NON-DESTRUCTIVE MEASUREMENTS OF COSMOGENIC ²⁶Al, NATURAL ⁴⁰K AND FALLOUT ¹³⁷Cs IN ANTARCTIC METEORITES

Kazuhisa Komura, Masaki Tsukamoto and Masanobu Sakanoue

Low Level Radioactivity Laboratory, Kanazawa University, Wake, Tatsunokuchi-machi, Nomi-gun, Ishikawa 923-12

Abstract: Non-destructive γ-ray measurements have been made to determine cosmogenic ²⁶Al, natural ⁴⁰K and fallout ¹³⁷Cs activities in 15 Antarctic meteorites (14 from Yamato Mountains and 1 from Allan Hills). The ²⁶Al activities range from 72 to 29 dpm/kg. If we assume that the saturation activity of ²⁶Al in chondrites is 60, about 1/3 of the measured meteorites show the contents close to this value; however, the rest show lower values. A simple graphical method was applied to estimate the exposure and terrestrial ages based on ²⁶Al and ⁵³Mn data, and these ages are compared with exposure ages obtained by ²¹Ne measurements. The results are generally consistent with the ²¹Ne data. It must be noted that the Antarctic meteorites are highly contaminated with fallout ¹³⁷Cs derived from nuclear test explosions.

1. Introduction

The measurement of cosmogenic nuclides in Antarctic meteorites offers important information both for meteorite and glacial research. Since most of Antarctic meteorites fell a long time ago and have been buried in the ice sheet, detectable cosmogenic radionuclides are limited to only the long-lived ones, such as 14 C ($T_{1/2}$ =5700 y), 36 Cl ($T_{1/2}$ =3.1×10⁵ y), 26 Al ($T_{1/2}$ =7.2×10⁵ y), 10 Be ($T_{1/2}$ =1.5×10⁶ y), 53 Mn ($T_{1/2}$ =3.7×10⁶ y) and some others. Among these, 26 Al and 53 Mn can be determined rather easily by recent techniques. The 26 Al activity is usually determined non-destructively by its 1808 and/or 511 keV γ -rays, using singles or coincidence mode of measurement. On the other hand, 53 Mn can be determined sensitively by a neutron activation method. A number of measurements have been made for 53 Mn in Antarctic meteorites collected both from Yamato Mountains and Allan Hills (IMAMURA *et al.*, 1979; NISHIIZUMI *et al.*, 1978, 1979, 1980), and 26 Al from Allan Hills (EVANS *et al.*, 1979). However, only a small number of data are available concerning 26 Al activities in Yamato meteorites.

Since 1979, measurements of radionuclides in meteorite samples have been one of the research works at the Low Level Radioactivity Laboratory, Kanazawa University. Until now 15 Antarctic meteorites and 3 specimens of Kirin meteorite (Honda et al., 1980, 1982) have been measured. In this report, techniques and results of non-destructive Ge(Li) measurements of ²⁶Al, ⁴⁰K and ¹³⁷Cs are described.

2. Experimental

2.1. Sample

As a first step of our investigation, rather large Antarctic meteorites ALH-766, Y-7305, -74156 and -74191, were chosen to establish the method of measurement and data analysis. Where ALH stands for Allan Hills and Y for Yamato. Among these, the ²⁶Al activity of ALH-766 has already been measured by Evans *et al.* (1979). Since our technique was confirmed as reliable by tentative measurements of these samples, we started the measurement of Yamato-74 series meteorites. Meteorites having weights more than 100 g were chosen for the measurement in the order of catalog number compiled by Yanai (1979). They are Y-74001, -74007, -74010, -74011, -74035, -74036, -74037, -74038, -74080, -74082 and -74418. Among these Y-74010, -74011 and -74037 are diogenites and are paired (Yanai, 1979; Honda, 1981). Since the ⁵³Mn activity in most of these meteorites has already been measured (Imamura *et al.*, 1979; Nishiizumi *et al.*, 1978, 1979, 1980), ²⁶Al data are considered to be very useful to estimate the terrestrial ages.

