NOTE

Correlated calcium and titanium isotopic anomalies in Allende inclusions

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The isotopic anomalies recently observed in Ca(Z=20) and Ti(Z=22) in the Allende inclusions are related to each other and can be explained as due to a combined effect of mass-fractionation and cosmic-ray irradiation processes which occurred during the early history of the solar system.

INTRODUCTION

Isotopic anomalies have recently been reported in Ca (Lee et al., 1978) and Ti (Heydegger et al., 1979) in Allende mineral inclusions. Lee et al. (1978) have reported that the Ca anomalies did not appear to be readily explained by a simple model, but suggested that the observed anomaly implied isotopic heterogeneity on a scale of 10 to 10² km within the solar nebula. Kuroda (1979a,b) has recently attempted to explain the Ca anomalies in terms of the alteration of the isotopic ratios due to the process of neutron-capture, but the results were inconclusive. The purpose of this NOTE is to point out that the Ca and Ti isotopic anomalies are related to each other and can be explained as due to a combined effect of mass-fractionation and cosmic-ray irradiation processes which occurred during an ‘early’ irradiation period of the solar system.

RESULTS AND DISCUSSION

Table 2 compares the values of δi calculated for the Ca extracted from the Allende inclusions (Lee et al., 1978) with those for the normal Ca exposed to 15KeV neutrons. The data are expressed here in such a manner that δ40 = 0 and δ48 = 0. Lee et al. (1978) reported that their calcium data, when corrected for mass fractionation by using ⁴⁰Ca/²⁴Ca as a standard, showed nonlinear isotopic effects in ⁴⁰Ca of +13.5 per mil and in ⁴²Ca of +1.7 per mil for EK1-4-1. The second sample (C1) showed a ⁴⁰Ca depletion of about -2.9 per mil, but all other isotopes were normal. If the data are normalized as shown in Table 2, however, the largest anomalies are observed in EK1-4-1 and the δi values are all negative. This means that ⁴²Ca, ⁴⁴Ca, ⁴⁴Ca and ⁴⁶Ca are depleted in the Allende inclusions relative to the normal Ca.

Table 2 shows that a neutron flux as high as 1 × 10²³ (n/cm²) is required in order to enhance the abundance of ⁴³Ca, for example, by 4.2 parts per 10³. The neutron flux which has to be assumed here is by a factor of 10² to 10³ greater than the fluxes used in explaining the Gd, Sm and Nd isotopic anomalies (Kuroda, 1979c,d,e). Moreover, an exposure of normal Ca to 15 KeV neutrons does not result in significant alterations of the relative abundances of...
Table 1. Relative abundances \((N)\) of the isotopes of \(\text{Ca}\) and their neutron-capture cross-sections

<table>
<thead>
<tr>
<th>(\text{Ca}^{40})</th>
<th>(\text{Ca}^{41})</th>
<th>(\text{Ca}^{42})</th>
<th>(\text{Ca}^{43})</th>
<th>(\text{Ca}^{44})</th>
<th>(\text{Ca}^{46})</th>
<th>(\text{Ca}^{48})</th>
</tr>
</thead>
<tbody>
<tr>
<td>(N)</td>
<td>1.00000</td>
<td>0.0004618</td>
<td>0.0066212</td>
<td>0.0013753</td>
<td>0.0212076</td>
<td>0.0000322</td>
</tr>
<tr>
<td>(\sigma(15\text{KeV})(\text{mb}))</td>
<td>4</td>
<td>25</td>
<td>12</td>
<td>20</td>
<td>8</td>
<td>4</td>
</tr>
</tbody>
</table>

