$^{244}\text{Pu}$ abundances in meteorites (and lunar samples) potentially provide a radiochronometer for the incorporation of r-process nucleosynthetic material in the early solar system and for meteorite formation and evolution at times $\approx 4$ b.y. ago. However, $^{244}\text{Pu}$ radiochronology requires a suitable stable or long-lived reference element (or isotope) which does not chemically fractionate from plutonium during processes leading to meteorite formation. Candidates for $^{244}\text{Pu}$ normalization include U, Th and light REE such as Ce and Nd.

Most reported $^{244}\text{Pu}$ results have been normalized to uranium. However, Pu/U fractionation has been observed in at least three meteorites (1, 2, 3). Recently, Marti et al. (4) suggested that $^{244}\text{Pu}$ follows the light REE in Angra dos Reis and that $^{244}\text{Pu}/\text{Nd}$ ratios in an Allende inclusion and Juvinas basaltic achondrite are similar to those in Angra dos Reis.

While fractionation among Pu, U, Th and REE have not been extensively studied, they should be minimal in samples which contain nearly constant proportions of refractory elements. Thus the $^{244}\text{Pu}/^{238}\text{U}$ ratio of 0.015 measured in the chondrite St. Severin (5) has often been used as a 'solar system' value. Recently, it was proposed (6) that coarse-grained refractory inclusions in the Allende meteorite, which contain on average relatively uniform refractory element enrichments over CI chondrites, might also represent the solar system $^{244}\text{Pu}/\text{U}$ ratio. A reported $^{244}\text{Pu}/^{238}\text{U}$ ratio of 0.016 in one of these inclusions (7) tentatively supports this view.

In view of the importance of coarse-grained inclusions in $^{244}\text{Pu}$ studies and the suggestion that $^{244}\text{Pu}/\text{REE}$ ratios may be useful for radiochronology, we report in this paper $^{244}\text{Pu}$, U, Ce and Nd abundances for refractory inclusions from the Allende meteorite.

$^{244}\text{Pu}/^{238}\text{U}$ ratios derived from fission track analyses in individual grains from six coarse-grained inclusions are summarized in Figures 1 and 2. The first three inclusions listed in the legend of each figure are described in detail by Podosek et al. (8); the last three inclusions listed are those originally described by Gray et al. (9). All inclusions are type B in the classification of Grossman (10).

The zero-order trend seen in Figures 1 and 2 is, roughly, a slope of -1, which indicates $^{244}\text{Pu}$ abundances are less variable than uranium abundances. $^{244}\text{Pu}/^{238}\text{U}$ ratios are not constant for a given mineral phase from one inclusion to another. In 4656-11, D7 and C1, significant grain-to-grain $^{244}\text{Pu}/^{238}\text{U}$ variations are present for grains of the same mineral. In all six inclusions, average $^{244}\text{Pu}/^{238}\text{U}$ ratios in melilite are different from those in clinopyroxene.

It is unlikely the non-uniform $^{244}\text{Pu}/^{238}\text{U}$ ratios resulted from different formation times, especially for grains from the same inclusion. Neither can differences in the track retention and etching properties of melilite and clinopyroxene explain the non-uniformities (2). It thus appears that $^{244}\text{Pu}$ and uranium were chemically fractionated during the solidification of melilite and clinopyroxene in these inclusions.
In three of these inclusions, Shirck (2) found half the uranium was not in melilitite and clinopyroxene. Therefore, track measurements cannot provide $^{244}\text{Pu}/^{238}\text{U}$ ratios for whole inclusions and it is still possible that $^{244}\text{Pu}/^{238}\text{U}$ is constant on the average in each inclusion.

In Table 1 we summarize available $^{244}\text{Pu}$, U, Ce and Nd data from Allende inclusions and include data from St. Severin and Angra dos Reis for comparison. REE analyses for the inclusions, discussed by Podosek et al. (8), were performed by D. Blanchard at Johnson Space Center. $^{244}\text{Pu}$ abundances are based on xenon analyses, and the total uranium abundances were done by induced fission track analysis.