2.2. Fabrication of mockup for detection efficiency calibration

In order to determine detection efficiency of γ -spectrometer, a mockup sample having the same shape and density as the meteorite must be made for each meteorite sample. For this purpose, usually, a mockup of plaster containing known amount of radionuclides is made to obtain absolute counting efficiency for the meteorite. However, in this work, oil-clay (fat clay) for clay works is used to make a mockup not only because it is easily molded by hand but also because it contains a certain amount of natural radionuclides belonging to the U- and Th-series and 40 K which can be used to obtain a counting efficiency curve.

In the case of large meteorites having weights of more than 500 g, the plaster mold for each meteorite was first prepared and then the mockup was fabricated using oil-clay. On the other hand, for small meteorites, a hand-made mockup of oil-clay was prepared directly by comparing carefully the shape with that of the meteorite.

In order to determine the ⁴⁰K concentration of oil-clay, a disk shaped source (50 mm in diameter and about 50 g in weight) was also prepared from the identical lot of each oil-clay as used for the mockup. Reagent-grade KCl was used as the primary standard for efficiency calibration.

2.3. Detector system and method of measurement

The γ -ray detector used is a 80 cc Ge(Li) detector (Ortec 8501–1523S) having energy resolution of 1.8 keV FWHM at 1.33 MeV and detection efficiency of 16% relative to 7.6 cm \times 7.6 cm NaI (Tl) detector. In order to reduce the background count due to external γ -rays from the surroundings, the Ge(Li) detector was heavily shielded with 5 cm thick mercury as an inner shield and 20 cm thick iron prepared by cutting the steel rotator of an electric generator fabricated in 1931 (SAKURAI and KOMURA, 1981). The typical background of the Ge(Li) detector is 0.003 cpm/keV at 1808 keV region, and the peak area of 1461 keV γ -ray due to 40 K is 0.020 cpm. The spectral data were stored in a full or a half memory of a 4096 channel pulse-height analyzer with the amplifier gain of 0.5 or 1 keV/channel. They were then transferred

and stored in floppy disk memory through mini-computer and were analyzed by an automatic peak search program developed by one of the authors (KOMURA, 1972).

The meteorite, and both the mockup and disc-shaped samples made of oil-clay were placed in turn on the end-cup of the Ge(Li) detector to measure the γ -rays emitted from these samples. Counting time required to attain adequate statistical accuracy depends both on the concentration of radionuclides and the geometrical efficiency of each sample. The meteorite sample requires counting time of about 1 week (10000 min) to attain statistical error of less than 5% for the 1808 keV γ -ray from ²⁶Al. On the other hand, mockup and disk samples require about 2 days of measurement to attain 1 to 2% accuracy for the 1461 keV peak due to ⁴⁰K. The relative efficiency curve was obtained by the counting rates of various γ -rays from ²¹⁴Pb and ²¹⁴Bi (daughters of ²³⁸U) present in the mockup sample. The accuracy of the counts for major peaks of these nuclides ranged from 2 to 5%, due to the branching ratio of each γ -ray and the energy dependency of intrinsic efficiency of the Ge(Li) detector. Besides these samples, the 1461 keV γ -ray from the KCl standard source was also measured with 1–2% of statistical accuracy to determine absolute K concentration in each oil-clay used for the mockup sample.

2.4. Determination of counting efficiency

The K concentration in oil-clay can easily be determined by comparing the count rates of disk samples made from oil-clay and from KCl. The emission rate of the 1461 keV γ -ray from 40 K is calculated to be 104.1 γ s/min/g-KCl by using the half-life of 1.28×10^9 y, isotopic abundance of 0.0117%, and a γ -ray branching ratio of 10.7% (Lederer and Shirley, 1978). The absolute counting efficiency of our Ge(Li) detector is $0.85\pm0.01\%$ at 1461 keV for 50 g of disk KCl source (50 mm $\phi\times13$ mm). The γ -ray peaks from 214 Pb and 214 Bi were used to obtain the relative counting efficiency curve for each mockup source and the absolute counting efficiency was obtained by normalizing the relative efficiency curve at 1461 keV.

