Cosmogenic radionuclides in the recently fallen Kobe (CK4) meteorite

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Cosmogenic radionuclides in the Kobe chondrite (CK4) fell on September 26th, 1999, have been measured immediately after the fall (21 hours) using a ultra low-background Ge-detector. Nineteen cosmogenic nuclides including the very short-lived 24Na were detected and their activities place constrains on the exposure history of the meteorite and reflect effect of solar modulation of galactic cosmic rays during solar maximum. Two nuclides, 28Mg and 43K were possibly detected for the first time and signals probably due to 56Ni and 57Ni were present although statistical uncertainties were very large. On the other hand, low activities of 60Co (~1 dpm/kg) and 26Al (~38 dpm/kg) mainly suggest a small preatmospheric size (≤10 cm) of the Kobe meteorite.

INTRODUCTION

Cosmogenic radioactive and stable nuclides in chondrites have preserved important records of their exposure history during the last ten million years (e.g., Michel, 1999; Vogt et al., 1990; Caffee et al., 1988). For example, activity of typical neutron-capture product, 60Co has been used as an indicator of their preatmospheric size (e.g., Eberhardt et al., 1963; Spergel et al., 1986), and those of spallation products, 22Na, 54Mn and 26Al etc. reflect irradiation conditions such as shielding effect (e.g., Bhandari et al., 1993; Michel et al., 1995), exposure age (e.g., Heimann et al., 1974; Herpers and Englert, 1983) and flux of cosmic-rays (e.g., Evans et al., 1982).

Five meteorite falls (chondrites) were recovered in Japan during last decade. Measurements of γ-emitting radionuclides in these meteorites have been carried out by Shima et al. (1993) and Komura et al. (1996a, b) by non-destructive method using low-background Ge-detectors.

On September 26th (20:23 local time), 1999, 20 fragments of a meteorite (weighing 136 g in all) fell through the roof into a room located in Kita-ku, Kobe City, Hyogo Prefecture, Japan. This meteorite was internationally registered by one of authors (N.N.) as “the Kobe meteorite”. Based on petrographical and mineralogical examinations, the meteorite was classified as very rare Karoonda (Kallemeyn et al., 1991) type (CK4) (Nakamura et al., 2000a, b).

In this paper, we present the results obtained by non-destructive γ-ray measurements of the largest fragment of the Kobe meteorite (~65 g, fragment-A) and discuss its exposure history including the period just before the collision with the Earth.

GAMMA-RAY MEASUREMENT

The meteorite fragments were brought to Forensic Science Laboratory (FSL) of Hyogo Prefectural Police Headquarters on Sep. 27th, 1999. Major fragments were photographed, weighed and subjected to X-ray fluorescence analysis to deter-
mine their bulk chemical composition. After general inspection at FSL, the largest fragment (fragment-A, ~65 g) (Fig. 1) was transferred to the Ogoya Underground Laboratory of Low Level Radioactivity Laboratory (LLRL) of Kanazawa University at Komatsu, Ishikawa Prefecture, located ~300 km NE of Kobe to evaluate radioactivity induced by cosmic-ray interactions with the meteorite.

Since levels of the cosmogenic nuclides in the meteorites are extremely low (<1~100 dpm/kg: disintegration per minute per kg sample), the Kobe meteorite was measured using an ultra low background Ge-detector located in the underground laboratory (270 mwe: meters water equivalent of depth) in the tunnel of the former Ogoya Copper Mine (see details in Komura et al., 1996a). The Ge-detector (EURISYS EGPC 90-220-R) has relative efficiency of 93.5% and resolution of 2.0 keV at 1.33 MeV.

Gamma-ray spectrum was recorded starting 17:25 pm on September 27th (21 hours after the

Fig. 1. The largest fragment of the Kobe meteorite (fragment-A, ~65 g) examined in this study. This photograph was provided by courtesy of Forensic Science Laboratory.

Fig. 2. Gamma-ray spectrum of the Kobe meteorite measured 5500 minutes at Ogoya Underground Laboratory.
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Table 1. Activities of short-lived nuclides (\(^{24}\)Na, \(^{28}\)Mg, \(^{43}\)K and \(^{57}\)Ni), cosmogenic radionuclides and natural nuclides (corrected at the time of fall) in the Kobe meteorite

