THE REACTION PRODUCTS OF LUNAR URANIUM AND COSMIC RAYS*
P. R. Fields, H. Diamond, D. N. Metta, and D. J. Rokop, Chemistry Division, Argonne National Laboratory, Argonne, Illinois 60439.

Radioactive and stable nuclei, formed during cosmic ray irradiation have been extensively studied, and the results analyzed in terms of complex models of the variation of flux with depth, time, temperature and lunar composition.\(^{(1,2)}\) An earlier paper\(^{(3)}\) discussed a sample with very high \(^{236}\)\(^{\text{U}}\) and \(^{237}\)\(^{\text{Np}}\) contents that must have come from solar cosmic proton irradiation of \(^{238}\)\(^{\text{U}}\) and that required more proton flux than was used to explain \(^{238}\)\(^{\text{Pu}}\) activity. This paper reports tentative explanations of the incidence of \(^{236}\)\(^{\text{U}}\). A histogram shows a grouping of \(^{236}\)\(^{\text{U}}\):\(^{238}\)\(^{\text{U}}\) values around \(5 \times 10^{-9}\) and an estimate of the lunar neutron flux is derived from this grouping. Further measurements of \(^{237}\)\(^{\text{Np}},^{238}\)\(^{\text{Pu}},^{239}\)\(^{\text{Pu}}\) and \(^{244}\)\(^{\text{Pu}}\) in lunar samples are reported.

Experimental and Results

Table 1 gives the results of \(^{236}\)\(^{\text{U}},^{237}\)\(^{\text{Np}},^{239}\)\(^{\text{Pu}}\) and \(^{244}\)\(^{\text{Pu}}\) measurements not reported in reference 3. Sample 14259,132 was requested because of its exceptionally high \(^{26}\)\(^{\text{Al}}\) content \((222 \pm 9\) dpm/kg\(^{(4)}\)). Duplicate measurements in this sample show satisfactory agreement in the unexpectedly low \(^{236}\)\(^{\text{U}}\):\(^{238}\)\(^{\text{U}}\) ratios and in limits to \(^{237}\)\(^{\text{Np}}\) content.

In an attempt to verify the high \((2.33 \times 10^{-7}),^{236}\)\(^{\text{U}}\):\(^{238}\)\(^{\text{U}}\) ratio of 12070,91\(^{(5)}\), another sample 12070,3, derived from sweepings of a hood floor at LRL after 12070 operations, was examined. In this sample, the \(^{236}\)\(^{\text{U}}\):\(^{238}\)\(^{\text{U}}\) ratio was less than one-half that of 12070,91. In addition, the absence of \(^{237}\)\(^{\text{Np}}\) set a 20-fold lower limit for this isotope than had been clearly observed in 12070,91. An attempt to measure the \(^{26}\)\(^{\text{Al}}\) activity in the residue of 12070,91 is in progress.

One of the portions of 14259,132 was processed without adding any isotopic diluents until the plutonium had been separated. The alpha activity from this plutonium fraction was examined for a month, and a limit of \(<2 \times 10^{-16}\) grams of \(^{238}\)\(^{\text{Pu}}\) (87.4 yr) per gram of sample was set.

Discussion

Lunar Neutron Flux

Fig. 1 illustrates that many of the available \(^{236}\)\(^{\text{U}}\):\(^{238}\)\(^{\text{U}}\) ratios cluster between 3 and 10 \(\times 10^{-9}\). This suggests that these low \(^{236}\)\(^{\text{U}}\) contents result principally from \(^{235}\)\(^{\text{U}}(n,\gamma)\)^{236}\(^{\text{U}}\). The neutron reactions are much less sensitive to depth than are charged particle reactions and hence, much less sensitive to the gardening histories of samples. If this were true, we can estimate a lower limit to the average thermal neutron flux experienced at a poorly defined average gardening depth, assuming a 106 barn capture cross section for \(^{235}\)\(^{\text{U}}\) and a steady neutron irradiation for a period extending back for many times the 2.34 \(\times 10^{7}\)

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yr half-life of $^{236}U$. The mean ratio $^{236}U:^{238}U = 5 \times 10^{-9}$ then leads to a lunar thermal neutron flux of 6 neutrons/(cm$^2$.sec). This is equivalent to a capture rate of $3 \times 10^{-5}$ captures per $^{235}U$ atom per aeon, which is 5-fold higher than the maximum number of captures (at OK) for a 100 barn $1/\nu$ absorber calculated by Lingenfelter et al. (2) It must be emphasized that the identification of the grouping of $^{236}U:^{238}U$ ratios with neutron captures is not well established. However, there are no neutron capture products which can be unambiguously related to the lunar thermal neutron flux.