The only problem encountered in using mockups of oil-clay is the large differences of solid density between meteorite (ρ =3.3-3.5 g/cm³ as bulk sample) and oil-clay (ρ =2.0 g/cm³), which may affect the self-absorption of γ -rays within the sample. Though accurate estimation of self-absorption can only be made by using a mockup having the same factors for shape, density, and chemical composition as those of meteorite, the self-absorption for the meteorite sample was estimated simply by the following method in our study. As a first approximation, the complicated shape of the meteorite was simplified to a cylindrical shape, of which the diameter and thickness can be arbitrarily chosen so as to give the same volume as each meteorite sample. By using this model, self-absorption factors for the two cases, *i.e.* density of 3.3 and of 2.0 g/cm³, were calculated by a computer code based on a Monte Carlo method. Since the mass attenuation coefficient at 1 to 2 MeV region of γ -ray is approximately the same for most of the materials, mass attenuation coefficient of soil (BECK *et al.*, 1972) was used for the calculation.

3. Results

Typical spectra obtained by non-destructive Ge(Li) measurement are shown in

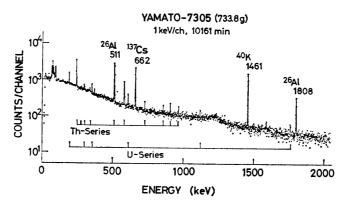


Fig. 1. Ge(Li) spectrum of Yamato-7305. ²⁸Al activity and K content were measured to be 48 dpm/kg and 0.088%, respectively. The γ-ray peak at 662 keV indicates contamination of the meteorite with fallout ¹⁸⁷Cs due to nuclear tests in the atmosphere. Other peaks belonging to U- and Th-series nuclides are the background of our detector system.

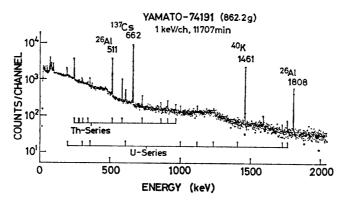


Fig. 2. Ge(Li) spectrum of Yamato-74191. The activity of fallout ¹⁸⁷Cs is very high in this meteorite. Count rate of 662 keV peak is 1.774 cpm, which is the highest among the samples measured.

Figs. 1 and 2 for Y-7305 and Y-74191, respectively. As seen in these figures, the 1808 keV peak due to cosmogenic 26 Al, and the 1461 keV peak due to natural 40 K are clearly observed in the spectra. Besides the 1808 keV γ -ray, the 511 keV γ -ray (annihilation photon) is also emitted from 26 Al. The 511 keV peak is clearly seen in the spectra; however, this peak was not used to determine 26 Al because the contribution of background to this peak is rather high in our detector system (about half of total counts). The 662 keV peak due to 137 Cs ($T_{1/2}$ =30 y) shows the contamination of the meteorite with fallout nuclides produced by the nuclear test explosion performed in the atmosphere. The contribution of background is negligibly small for the 1461 keV peak due to 40 K for most of the meteorites except the diogenite, in which 40 K content is nearly at the detection limit of our detector system. Many other peaks due to U- and Th-series nuclides are in the background of our detector system.

