NOTE

Isotopic compositions of calcium, barium, neodymium and xenon in meteorites

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Isotopic anomalies observed in meteorites for Ca, Ba, Nd and Xe are explained as due to the alteration of the isotopic ratios by a combined effect of mass fractionation, neutron-capture and cosmic-ray irradiation processes, which took place prior to and during the period of formation of the solar system. The neutron-capture process appears to have occurred at a temperature higher than that of the earth's surface.

INTRODUCTION

It has recently been pointed out that the isotopic compositions of Ca, Ba and Nd in the Allende inclusion EK 1-4-1 resemble those of the terrestrial Ca, Ba and Nd exposed to a neutron flux (KURODA, 1979). The thermal neutron-capture cross-section values were used in the previous report to calculate the variations of the isotopic ratios. It appears, however, that much improved agreements between the calculated and observed isotopic ratios can be obtained if the 15KeV cross-sections values for the neutron-capture processes are used. The results from the calculations for the isotopic anomalies observed for Ca, Xe, Ba and Nd are reported here.

of anomalous neodymium found in Allende EK 1-4-1 by McCULLOCH and WASSERBURG (1978) and normal neodymium exposed to a neutron flux. The values of 15KeV neutron-capture cross-sections given by BURBIDGE *et al.* (1957) are used here. This does not mean that the neutron-capture processes occurred at temperatures as high as 10^8 K. The 15KeV values are used here, because the cross-section values for the temperatures at which the neutron captures took place are unknown. The agreement between the calculated and observed values appears to be satisfactory. There is a small excess of ^{143}Nd in Allende EK 1-4-1, which can be explained as due to the alpha-decay of 1.07×10^{11} -year ^{147}Sm .

Table 2 compares the isotopic composition

RESULTS AND DISCUSSION

Table 1 compares the isotopic compositions

Table 1. Isotopic composition of neodymium in Allende EK 1-4-1^(a)

Sample	ϵ^{142}	ϵ^{143}	ϵ^{144}	ϵ^{145}	ϵ^{146}	ϵ^{148}	ϵ^{150}
(1) EK 1-4-1 SCa	-9.6 ± 0.3	$+24.1 \pm 0.4$	$\equiv 0$	$+2.7 \pm 0.6$	-12.4 ± 0.4	-1.5 ± 0.5	-9.6 ± 0.3
(2) EK 1-4-1 SCb	-9.9 ± 0.3	$+23.5 \pm 0.4$	$\equiv 0$	$+2.4 \pm 0.4$	-12.5 ± 0.4	-1.5 ± 0.9	-9.9 ± 0.3
(3) EK 1-4-1 MEL	-9.2 ± 0.4	-0.1 ± 0.7	$\equiv 0$	$+2.3 \pm 0.9$	-12.9 ± 0.5	-0.5 ± 1.0	-9.2 ± 0.4
(4) Normal Nd exposed to a neutron flux ^(b)	-0.81	-7.4	$\equiv 0$	+19.2	-0.54	-9.2	-9.3

(a) The data reported by McCULLOCH and WASSERBURG (1978). The differences in the isotopic ratios are expressed in parts per 10^4 .

(b) Calculated values based on the 15KeV neutron-capture cross-sections (BURBIDGE *et al.*, 1957).

Table 2. Isotopic composition of barium in Allende EK 1-4-1^(a)

	Sample	ε130	ε132	ε134	ε135	ε136	ε137	ε138
(1)	EK 1-4-1 SCa	-34.0 ± 7.0	-28.0 ± 14.0	-17.8 ± 1.2	≡ 0	-9.7 ± 0.6	+7.9 ± 0.4	≡ 0
(2)	Normal Ba exposed to a neutron flux ^(b)	-10.8	-11.3	-11.2	-33.1	+20.1	-27.5	≡ 0

(a) The data reported by McCULLOCH and WASSERBURG (1978). The differences in the isotopic ratios are expressed in parts per 10⁴.