Table 2. Isotopic composition of \(\text{Ca}\) in the Allende inclusions

<table>
<thead>
<tr>
<th>Sample</th>
<th>(\delta 40)</th>
<th>(\delta 42)</th>
<th>(\delta 43)</th>
<th>(\delta 44)</th>
<th>(\delta 46)</th>
<th>(\delta 48)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1) EK1-4-1 PYX-A</td>
<td>(\Xi 0)</td>
<td>(-1.6 \pm 0.3)</td>
<td>(-4.1 \pm 0.6)</td>
<td>(-6.8 \pm 0.3)</td>
<td>(-23 \pm 13)</td>
<td>(\Xi 0)</td>
</tr>
<tr>
<td>(2) CI-Si</td>
<td>(\Xi 0)</td>
<td>(+0.5 \pm 0.3)</td>
<td>(+0.8 \pm 0.5)</td>
<td>(+1.4 \pm 0.1)</td>
<td>(-11 \pm 7)</td>
<td>(\Xi 0)</td>
</tr>
<tr>
<td>(3) WA-AN</td>
<td>(\Xi 0)</td>
<td>(-0.2 \pm 0.2)</td>
<td>(-0.4 \pm 0.5)</td>
<td>(+0.2 \pm 0.2)</td>
<td>(-0.4 \pm 13)</td>
<td>(\Xi 0)</td>
</tr>
<tr>
<td>(4) Normal (\text{Ca}) exposed to 15 KeV neutrons (\text{MeV}^{-1})</td>
<td>(\Xi 0)</td>
<td>(+1.0)</td>
<td>(+4.2)</td>
<td>(-0.3)</td>
<td>0.0</td>
<td>0.0</td>
</tr>
</tbody>
</table>

\((a)\) The total neutron flux was assumed to be such that the abundance of \(\text{Ca}^{40}\) became depleted by 4.0 parts per \(10^3\). This corresponds to a total flux of \(1 \times 10^{23}\) neutrons per \(\text{cm}^2\).

\(\text{Ca}^{44}, \text{Ca}^{46}\) and \(\text{Ca}^{48}\).

The 15 KeV neutron-capture cross-sections of the \(\text{Ca}\) isotopes are generally quite small ranging from 4 to 20 mb, as shown in Table 1. Hence it is likely that the neutron-capture process plays a minor role in altering the isotopic ratios. The fact that the values of \(\delta 42, \delta 43, \delta 44\) and \(\delta 46\) are all negative for the Allende inclusions EK1-4-1 may thus be attributed to the addition of spallogenic or cosmic-ray-produced \(\text{Ca}\) isotopes to the solar system materials during an early irradiation period. If the Allende inclusions were exposed to a smaller cosmic-ray flux than the average solar system materials, the cosmic-ray-produced \(\text{Ca}\) isotopes may be deficient in the Allende inclusions relative to the normal \(\text{Ca}\).

Kuroda (1979c) assumed that the average earth material was exposed to a greater time-integrated neutron flux than the Allende inclusions during an early irradiation period and the difference in the neutron flux amounted to \(1 \times 10^{20}\) \(\text{n/cm}^2\). He also estimated that the neutron flux was equivalent to an exposure to a total cosmic-ray flux which is equal to that observed on the present day earth for a period of \(1.22 \times 10^{12}\) years. The amount of cosmic-ray-produced \(\text{Ca}\) isotopes accumulated in the average earth material during an early irradiation period can not be calculated rigorously, since the cosmic-ray production rates for the \(\text{Ca}\) isotopes are not known. It may be possible, however, to carry out an order-of-magnitude calculation of the amount of cosmic-ray-produced \(\text{Ca}\) isotopes from the known cosmic-ray production rate of \(\text{Ar}^{38}\) in meteorites.

Bogard and Cressy (1973) reported that the cosmic-ray production rate of \(\text{Ar}^{38}\) in various chondrites to be about \(6 \times 10^{-10}\) cc STP per gram per \(10^6\) years. They also showed that the relative contributions from various elements in the meteorites can be calculated from the equation

\[
(\text{Ar}^{38})_c = k \cdot \left\{ 2.60(\text{K}) + 0.368(\text{Ca}) \\
+ 0.081(\text{Ti} + \text{Cr} + \text{Mn}) \\
+ 0.0213(\text{Fe} + \text{Ni}) \right\}
\]  

(1)

where \(k\) is the reciprocal of the exposure age of the Bruderheim meteorite.