Results of γ -ray measurements are summarized in Table 1 together with the weight and class of each meteorite. The ²⁶Al activities are given as dpm/kg, while ⁴⁰K activity is converted to % K, where the amount of cosmogenic ⁴⁰K in stone meteorites is known to be negligibly small for a bulk sample. The ¹³⁷Cs activity is given in cpm, since this

The second secon				2000			
			Managaina	Measured radioactivities			
Sample	Class	Weight (g)	Measuring time (min)	²⁶ Al (dpm/kg)	⁴⁰ K (%)	137Cs (cpm)	
Yamato-7305	L6	733.8	10161	48±4	$0.088 \pm .005$	$0.415 \pm .012$	
-74001	H5	244.8	10772	38 ± 3	$0.067 \pm .004$	$0.440 \pm .00$	
-74007	L6	147.3	10830	62 ± 5	$0.080 \pm .005$	$0.267 \pm .00^{\circ}$	
-74010	Diogenite	275.9	6228	72 ± 5	$0.001\pm.002$	$0.116 \pm .003$	
-74011	Diogenite	197.7	12396	61 ± 4	$0.001\pm.002$	$0.117 \pm .004$	
-74035	L6	101.0	12533	53 ± 4	$0.069 \pm .005$	$0.263 \pm .00$	
-74036	L6	195.0	10579	$29\!\pm\!3$	$0.085 \pm .004$	$0.598 \pm .00$	
-74037	Diogenite	544.6	5497	$66{\pm}4$	LTD*	$0.140 \pm .00$	
-74038	H5-6	198.4	14523	35 ± 3	$0.046 \pm .005$	$0.354 \pm .00$	
-74080	L6	236.0	9645	41 ± 3	$0.081 \pm .003$	$0.122 \pm .00$	
-74082	H4-5	158.6	10244	51 ± 5	$0.065 \pm .003$	$0.439 \pm .019$	
-74156	H4	707.7	9833	35 ± 3	$0.065 \pm .004$	$0.551 \pm .01$	
"		463.5	7441	$40\!\pm\!4$	$0.077 \pm .007$	$0.234 \pm .00$	
-74191	L3	862.2	11707	71 ± 5	$0.104 \pm .006$	$1.774\pm.03$	
-74418	Н6	340.1	10112	38 ± 3	$0.061 \pm .003$	$0.849 \pm .01$	
Allan Hills-766	H6	562.8	12342	54 ± 4	$0.086 \!\pm\! .005$	$0.469 \pm .00$	
				51 + 1**			

Table 1. ²⁸Al, ⁴⁰K and ¹⁵⁷Cs activities in Antarctic meteorites measured by non-destructive Ge(Li) measurement.

nuclide is considered to be concentrated only on the surface portion of the meteorite sample.

Errors cited in Table 1 consist both of counting statistics of measurement of the meteorite, the mockup, and the KCl standard, and of systematic errors due to the reproducibility of counting geometry and self-absorption correction. Uncertainties in nuclear data, *i.e.* half-lives, γ -ray branching ratio, and isotopic abundance, etc., are not considered in error calculation.

4. Discussion

4.1. ²⁶Al activity

The cosmogenic nuclide 26 Al is produced in meteorites mainly through 28 Si(n, p2n) 26 Al and 27 Al(n, 2n) 26 Al reactions by the interaction of cosmic rays with target silicon and aluminum. The 26 Mg(p, n) 26 Al reaction and spallation reactions of heavier elements also contribute to 26 Al production. Since the elemental abundance of silicon and aluminum in chondrite varies only a little not withstanding with the class of the meteorite (38–40% SiO₂ and 2–3% Al₂O₃), the saturation activity of 26 Al is considered to be nearly equal. On the other hand, diogenite contains high amount of SiO₂(\sim 50%) and saturation activity of 26 Al is expected to be little higher than that of chondrite. The 26 Al activities of 45–70 dpm/kg have been observed in stone meteorites soon after fall, and about 60 dpm/kg is commonly accepted as an average saturation activity of 26 Al. If the preatmospheric meteorite body is large, low saturation activity in near surface region due to 2π irradiation and in deep layer due to shielding effect must be considered.

^{*} Less than detectable. ** Evans et al. (1979).