(b) Calculated values based on the 15KeV neutron-capture cross-sections (BURBIDGE *et al.*, 1957).

of anomalous barium found in Allende EK 1-4-1 by McCULLOCH and WASSERBURG (1978) and normal barium exposed to a neutron flux. It appears that small excesses of ¹³⁵Ba and ¹³⁷Ba are present in Allende EK 1-4-1, as it was mentioned in the previous report (KURODA, 1979). These excesses may be the products of the neutron-capture reactions

and

Table 3 compares the isotopic compositions of anomalous calcium found in Allende EK 1-4-1 by LEE *et al.* (1978) and normal calcium exposed to a neutron flux. It is interesting to note here that there is a small excess of ⁴²Ca in Allende EK 1-4-1, which may be due to the contribution from the reaction

The cosmic abundances (N) of ⁴¹K and ⁴⁴Ca are 219 and 1040 (relative to Si = 10⁶), respec-

tively (BURBIDGE *et al.*, 1957). In the calculations shown in Table 3, it was assumed that the total neutron flux was such that the abundance of ⁴⁴Ca was depleted by 0.80 per mil due to the ⁴⁴Ca(n, γ)⁴⁵Ca reaction. The 15KeV cross-sections (σ) for ⁴¹K and ⁴⁴Ca are 25mb and 8 mb, respectively. The ratio of Nσ values for ⁴¹K and ⁴⁴Ca is then

$$\frac{(219)(25)}{(1040)(8)} = 0.6581.$$

If the amount of excess ⁴²Ca corresponding to the above ratio is added to the ⁴²Ca in the neutron-irradiated Ca in Table 3, a value of δ42 = +1.2 (per mil) is obtained. The enrichment of ⁴²Ca thus calculated is in good agreement with the observed value δ42 = +1.5 ± 0.3 (per mil) reported by LEE *et al.* (1978).

Table 4 compares the isotopic compositions of the anomalous xenon in the Murray meteorite (KURODA *et al.*, 1974) and the atmospheric xenon exposed to a neutron flux. Note that the agreement between the calculated and observed values is not at all satisfactory, except for the abundance at mass number 130. A vastly improved agreement between the calculated and observed values can be obtained, however, if a comparison is made with a sample of atmospheric xenon mixed with iodine and tellurium in the ratio of their cosmic abundances and a correction is applied for a mass-dependent fractionation. The abundances of ¹²⁸Xe, ¹²⁹Xe

Table 3. Isotopic composition of calcium in Allende EK 1-4-1^(a)

	Sample	δ40	δ42	δ43	δ44	δ46	δ48
(1)	EK 1-4-1 PYX-A	≡ 0	+1.5 ± 0.3	+0.9 ± 0.5	≡ 0	-13 ± 13	+13.6 ± 0.5
(2)	Normal Ca exposed to a neutron flux ^(b)	+0.27	-0.53	+4.44	≡ 0	+0.26	+0.27

(a) The data reported by LEE *et al.* (1978). The differences in the isotopic ratios are expressed in parts per 10³.

(b) Calculated values based on the 15KeV neutron-capture cross-sections (BURBIDGE *et al.*, 1957).

Table 4. Isotopic composition of xenon in the Murray meteorite compared with that of the neutron-irradiated atmospheric xenon ^(a)

Sample	δ_{124}	δ_{126}	δ_{128}	δ_{129}	δ_{130}	δ_{131}	δ_{132}	δ_{134}	δ_{136}
(1) Murray	+396	+301	+193	+80.8	+101	+77.1	+29.7	+11.7	$\equiv 0$
(2) Atmospheric Xe exposed to a neutron flux ^(b)	-0.31	-3.6	-3.5	-13.7	+86.3	-13.3	+7.5	-3.6	$\equiv 0$

(a) The Murray data reported by KURODA *et al.* (1974). The differences in the isotopic ratios are expressed in parts per 10^3 .

(b) Calculated values based on the 15KeV neutron-capture cross-sections (BURBIDGE *et al.*, 1957).

