows excellent agreement of all ratios with literature values and shows typical uncertainties of 5-20 ppm. For the terrestrial rock standard, JB-2, corrections for ^{144}Nd, ^{148}Nd and ^{150}Nd interferences based on the measured ^{143}Nd/^{152}Sm ratio lead to overcorrection of ^{144}Sm, ^{148}Sm and ^{150}Sm by a factor of ~2. Although this may suggest that the interference at mass 143 is not entirely due to ^{143}Nd, we do not fully understand this discrepancy. We assume for now that the elevated ^{144}Sm/^{152}Sm ratio is the best measure of the amount of Nd interference and made corrections for ^{148}Nd and ^{150}Nd interferences accordingly. We will strive to further reduce the Nd interference. Despite these uncertainties in the Nd corrections, the ^{149}Sm/^{152}Sm and ^{150}Sm/^{152}Sm ratios in the Sm standard as well as the terrestrial rock sample are already reliable enough to detect isotopic shifts of the magnitude expected in lunar meteorites. Table 2 shows that the Gd isotopic ratios in the Gd standard are also consistent with values measured by others [6,7], but more works is needed to obtain reliable Gd results from rock samples.

Future work. We have selected several lunar meteorites from the Antarctic meteorite collection and hope to present preliminary Sm isotopic data for a few of these samples at the meeting. We also anticipate to measure Gd isotopic ratios in terrestrial rocks and possibly lunar meteorites in the next few months.

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References: [1] Nishiizumi K. (2004) *Meteorit. Planet. Sci*. 39, A77. [2] Lorenzetti S. et al. (2005) *Meteorit. Planet. Sci.* 40, 315-327. [3] Woolum D. S. and Burnett D. S. (1974). *Earth Planet. Sci. Lett.* 21, 153-163. [4] Nishiizumi K. et al. (1997) *Earth. Planet Sci. Lett*. 148, 545-552. [5] Hidaka H. et al. (1995) *Anal. Chem*. 67, 1437-1441. [6] Russ G. P. et al. (1971) *Earth. Planet Sci. Lett*. 13, 53-60. [7] Hidaka H. et al. (2000) *Meteorit. Planet. Sci*. 35, 581-589.

Table 1. Measured Sm isotope ratios in Ames Sm metal (STD) and terrestrial basalt (JB-2) in comparison to literature values [5,6]. Measured ratios are corrected for instrumental mass fractionation effects using ^{147}Sm/^{152}Sm=0.56083 [6].

Sample	^{143}Nd/^{152}Sm	^{144}Sm/^{152}Sm	^{148}Sm/^{152}Sm	^{149}Sm/^{152}Sm	^{150}Sm/^{152}Sm	^{154}Sm/^{152}Sm	^{155}Gd/^{152}Sm
STD-a	0.000033	0.114932±7	0.42044 ± 1	0.51683 ± 3	0.275997±7	0.85080 ± 2	–
STD-b	0.000026	0.114954±3	0.420446±4	0.516845±5	0.275998±4	0.850792±8	0.000005
Lit. [6]	–	0.11499 ± 1	0.42045 ± 2	0.51686 ± 2	0.27600 ± 2	0.85079 ± 3	–
JB-2 *	0.000195	0.114954±5	0.420456±8	0.516861±8	0.275997±6	0.85077 ± 2	0.000740
JB-2 [5]		0.114952±3	0.420461±4	0.516855±5	0.275993±5	0.850834±3	

*corrections for Nd interferences in JB-2 at masses 148 and 150 were made based on assumption that elevated ^{144}Sm/^{152}Sm ratio of 0.115144 is due to ^{144}Nd.

Table 2. Measured Gd isotope ratios in Ames Gd metal (STD) in comparison to literature values [6]. Measured ratios are corrected for instrumental mass fractionation effects, using ^{156}Gd/^{160}Gd=0.93610 [6].

Sample	^{150}Sm/^{160}Gd	^{152}Gd/^{160}Gd	^{154}Gd/^{160}Gd	^{155}Gd/^{160}Gd	^{157}Gd/^{160}Gd	^{158}Gd/^{160}Gd	^{162}Dy/^{160}Gd
Lit. [6]	–	0.00928±8	0.09974 ± 3	0.67687 ± 5	0.71588 ± 4	1.13582 ± 5	–
STD-a	0.00001	0.009279±1	0.099719±2	0.676940±50	0.715854±10	1.135851±8	<0.000005
STD-b	<0.00001	0.009280±1	0.099728±2	0.676944 ± 5	0.715871 ± 4	1.135864±5	<0.000005