As given in Table 1, 26 Al activities of Antarctic meteorites measured by the present study ranged from 72 to 29 dpm/kg. Since the 26 Al activity data of Y-74156 measured in 1979 was questioned by Honda (priv. commun., 1982), this sample was remeasured for a smaller specimen. However, new value is consistent with the former one within the counting error. The 26 Al activity of 54 ± 4 dpm/kg for ALH-766 agrees well with 51 ± 1 dpm/kg measured by Evans *et al.* (1979).

Table 1 shows that one-third of the meteorite samples (Y-74007, -74010, -74011, -74037 and -74191) show ²⁶Al activities near the saturation values of 60–70 dpm/kg. Four samples (Y-7305, -74035, -74082 and ALH-766) have ²⁶Al activities of around 50 dpm/kg and the rest (Y-74001, -74036, -74038, -74080, -74156 and -74418) have much lower values. The ²⁶Al and ⁴⁰K activities of three diogenites (Y-74010, -74011 and -74037) are nearly equal, indicating that they are fragments of the same meteorite body as suggested by mineralogical evidence (Yanai, 1979) and by ⁵³Mn and ²¹Ne data (Honda, 1981).

4.2. Estimation of exposure and terrestrial ages

Determination of exposure and terrestrial ages of meteorite samples is very useful to investigate the frequency of meteorite fall, spread of a shower, and their accumulation mechanism in special areas in Antarctica, etc. The exposure histories of Antarctic meteorites have been successfully investigated by cosmogenic long-lived nuclides (IMAMURA et al., 1979; NISHIIZUMI et al., 1979), cosmogenic ⁴⁰K in a magnetic fraction (NITOH et al., 1979), and rare-gas components (TAKAOKA and NAGAO, 1978; NAGAO and TAKAOKA, 1979; TAKAOKA et al., 1981). If the exposure age is sufficiently long as compared with the half-lives of the radionuclides, the terrestrial age can be estimated simply by the ratio of measured to saturation activity. In an ideal case, the terrestrial age can in principle be estimated from the data of a single nuclide with the proper half-life. But this method sometimes leads to erroneous results, therefore data of two or more nuclides must be used to obtain a reliable age. In special cases, however, a complicated exposure history must be considered to explain consistently the measured activities (NISHIIZUMI et al., 1979; IMAMURA et al., 1979).

Since ⁵³Mn data are available for most meteorite samples studied in this work (compiled by Honda, 1981), estimation of both exposure and terrestrial ages can be made by a simple graphical method described below. In the first step, the ²⁶Al and ⁵³Mn activities were plotted on a logarithmic or linear scale section paper as the activity ratio against their saturation values. Figures 3 and 4 are the examples of the logarithmic case. Here, the saturation activities of ²⁶Al and ⁵³Mn are arbitrarily taken, respectively, to be 60 dpm/kg and 430 dpm/kg-Fe. It is well known that the saturation activities of ²⁶Al and ⁵³Mn depend highly on the size of the preatmospheric body and the depth in it. Therefore, present method is valid only in the case that proper values are taken for saturation activities of ²⁶Al and ⁵³Mn. If the chemical composition of all the meteorite samples is known, it seems better to use ²⁶Al activity normalized by SiO₂ content, just like the case of ⁵³Mn. For simplicity, error is not considered in these values in Fig. 4.

As shown in Fig. 3, the increase of ²⁶Al and ⁵³Mn activities is given by a thick line curve with an arrow. After the meteorite fall, the ²⁶Al and ⁵³Mn activities decrease

²⁶Al Activity (relative to saturation value)

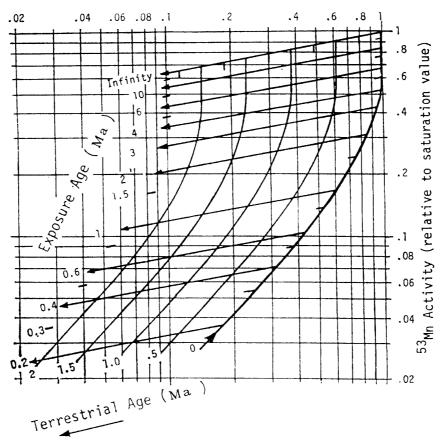
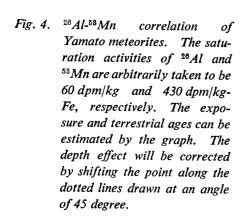