In the fine-grained (4656-II) and coarse-grained (3666-II) Allende inclusions, which have very different $^{244}\text{Pu}/^{238}\text{U}$ ratios, $^{244}\text{Pu}/\text{Ce}$ and $^{244}\text{Pu}/\text{Nd}$ are much more constant. The same is true for whitlockite and pyroxene from Angra dos Reis. However, $^{244}\text{Pu}/\text{Ce}$ and $^{244}\text{Pu}/\text{Nd}$ are much larger in St. Severin. It appears, $^{244}\text{Pu}$ may follow neodymium or cerium in some circumstances, but not always.

We suggest that $^{244}\text{Pu}$ geochemical behavior can best be evaluated by combined $^{244}\text{Pu}$, REE and U analyses on suites of samples. If $^{244}\text{Pu}/\text{U}$ is plotted versus Ce/U (or Nd/U) for all samples, $^{244}\text{Pu}$ geochemical behavior and abundances may possibly be inferred from the data. For example, a linear array of data points would suggest the samples incorporated refractory trace elements from a common source at the same point in time, Pu$^{3+}$ predominantly following cerium (or neodymium) and Pu$^{4+}$ predominantly following uranium. If Pu$^{3+}$ and Pu$^{4+}$ were completely partitioned, the slope of the array would be equal to $^{244}\text{Pu}^{3+}/\text{Ce}$ (or Nd) in the source material from which the samples formed. The intercept would represent $^{244}\text{Pu}^{4+}/\text{U}$. A non linear array would imply one or more of the above conditions was not satisfied.

The obvious samples for initial study are refractory inclusions from Allende which appear to represent early refractory condensates in the solar nebula. In the nebula, Podosek et al. (8) estimate Pu$^{3+}$/Pu$^{4+}$ = 0.5, which should lead to a measurable positive slope in the array of data if Pu$^{3+}$ followed cerium (or neodymium) and Pu$^{4+}$ followed uranium. At present, only the data in Table 1, two points, are available for these inclusions. A line connecting the corresponding points in a $^{244}\text{Pu}/\text{U}$ vs Ce/U (or Nd/U) plot would have a positive slope, but clearly more data are needed. Analyses of this type may eventually provide useful $^{244}\text{Pu}$ normalization even when no single element normalization is possible.

REFERENCES

(2) Shirck, J. (1975), Ph.D. Thesis, Washington University, St. Louis, MO.
(7) Drozd, R. J., Morgan, C. J., Podosek, F. A. Poupeau, G., Shirck, J. R. and

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Fig. 1. 244Pu/238U vs. U from fission track analyses in melilitte from Allende inclusions.

Fig. 2. 244Pu/238U vs. U from fission track analyses in clinopyroxene from Allende inclusions.

TABLE 1: METEORITIC ABUNDANCE RATIOS OF 244Pu TO CE, Nd AND U.

<table>
<thead>
<tr>
<th>SAMPLE</th>
<th>244Pu/Ce</th>
<th>244Pu/Nd</th>
<th>244Pu/U</th>
</tr>
</thead>
<tbody>
<tr>
<td>4656-13a</td>
<td>1.1x10^-4</td>
<td>1.4x10^-4</td>
<td>&gt; .14</td>
</tr>
<tr>
<td>3666-11a</td>
<td>1.3x10^-4</td>
<td>1.9x10^-4</td>
<td>.016</td>
</tr>
<tr>
<td>St. Severinb</td>
<td>5.1x10^-4</td>
<td>7.3x10^-4</td>
<td>.015</td>
</tr>
<tr>
<td>Angra dos Reis c</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Phosphate</td>
<td>1.5x10^-4</td>
<td>.0001</td>
<td></td>
</tr>
<tr>
<td>Pyroxene</td>
<td>1.5x10^-4</td>
<td>.006</td>
<td></td>
</tr>
</tbody>
</table>

aPu, Ce, Nd and U, this paper
bPu (5), Ce and Nd (11), U (12)
cPu and Nd (13), U (14)