LUNAR $^{236}$U AND SOLAR COSMIC RAY FLUX. * P. R. Fields, H. Diamond, D. N. Metta and D. J. Rokop, Chemistry Division, Argonne National Laboratory, Argonne, Illinois 60439.

$^{236}$U (2.34 x 10$^6$ yr half-life) and $^{237}$Np (2.14 x 10$^6$ yr) have been found widely in lunar samples (1,2). Their abundances have implied a larger galactic cosmic ray induced neutron flux, and/or a larger solar cosmic ray proton flux than can be inferred from recent direct observations, or from other cosmogenic radionuclides in lunar and meteoritic samples. The $^{236}$U:$^{238}$U ratio at three depths in rock 12002 are reported. Different samples of 12070 have varying $^{236}$U:$^{238}$U ratios, although there is very good agreement between separate measurements of the same sample. It is suggested that the very fine fragments contain a component derived from ablation and that these fines are not homogeneously distributed in 12070. Re-examination of the $^{236}$U:$^{238}$U ratios in dummies support the validity of the measurements. A small intermittent reagent blank has inhibited the presentation of new results from this laboratory; results derived from uranium samples processed elsewhere are presented.

Experimental

Before and after each group of samples, the $^{236}$U:$^{238}$U ratio of Port Hope uranium ($^{236}$U:$^{238}$U = 6 x 10$^{-10}$) (3) is measured to insure against machine-related spurious signals. Dummy uranium samples with small amounts of $^{236}$U are used to check the position of the $^{236}$U peak. In some samples spurious counts at the mass 236 position were observed upon initial heating. These counts are not present upon subsequent heating. All data in which the temperature fractionation of $^{236}$U has not been tested have been rejected in this and in all earlier papers. Each group of samples are also processed along with reagent blanks containing iron and 3 µg of Port Hope uranium. Some of these reagent blanks have recently been as high as $^{236}$U:$^{238}$U = 25 x 10$^{-9}$, but the results are not reproducible, and the source of the spurious $^{236}$U has not yet been traced. Consequently, no new results (other than 12070, 136 below) are reported for samples processed in this laboratory.

Uranium, or uranium-rich fractions obtained from other laboratories have been measured, and are reported in Table 1. These are not accompanied by full reagent blanks, but the low values of $^{236}$U:$^{238}$U in each set support the assumption that the higher values of $^{236}$U arise from cosmic ray irradiation of lunar uranium.

The tube and pipette from which 12070,91 were originally loaded onto the mass spectrometer was leached and this was again mass spectrometrically examined (three years after the original measurement). In addition a new sample, 12070,136 was processed, in a manner reported earlier (4) and the $^{236}$U:$^{238}$U was measured.

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Results and Discussion

There is no clear pattern in the $^{236}$U:$^{238}$U ratios of the layers of 12002, that resembles either solar cosmic ray proton reactions, or galactic cosmic ray induced neutron reactions. Examples of both kinds cosmogenic reactions are clearly seen for successive layers of 12002 in references 5 and 6.

The ablation products of the lunar surface should be rich in solar cosmic-ray induced radioactivity and this may be consistent with finding very high $^{236}$U:$^{238}$U ratios in fines 12070,91 and 15080,2. There was however, no large amount of $(7.4 \times 10^5 \text{ yr})$ $^{26}$Al in 12070,91 (7) and a large amount of solar-cosmic-ray produced $^{26}$Al would be expected unless much of the irradiation that produced $^{236}$U occurred more than 2 million years ago.

There have been five analyses of 12070 uranium with anomalous results. Originally, 12070,91 was analyzed and $^{236}$U:$^{238}$U = $(215 \pm 15) \times 10^{-9}$ was reported (1). Recently, the residues obtained by leaching the glass from which the sample was loaded into the mass spectrometer were again measured at $(200 \pm 15) \times 10^{-9}$ in excellent agreement with the earlier measurement. The fraction of a small portion of 12070,91 from which all of the finest fragments had been removed had a $^{236}$U:$^{238}$U ratio of only $(31 \pm 6) \times 10^{-9}$ (1). Sample 12070,3, the sweepings of the box in which 12070 was processed, gave an poor analysis: $^{236}$U:$^{238}$U = $(24 \pm 5) \times 10^{-9}$. No good explanation for these results exists: there may be a correlation between high $^{235}$U:$^{238}$U values and the finest particles, but this hypotheses does not have much experimental backing. A very high $^{237}$Np (reported in reference (1)) in the original 12070,91 provides support for a cosmic ray irradiation that produced both nuclides.

Earlier work has led to a postulated large galactic cosmic-ray-induced neutron flux 10-70 million years ago. Such a flux could have produced perceptable amounts of 108 yr $^{146}$Sm in selected lunar samples. An attempt to find this nuclide is incomplete.

References
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7. $^{26}\text{Al}$ count kindly provided by J. S. Eldridge, 1973.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$^{236}\text{U}/^{238}\text{U}$</th>
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<tbody>
<tr>
<td>15505,25$^1$</td>
<td>Rock, St. 9 crater ejects (23 ± 3) x $10^{-9}$</td>
</tr>
<tr>
<td>15080,2$^1$</td>
<td>Fines Station 2 (122 ± 20) x $10^{-9}$</td>
</tr>
<tr>
<td>14318,26$^1$</td>
<td>Rock, point H (5 ± 2) x $10^{-9}$</td>
</tr>
<tr>
<td>15515,13$^1$</td>
<td>Breccia (76 ± 6) x $10^{-9}$</td>
</tr>
<tr>
<td>12022$^2$-OP-1</td>
<td>Rock, 0-1 mm from top (15 ± 4) x $10^{-9}$</td>
</tr>
<tr>
<td>12002$^2$-OP-5</td>
<td>Rock, 9-20 mm from top (25 ± 5) x $10^{-9}$</td>
</tr>
<tr>
<td>12002$^2$-OP-6</td>
<td>Rock, 20-60 mm from top (7 ± 2) x $10^{-9}$</td>
</tr>
<tr>
<td>12070,136</td>
<td>course &quot;fines&quot; (24 ± 5) x $10^{-9}$</td>
</tr>
<tr>
<td>12070,91,2</td>
<td>rerun (200 ± 15) x $10^{-9}$</td>
</tr>
</tbody>
</table>

$^1$Purified uranium from Rosholt (reference 8).
$^2$Uranium-rich fraction from Finkel (reference 5).