Fig. 3. Growth and decay curves of ²⁸Al and ⁵³Mn activities. The ²⁸Al and ⁵³Mn activities grow along the thick line curve. After the meteorite fall, their activities decrease along the straight line from right to down-left direction. The height of turning point corresponds to the exposure age.

by their own half-lives. If the logarithmic scale is used, the decay of ²⁶Al and ⁵³Mn is given by the straight line from right side to lower-left direction in the figure. The height of the turning point depends on the exposure age. In Fig. 4, the data of Yamato meteorites are shown on the enlarged figure of logarithmic scale. Since Y-74155 and -74156 are paired, ⁵³Mn value of the former is used for Y-74156. The error bar given in the figure accounts only for the statistical error of measurements, while the uncertainty of saturation activities assumed here is not considered for both ²⁶Al and ⁵³Mn. The exposure and terrestrial ages can be estimated easily from the scales given at left and lower part of the figure. In ideal cases, all the data points must be confined by the area surrounded by upper and right-most curves. However, as seen from Fig. 4, Y-74011, -74037 and -74191 data are beyond this area, and Y-74035 seems to have a little lower ²⁶Al activity than that expectable from ⁵³Mn activity. These anomalies may be explained by using the proper value for saturation activities, which will be obtained by considering the depth profile of nuclear interactions in the meteorite body (IMAMURA *et al.*, 1973; NISHIIZUMI *et al.*, 1979). If both the ²⁶Al and ⁵³Mn-producing



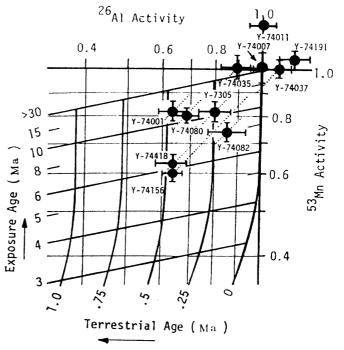


Table 2. Exposure and terrestrial ages estimated by graphical method.

		Activity (d	pm/kg)			Expos. age
Sample	Class	$(T_{1/2} = .72 \text{ Ma})$	⁵⁸ Mn ^a (3.7 Ma)	Terrest. age corr. (Ma) age	Expos. age (Ma)	by other method (Ma)
Adopted saturatio	n	The state of the s	A CONTRACTOR OF THE STATE OF TH		The state of the s	THE NATIONAL ASSESSMENT OF THE PROPERTY OF A SHARE THE
value		60	430			
Yamato-7305	L6	48 ± 4	352 ± 18	0.23(.1)	9-13	22 ^b
-74001	H5	38 ± 3	351 ± 16	0.46	11-17	
-74007	L6	62 ± 5	433 ± 19	< 0.1	>15	
-74035	L6	$53{\pm}4$	440 ± 19	0.24(.2)	>15	92°
-74036	L6	29 ± 3		0.68		
-74038	H5-6	35 ± 3		0.57		
-74080	L6	41 ± 3	345 ± 15	0.35 (.2)	10–14	49°, 46ª
-74082	H45	51±5	320 ± 13	0.13	7-9	<i>i</i> , 10
-74155	H4		255 + 15		, ,	50
-74156	H4	38+2°	- Combour	0.45 (.5)	5-7	5
-74191	L3	71 ± 5	450 ± 23	<0.1 (<.1)	10	12 ^b
-74418	Н6	38 ± 3	275 ± 11	0.46 (.5)	6–7	7°, 7ª
-74010	Diogenite	72±5		<0.1	0 /	, , ,
-74011	Diogenite	61+4	525 ± 23	<0.1 (<.1)	:>15	32f
-74037	Diogenite	66±4	445+16	<0.1	>15	32-
	Averages	68	470	<0.1	>15	
Allan Hills-766	H6	54+4	110	0.12 (.1)	~13	1.Ch
	110	51±1 ^g		0.12 (.1)		16 ^h

- a: Taken from compiled data by Honda (1981).
- b: TAKAOKA and NAGAO (1978).
- с: Такаока et al. (1981).
- d: Nітон et al. (1980).