Table 5. Relationship between the isotopic compositions of the anomalous xenon in the Murray meteorite and the atmospheric xenon ^(a)

Sample	δ_{124}	δ_{126}	δ_{128}	δ_{129}	δ_{130}	δ_{131}	δ_{132}	δ_{134}	δ_{136}
(1) Atmospheric Xe exposed to a neutron flux ^(b)	-0.31	-3.6	-3.5	-13.7	+86.3	-13.3	+7.5	-3.6	$\equiv 0$
(2) A mixture of Xe, I and Te in the cosmic-abundance ratios exposed to a neutron flux	-0.31	-3.6	+88.3	-11.5	+86.3	-10.4	+7.5	-3.6	$\equiv 0$
(3) Corrected for mass-fractionation ^(c)	+91.5	+72.9	+150	+44.1	+132	+27.9	+38.1	+11.7	$\equiv 0$
(4) Murray	+396	+301	+193	+80.8	+101	+77.1	+29.7	+11.7	$\equiv 0$

(a) The Murray data reported by KURODA *et al.* (1974). The differences in the isotopic ratios are expressed in parts per 10^3 .

(b) Calculated values based on the 15KeV neutron-capture cross-sections.

(c) It is assumed here that the difference between the δ_{134} values of irradiated atmospheric xenon (-3.6 per mil) and the xenon in Murray (+11.7 per mil) is due to the fact that the lighter isotopes in the atmospheric xenon are systematically mass-fractionated relative to the heavier isotopes by $(3.6 + 11.7)/2 = 7.65$ per mil per unit mass difference.

and ^{131}Xe are enhanced by the contributions from the reactions

and

The contribution from the neutron-capture reaction on ^{130}Ba is negligibly small in this case, because of the very small cosmic abundance of ^{130}Ba ($N = 0.00370$ relative to $\text{Si} = 10^6$).

The fact that the agreements between the calculated and observed values are still not very good for the δ values at mass numbers 124,

126, 129 and 131 can be accounted for as being due to the presence of excesses of the cosmic-ray-produced xenon isotopes at these mass numbers and also a small contribution from the decay of extinct nuclide ^{129}I at mass number 129 (KURODA *et al.*, 1974).

It is of special interest to make a rough estimate of the total neutron flux which is required to produce the isotopic anomalies observed in these meteorites. The thermal neutron-capture cross-section for ^{136}Xe is 150mb and the 15KeV value is 4.9mb. Let us assume that the neutron-capture processes have occurred at an intermediate neutron energy and the cross-section value at that temperature corresponded to the average of the two extreme values, namely, about 80mb. In the calculations shown in Table 5, the abundance of ^{136}Xe is depleted by 0.049 per mil relative to the original abundance. The total neutron flux (Φ), which is required to produce such an

alteration of the ^{136}Xe abundance is then

$$\Phi = \frac{0.000049}{80 \times 10^{-26}} = 6.1 \times 10^{20} (\text{n/cm}^2).$$

The value of total neutron flux thus calculated is similar to a value of 9.1×10^{20} (n/cm^2), which was obtained by a quite different method in the previous report (KURODA, 1979). It is also interesting to note here that these values are comparable to the neutron flux of the Oklo natural reactors. HAGEMANN *et al.* (1975) have estimated the total integrated neutron flux for the Oklo reactors to be from 8.1×10^{20} to 1.32×10^{21} (n/cm^2).

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REFERENCES

- BURBIDGE, E. M., BURBIDGE, G. R., FOWLER, W. A., and HOYLE, F. (1957) Synthesis of the elements in stars. *Rev. Mod. Phys.* **29**, 547–650.
- HAGEMANN, R., DEVILLERS, C., LUCAS, M., LECOMTE, T., RUFFENACH, J.-C. (1975) Estimation de la durée de la réaction. Limitation imposées par les données neutroniques. *Le Phenomene D'Oklo*, International Atomic Energy Agency, Vienna, 415–423.
- KURODA, P. K. (1979) Isotopic anomalies in the early solar system. *Geochem. J.* **13**, 83–90.
- KURODA, P. K., BECK, J. N., EFURD, D. W., and MILLER, D. K. (1974) Xenon isotope anomalies in the carbonaceous chondrite Murray. *J. Geophys. Res.* **79**, 3981–3992.
- LEE, T., PAPANASTASSIOU, D. A., and WASSERBURG, G. J. (1978) Calcium isotopic anomalies in the Allende meteorite. *Astrophys. J.* **220**, L21–L25.
- MCCULLOCH, M. T., and WASSERBURG, G. J. (1978) Barium and neodymium isotopic anomalies in the Allende meteorite. *Astrophys. J.* **220**, L15–L19.