The amount of \((\text{Ar}^{38})_c\) produced by the elements heavier than \(\text{Ca}\) can be calculated from equation (1) to be about 3 percent of the total or \(2 \times 10^{-11}\) cc STP per gram per \(10^6\) years. If it is assumed that the production rate of \(\text{Ca}^{44}\), for example, is equal to that of \(\text{Ar}^{38}\), the amount of \((\text{Ca}^{44})_c\) which is expected to be accumulated in the meteorites can be calculated to be \(3.8 \times 10^{-14}\) gram per gram per \(10^6\) years. The amount of \((\text{Ca}^{44})_c\) produced during a period of \(1.22 \times 10^{12}\) years then equals to \(4.6 \times 10^{-8}\) (g/g).
The average Ca content of all meteorites is about 1.1 percent (RANKAMA and SAHAMA, 1950) and hence the amount of $^{42}$Ca originally present in the meteorites is $7.15 \times 10^{-5}$ (g/g). Thus the abundance of $^{42}$Ca in the average solar system material should be enhanced by

$$\frac{4.6 \times 10^{-6}}{7.15 \times 10^{-5}} = 0.64 \times 10^{-3}$$

or 0.64 per mil relative to the Allende inclusions. This corresponds to a value of $(\delta^{42}\text{Ca}) = -0.64$ (per mil), which agrees within a factor of 2 to 3 with the observed value of $(\delta^{42}\text{Ca}) = -1.6 \pm 0.3$ (per mil) for the Allende inclusion EK1-4-1 PYX-A shown in Table 2. The agreement between the calculated and observed values should perhaps be considered as remarkable in view of the many uncertainties involved in these calculations.

Heydegger et al. (1979) have recently reported that some inclusion materials from the Allende meteorite had a statistically significant enhancement of the order of one part per mil in the 50/49 ratio, probably due to a nucleogenic anomaly in $^{50}$Ti abundance. It is quite likely, however, that the isotopic anomalies observed in Ca(Z=20) and Ti(Z=22) are similar in nature. The 15KeV neutron-capture cross-sections of $^{46}$Ti, $^{47}$Ti, $^{48}$Ti, $^{49}$Ti and $^{50}$Ti are 10, 12, 6, 4, 8 and 4 (millibarns), respectively (Burridge et al., 1957), and they are similar to the cross-section values of Ca isotopes shown in Table 1. Since the neutron-capture cross-sections of the Ti isotopes are generally quite small, it is likely that the spallation reactions play a dominant role in producing the Ti anomalies, just as in the case of the Ca isotopes.

It is therefore interesting to carry out an order-of-magnitude calculation of the amount of spallogenic or cosmic-ray-produced Ti isotopes expected to be accumulated in the average solar system material during the early irradiation period.

The average Ti, Cr and Mn contents of all meteorites are 0.08, 0.12 and 0.15 percent, respectively (RANKAMA and SAHAMA, 1950), and hence the amount of $(^{38}\text{Ar})_c$ produced by the elements heavier than Ti can be calculated from equation (1) to be about 2.7 percent of the value of $6 \times 10^{-16}$ cc STP per gram per 10$^6$ years or $1.6 \times 10^{-11}$ cc STP per gram per 10$^6$ years.

The cosmic-ray production ratio for the Ti isotopes is unknown, but let us assume for the time being that the heaviest isotope $^{50}$Ti is not affected, whereas the relative abundance of one of the lighter isotopes, for example, $^{49}$Ti is altered significantly by the addition of the cosmic-ray-produced component. If it is further assumed that the production rate of $^{49}$Ti is equal to that of $(^{38}\text{Ar})_c$, from the elements heavier than Ti, we have a value of $3.5 \times 10^{-14}$ gram per gram per 10$^6$ years for the cosmic-ray production rate of $^{49}$Ti. The amount of $(^{49}\text{Ti})_c$ produced during a period of $1.22 \times 10^{12}$ years then equals $3.5 \times 10^{-8}$ (g/g).

The average Ti content of all meteorites is 0.08 percent (RANKAMA and SAHAMA, 1950) and hence the amount of $^{49}$Ti originally present in the meteorites is $4.4 \times 10^{-5}$ (g/g). Thus the abundance of $^{49}$Ti in the average solar system material should be enhanced by

$$\frac{3.5 \times 10^{-8}}{4.4 \times 10^{-5}} = 0.8 \times 10^{-3}$$

or 0.8 per mil relative to the Allende inclusions, in good agreement with the finding of Heydegger et al. (1979) that some inclusion materials from the Allende meteorite had a statistically significant enhancement of the order of one part per mil in the 50/49 ratio.

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References


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