- e: Average of two measurements.
- f: Nagao and Takaoka (1979).
- g: Evans et al. (1979).
- h: SCHULTZ (1978).

reactions have nearly the same depth profile, the correction can be made simply by shifting the point to the lower-left or upper-right direction along the dotted 45° line.

The exposure and terrestrial ages thus estimated are given in Table 2, together with the exposure ages estimated by 21Ne (TAKAOKA and NAGAO, 1978; NAGAO and Такаока, 1979; Такаока et al., 1981; Shultz, 1978) and 40K (Nitoh, et al., 1980). The corrected terrestrial ages are given in parentheses. As described above, the saturation activities of 26Al and 53Mn are uncertain, therefore, the terrestrial and exposure ages estimated by the graphical method are not quite sure. Y-74007, -74011, -74037 and -74191 are considered to have short terrestrial ages less than 0.1 Ma, since both their ²⁶Al and ⁵³Mn activities are higher than the saturation values assumed here. The exposure ages estimated by the graphical method agree well with other methods for Y-74156 and Y-74418, while rather low values were obtained for Y-7305 and Y-74080 by this method. These differences may be explained by the shielding effect which gives low saturation activity. The terrestrial ages corrected by using exposure ages estimated by other methods become 0.1 and 0.2 Ma for Y-7305 and Y-74080, respectively. The terrestrial age of Y-74156 by the graphical method is estimated to be 0.5 Ma. According to Honda (priv. commun., 1982), the ³⁶Cl activity measured recently for metal phase of Y-74155 indicates that the terrestrial age must be less than 0.1 Ma. If this value is adopted, the exposure age by the graphical method becomes 15 Ma, which seems inconsistent with the 21 Ne data. Y-74036 has the lowest 26 Al activity among the measured meteorites, which corresponds to an apparent terrestrial age of 0.7 Ma. Measurements of ³⁶Cl and ⁵³Mn activities and also ²¹Ne data are strongly required to obtain a reliable age.

As described above, the graphical method is very simple and may be used to estimate roughly relations among exposure and terrestrial ages and shielding simultaneously. This method can be applied to various combination of cosmogenic nuclides, such as ²⁶Al-³⁶Cl, ²⁶Al-¹⁰Be, ³⁶Cl-¹⁰Be, ²¹Ne-⁴⁰K, etc. A three-dimensional expression using ³⁶Cl-²⁶Al-⁵³Mn, ²⁶Al-⁵³Mn-²¹Ne, etc. will give useful information to investigate complex exposure and terrestrial histories and shielding of meteorite samples.

4.3. Contamination of meteorites with fallout nuclides

A fissiogenic nuclide ¹³⁷Cs with half-life of 30 y has been scattered on a global scale along with many kinds of fissiogenic nuclides by nuclear explosions performed in the atmosphere during the past three decades. As known from Ge(Li) spectra shown in Figs. 1 and 2, and Table 1, all the meteorites measured are highly contaminated with this nuclide. The ¹³⁷Cs activity of Y-74191 is the highest and those of three diogenites and Y-74080 are relatively low. The contamination is considered to be limited only to the surface portion unless the meteorite has been considerably weathered. But there seems no apparent correlation between ¹³⁷Cs activity and surface area (or weight) of meteorite.

Since 137 Cs can be detected easily by non-destructive γ -spectrometry, this nuclide can be used as an indicator for the contamination of a meteorite sample with natural and artificial fallout. However, this indicator can be used only for the last three decades because nuclear tests began only about 30 years ago.