**Short-lived nuclides**

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>keV</th>
<th>Half-life (cpm)</th>
<th>(cpm)</th>
<th>(cpm)</th>
<th>(cpm)</th>
<th>Saturation activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{24})Na</td>
<td>1368</td>
<td>15.0 h</td>
<td>0.0205 ± 0.0045</td>
<td>0.0054 ± 0.0018</td>
<td>0.0035 ± 0.0011</td>
<td>27 ± 7</td>
</tr>
</tbody>
</table>
| \(^{28}\)Mg | 1342 | 20.9 h | 0.0049 ± 0.0022 | 0.0024 ± 0.0012 | 0.0021 ± 0.0008 | 13 ± 5 | 15 ± 7 | \n
**Cosmogenic radionuclides**

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>keV</th>
<th>Half-life (cpm)</th>
<th>(cpm)</th>
<th>(dpm/kg)</th>
<th>(dpm/kg)</th>
</tr>
</thead>
</table>
| \(^{44}\)Sc | 272 | 2.44 d | 0.0161 ± 0.0027 | 13 ± 2 | \n
**Natural (U, Th, K) nuclides**

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>keV</th>
<th>(concentration)</th>
<th>(concentration)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U</td>
<td>((^{235}Pa))</td>
<td>295</td>
<td>0.0229 ± 0.0039(^{8})</td>
</tr>
<tr>
<td>Th</td>
<td>((^{233}Pa))</td>
<td>911</td>
<td>0.0306 ± 0.0031(^{8})</td>
</tr>
<tr>
<td>K</td>
<td>((^{238}Ac))</td>
<td>1461</td>
<td>1.28*10(^{6}) y</td>
</tr>
</tbody>
</table>

Errors noted are 1σ.

\(^{5}\)Net counts.

\(^{8}\)Contribution of \(^{56}\)Ni was subtracted.

\(^{1}\)Bhandari et al. (1989); \(^{2}\)Yoneda, S. (personal communication); \(^{3}\)Komura et al. (1996a); \(^{4}\)Evans et al. (1982), Cressy, Jr. (1970), Shedlovsky et al. (1967) et al.; \(^{5}\)Mason (1979).
fall) to Oct. 1st 13:18 pm (5500 min.) with a 4K channel pulse height analyzer. The spectrum was recorded several times every day to evaluate activities of short-lived radionuclides with half-life ranging from less than a day to a few days.

RESULTS AND DISCUSSION

The $\gamma$-ray spectrum (5500 min.) of the Kobe meteorite obtained in this study is shown in Fig. 2 and the activities of cosmogenic nuclides at the time of fall (dpm/kg) are summarized in Table 1 together with those of other recent meteorites (Evans et al., 1982; Cressy, Jr., 1970; Shedlovsky et al., 1967 etc.). All values are decay corrected to the time of fall and the errors cited in Table 1 are 1σ statistical errors of $\gamma$-ray counting.

Nuclides detected

Natural $U$, $Th$ and $K$: Natural $U$, $Th$ and $K$ in the Kobe meteorite were determined to be $29 \pm 2$ ppb, $51 \pm 4$ ppb and $0.029 \pm 0.002\%$ by $\gamma$-ray peaks of $^{214}$Pb (295 and 352 keV) for $U$, $^{212}$Pb (238 keV) and $^{228}$Ac (911 keV) for $Th$ and $^{40}$K (1461 keV) (Table 1). The concentrations of $U$ and $Th$ are little higher and lower, respectively, compared with those of typical values of C3 chondrites (Mason, 1979). The $K$ value agrees with those of typical CK chondrites, as well as that of the Karoonda (CK) (~0.03%; Kallemeyn et al., 1991) and also with the value obtained for the Kobe fragment-E (Nakamura et al., 2000a) by EPMA analysis.

Cosmogenic nuclides: Despite the short counting time, 19 nuclides were detected by non-destructive $\gamma$-ray spectrometry including $^{24}$Na ($t_{1/2} = 15.0$ h), $^{28}$Mg (20.9 h), $^{43}$K (22.3 h), $^{44m}$Sc (2.44 d), $^{47}$Sc (3.35 d), $^{52}$Mn (5.59 d), $^{56}$Ni (5.9 d), $^{48}$V (16.8 d), $^{51}$Cr (27.8 d), $^{7}$Be (53.3 d), $^{58}$Co (70.8 d), $^{56}$Co (77.3 d), $^{46}$Sc (83.8 d), $^{57}$Co (272 d), $^{54}$Mn (312 d), $^{22}$Na (2.60 y), $^{60}$Co (5.26 y), $^{44}$Ti (60.4 y) and $^{26}$Al ($7.2 \times 10^5$ y), although many of them have large statistical error.

Long-lived nuclides with half-life longer than $10^5$ y ($^{10}$Be, $^{36}$Cl and $^{26}$Al) of the Kobe meteorite were determined by AMS (accelerator mass spectrometry) at Lawrence Livermore National Laboratory. Activity of $^{26}$Al (37.8 dpm/kg) obtained in this work by non-destructive $\gamma$-ray spectrometry agreed well with the value measured by AMS (fragments C-2 and C-3; ~38 dpm/kg: Caffee et al., 2000).