We can interpret our 14259,132 $^{236}U:^{238}U$ ratio to imply a flux of 3 thermal neutrons/(cm$^2$.sec). Combining this with the nyt = $1.22 \times 10^{16}$ n/cm$^2$ determined by Lugmair and Marti (5) from $^{158}Gd:^{157}Gd$ ratios gives a thermal neutron exposure age of $1.2 \times 10^8$ yr for 14259.

Thermal neutrons produce 5.4 times as many fissions in $^{235}U$ as they do captures. This means that there are 11.5 thermal-neutron induced fissions for each lunar uranium spontaneous fission. This comprises a lower limit to any estimate of induced fissions because all other mechanisms for $^{236}U$ production are accompanied by several orders of magnitude more fissions per non-fission nuclear reaction. We see in figure 1 that $^{236}U$ is widely distributed in lunar samples, so we expect induced fissions to be the most important source of fission tracks.

The absence of $2.439 \times 10^4$ yr $^{239}Pu$ in lunar samples set limits to the average thermal neutron flux within the last 50,000 years. These limits in neutrons/(cm$^2$.sec) are: <100 for 15021,79; <56 for 15271,51; <26 for 14259,132; <130 for 61221,22; <270 for 69941,18; and <70 for both 61241,10 and 69961,19.

Mechanisms

One might use the grouping evident in figure 1 to distinguish between those samples exposed to solar cosmic rays within the top cm of the moon and those that remained buried most of the time. Charged particle production of $^{236}U$ should find comparable data in the track records, $^{26}Al$, $^{53}Mn$, $^{36}Cl$ and $^{237}Np$. Of these $^{26}Al$ has the best documentation, but no correlation is apparent. Similarly, there is no evidence supporting the suggestion in reference 2 that there should be a negative correlation between the iron and titanium content and the number of neutron captures.

REFERENCES

(3) FIELDS P. R., DIAMOND, H., METTA D. N., and ROKOP D. J. Proc. 3rd Lunar Sci. Conf. Vol. 2, pp. 1637-1644,
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MIT Press (1972)


Table 1. $^{236}$U, $^{237}$Np, $^{239}$Pu and $^{244}$Pu in lunar samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>$^{236}$U, $^{238}$U ($\times 10^{-9}$)</th>
<th>$^{237}$Np ($\times 10^{-15}$)</th>
<th>$^{239}$Pu (b) ($\times 10^{-16}$)</th>
<th>$^{244}$Pu (b) ($\times 10^{-16}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>15021,79</td>
<td>7.6±1.0 (b)</td>
<td>(8±3)×$10^{-15}$</td>
<td>&lt;4×$10^{-15}$</td>
<td>&lt;6×$10^{-15}$</td>
</tr>
<tr>
<td>15271,51</td>
<td>7.4±1.0 (b)</td>
<td>&lt;2×$10^{-15}$</td>
<td>&lt;2×$10^{-15}$</td>
<td>&lt;3×$10^{-15}$</td>
</tr>
<tr>
<td>14259,132(I)</td>
<td>4.2±0.4 (b)</td>
<td>&lt;1×$10^{-15}$</td>
<td>&lt;3×$10^{-15}$</td>
<td>&lt;3×$10^{-15}$</td>
</tr>
<tr>
<td>14259,132(II)</td>
<td>3.5±0.4 (b)</td>
<td>&lt;1×$10^{-15}$</td>
<td>&lt;3×$10^{-15}$</td>
<td>&lt;3×$10^{-15}$</td>
</tr>
<tr>
<td>12070,3</td>
<td></td>
<td>&lt;5×$10^{-15}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Reagent Blank</td>
<td>1.3±0.7</td>
<td>(4.5±0.5)×$10^{-15}$</td>
<td>(1.5±0.5)×$10^{-15}$</td>
<td>&lt;1×$10^{-15}$</td>
</tr>
<tr>
<td>(for above samples)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>61221,22</td>
<td>&lt;15 (c)</td>
<td>&lt;2×$10^{-15}$</td>
<td>&lt;2×$10^{-15}$</td>
<td>&lt;1.1×$10^{-15}$</td>
</tr>
<tr>
<td>69941,18</td>
<td>&lt;15 (c)</td>
<td>&lt;2×$10^{-15}$</td>
<td>&lt;2×$10^{-15}$</td>
<td>&lt;6×$10^{-15}$</td>
</tr>
<tr>
<td>61241,20</td>
<td>&lt;20 (c)</td>
<td>&lt;2×$10^{-15}$</td>
<td>&lt;1×$10^{-15}$</td>
<td>&lt;1×$10^{-16}$</td>
</tr>
<tr>
<td>69961,19</td>
<td>&lt;15 (c)</td>
<td>&lt;2×$10^{-15}$</td>
<td>&lt;1×$10^{-15}$</td>
<td>&lt;1×$10^{-16}$</td>
</tr>
</tbody>
</table>

(a) other data in text and in reference 1; (b) no correction for reagent blank; (c) preliminary value.

Figure 1

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