Evidences of $^{24}$Na, $^{28}$Mg and $^{43}$K

In this study, detection of short-lived nuclides was of particular interest because the $\gamma$-ray measurement was started just after 21 hours of the fall, which is the second quickest measurement (the Tsukuba meteorite was analyzed 9 hours after its fall). In the 1995 Neagari (L6) and the 1996 Tsukuba (H5-6) meteorites (Komura et al., 1996a, b; Yoneda et al., 1996), short-lived nuclides such as $^{48}$Sc, $^{44m}$Sc, $^{52}$Mn and $^{24}$Na were also detected. In the case of the Neagari meteorite, detection of $^{43}$K and $^{57}$Ni was ambiguous because of very poor statistics (Komura et al., 1996a).

Figure 3 shows the variation of 1300–1400 keV region of $\gamma$-ray spectrum together with background spectrum, where $\gamma$-ray peaks of $^{48}$V (1312 keV, 97.5%) (+$^{48}$Sc, 100%), $^{60}$Co (1332 keV, 100%), $^{28}$Mg (1342 keV, 52.6%), $^{24}$Na (1364 keV, 100%) and $^{57}$Ni (1378 keV, 81.7%) are expected to appear. As seen in the top spectrum (433 min.), 1312 keV and 1364 keV peaks are clearly seen and 1332 keV, 1342 keV and 1378 keV peaks are recognized in the second spectrum (1433 min.). Peak areas of 1312 keV and 1332 keV peaks increase monotonically with time, however, those of $^{28}$Mg, $^{24}$Na and $^{57}$Ni attained their saturation values because of most of short-lived atoms decay within 5500 minutes of counting. The 1328 keV, 1346 keV and 1351 keV peaks, which increase with counting time, could not be explained by radionuclides belonging to natural $U$, $Th$ and $Ac$ series and background peaks of the Ge-detector as seen in the bottom spectrum in Fig. 3.

Integral method was applied to evaluate the activities of short-lived nuclides such as $^{24}$Na, $^{28}$Mg and $^{43}$K (372 keV, 87%). In this method, peak area (net counts) was plotted as a function of counting time and increase of peak area was fitted to the theoretical calculation to estimate saturation value. Existence of $^{43}$K was suggested by
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this method although ambiguity was rather large. We considered that the $\gamma$-ray peak at 1378 keV seen in the second spectrum in Fig. 3 might have derived from $^{57}\text{Ni}$, however, observed peak area can also be explained by natural $^{214}\text{Bi}$ (3.9%). Therefore the inferred $^{57}\text{Ni}$ activity represents an upper limit.

**Constraints of exposure history**

Activity of cosmogenic nuclides in meteorites depends mainly on the preatmospheric size and shape, shielding condition (depth from the surface), elemental composition of meteorites, flux variation of cosmic-rays (11 years solar cycle; e.g., Evans et al., 1982; Bhandari et al., 1994) and exposure time and history. Preatmospheric size, shielding effect and the galactic cosmic-ray flux received by the Kobe meteorite in interplanetary space are discussed below based on $\gamma$-ray measurements.

**Shielding effects (preatmospheric size and depth):** $^{22}\text{Ne}/^{21}\text{Ne}$ ratio and activity of neutron-capture nuclide, $^{60}\text{Co}$, have been used as important indicators of the preatmospheric size of meteorites (Bhandari and Podar, 1982). For example, $^{60}\text{Co}$ activities of Allende (CV3) (53~260 dpm/kg, Rancitelli et al., 1969; Cressy, Jr., 1972) and Jilin (H5) (23~179 dpm/kg, Honda et al., 1980, 1982) with large preatmospheric sizes (recovered radius estimated, 70~80 cm) show remarkably high values and wide ranges. On the other hand, most chondrites show low values (<30 dpm/kg, typically <10 dpm/kg) (Inoue and Komura, 2001), suggesting that the preatmospheric size of these meteorites was small (radius <30 cm). The Kobe meteorite shows extremely low $^{60}\text{Co}$ activity (~1 dpm/kg), which suggests that preatmospheric size of this meteorite was even smaller (radius <~10 cm) or that it was located near the surface region of the meteoroid.

A large data base exists for spallation products (particularly $^{22}\text{Na}$, $^{54}\text{Mn}$ and $^{26}\text{Al}$) in meteorites, especially chondrites. Activities of most cosmogenic nuclides in the Kobe meteorite are generally lower compared to the values reported previously (Table 1). These low activities do not seem to be due to the variation in target element compositions of the Kobe meteorite, because its bulk composition is similar to other CV-CO group chondrites (Nakamura et al., 2000a). In the case of the Jilin meteorite, the low activities of the long-lived $^{26}\text{Al}$ (19~27 dpm/kg) were explained by its short exposure age (~0.5 $\times$ 10$^6$ y; Honda et al., 1980). On the other hand, the exposure age of the Kobe meteorite is reported to be long enough for $^{26}\text{Al}$ to have attained secular equilibrium with production although the estimates based on $^{21}\text{Ne}$, $^{38}\text{Ar}$ and $^{10}\text{Be}$-$^{21}\text{Ne}$ methods (22~56 $\times$ 10$^6$ y; Caffee et al., 2000) scatter widely. Therefore the low activity of $^{26}\text{Al}$ is mainly a consequence of its small preatmospheric size. The low activity of $^{60}\text{Co}$ of fragment-A is not inconsistent with this result. Depth profiles of some cosmogenic nuclides, e.g., $^{22}\text{Na}$, $^{10}\text{Be}$, $^{53}\text{Mn}$, $^{26}\text{Al}$ as a function of preatmospheric size and shielding depth (e.g., Bhandari et al., 1993; Murty et al., 1998) have

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Fig. 3. Gamma-ray spectra of the 1300~1400 keV region and background spectrum of the Ge-detector used in this work.
been extensively studied. The low activities of other spallation products (Table 1) could also be partly due to small preatmospheric size of the Kobe meteorite. The $^{22}$Ne/$^{21}$Ne ratio (1.09) of the Kobe meteorite is not consistent with small size of the meteoroid as indicated by its $^{60}$Co activity. This discrepancy can be explained if the meteoroid had a two stage exposure (Caffee et al., 2000).

**Effect of solar activity:** The flux of galactic cosmic-rays correlates inversely with the solar activity during a cycle of 11 years. Activities of short-lived radionuclides (especially $^{46}$Sc and $^{22}$Na) reflect the 11 years of solar cycle (Evans et al., 1982). In Fig. 4, the activities of spallation products in the Kobe meteorite are compared with unpublished data of the Tsukuba (H5-6) meteorite (the largest fragment No. 13, 177.5 g; Komura et al., 1996b: fragment No. 1, 62.4 g; $^{24}$Na, 155 ± 29 dpm/kg; personal communication, S. Yoneda). It is interesting that the activity ratios, $R = [\text{Kobe/Tsukuba}]$ of nuclides increase with their half-lives ($R = 0.2–0.9$) except for $^{44}$Ti ($t_{1/2} = 60.4$ y). The Kobe meteorite fell around the solar maximum and the Tsukuba meteorite fell near the solar minimum (on Jan. 8th, 1996). Especially, the low activity ratios of Kobe to Tsukuba ($< 0.7$) of $^{48}$V, $^{51}$Cr, $^{58}$Co, $^{56}$Co, $^{46}$Sc, $^{57}$Co and $^{54}$Mn (with half-life shorter than one year) and extremely low ratio of $^{24}$Na ($< 0.2$) ($t_{1/2} = 15$ h) indicate the influence of solar activity over 0.01–1 years and a few days before their fall on the Earth.

**Summary**

1. By “extremely” low background $\gamma$-ray measurement of the Kobe meteorite, 19 cosmogenic nuclides ($^{24}$Na, $^{28}$Mg, $^{43}$K, $^{44m}$Sc, $^{47}$Sc, $^{52}$Mn, $^{56}$Ni, $^{48}$V, $^{51}$Cr, $^{7}$Be, $^{58}$Co, $^{56}$Co, $^{46}$Sc, $^{57}$Co, $^{54}$Mn, $^{22}$Na, $^{60}$Co, $^{44}$Ti and $^{26}$Al) were detected. Detections of $^{28}$Mg and $^{43}$K are the first case for meteorites. Detection of $^{57}$Ni is quite uncertain because of statistical error.

2. Relatively low activities of neutron-induced $^{60}$Co (~1 dpm/kg) and spallation reaction products such as $^{26}$Al (~38 dpm/kg) suggest that the preatmospheric size of this meteorite was rather small ($r < ~10$ cm) or this fragment was from near the surface region of the meteoroid.

3. Low activities of short-lived spallation products ($t_{1/2} < 1$ y) in the Kobe meteorite, compared with the Tsukuba meteorite, are mainly explained by the 11 year variation in solar activity. In particular, extremely low activity of $^{24}$Na ($t_{1/2} = 15$ h) in the Kobe meteorite may reflect a high solar activity, during a few days before its fall.

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