Acknowledgments

The authors wish to thank Dr. K. Yanai and the National Institute of Polar Research for providing the Antarctic meteorites. We are grateful to Prof. M. Honda for encouraging and useful suggestions. We also thank students of our laboratory for their assistance in γ -ray measurements and preparation of mockups.

References

- BECK, H. L., DECAMPO, J. and GOGOLAK, C. (1972): In situ Ge(Li) and NaI (T1) *γ*-ray spectrometry. Health and Safety Laboratory, HASL-258.
- Evans, J. C., Rancitelli, L. A. and Reeves, J. H. (1979): ²⁸Al content of antarctic meteorites: Implications of terrestrial ages and bombardment history. Proc. Lunar Planet. Sci. Conf. 10th, 1061–1072.
- Honda, M. (1981): Terrestrial history of Antarctic meteorites recorded in the cosmogenic nuclides. Geochem. J., 15, 163-181.
- Honda, M., Horie, K., Imamura, M., Nishiizumi, K., Takaoka, N. and Komura, K. (1980): Irradiation history of the Kirin meteorite. Geochem. J., 14, 83–89.
- HONDA, M., NISHIIZUMI, K., IMAMURA, M., TAKAOKA, N., NITOH, O., HORIE, K. and KOMURA, K. (1982): Cosmogenic nuclides in the Kirin chondrite. Earth Planet. Sci. Lett., 57, 101–109.
- IMAMURA, M., FINKEL, R. C. and Wahlen, M. (1973): Depth profile of ⁵⁸Mn in the lunar surface. Earth Planet. Sci. Lett., **20**, 107–112.
- IMAMURA, M., NISHIIZUMI, K. and HONDA, M. (1979): Cosmogenic ⁵⁸Mn in Antarctic meteorites and their exposure history. Mem. Natl Inst. Polar Res., Spec. Issue, **15**, 227–242.
- Komura, K. (1972): Kô-bunkainô γ -sen supekutoru no jidô kaiseki puroguramu (Automatic peak search program of high resolution γ -ray spectrum). Institute for Nuclear Study, Univ. of Tokyo, INS-TCH-7.
- LEDERER, C. and SHIRLEY, V. S. (1978): Table of Isotopes. 7th ed. New York, John Wiley.
- NAGAO, K. and TAKAOKA, N. (1979): Rare gas studies of Antarctic meteorites. Mem. Natl Inst. Polar Res., Spec. Issue, 12, 207–220.
- NISHIIZUMI, K., IMAMURA, M. and HONDA, M. (1978): Cosmic ray induced ⁵⁸Mn in Yamato-7301(j), -7305(k) and -7304(m) meteorites. Mem. Natl Inst. Polar Res., Spec. Issue, **8**, 209–219.
- NISHIIZUMI, K., IMAMURA, M. and HONDA, M. (1979): Cosmic ray produced radionuclides in Antarctic meteorites, Mem. Natl Inst. Polar Res., Spec. Issue, 12, 161–177.
- Nishiizumi, K., Murrell, M. T., Arnold, J. R., Imamura, M. and Honda, M. (1980): Cosmogenic ⁵³Mn survey of Yamato meteorites. Mem. Natl Inst. Polar Res., Spec. Issue, 17, 202–209.
- NITOH, O., HONDA, M. and IMAMURA, M. (1980): Cosmogenic K-40 in Antarctic meteorites. Mem. Natl Inst. Polar Res., Spec. Issue, 17, 189-201.
- SAKURAI, J. and KOMURA, K. (1981): Secular change of background activity in Ge(Li) detector. Radioisotopes, 30, 471-474.
- SCHULTZ, L. (1978): Abstract. The 41st Ann. Meeting Meteoritical Soc., Aug. 14-17, 1978, Sudbury, Canada.
- Takaoka, N. and Nagao, K. (1978): Rare gas studies of Yamato-7301 (j), -7304 (m) and -7305 (k). Mem. Natl Inst. Polar Res., Spec. Issue, 8, 198-208.
- TAKAOKA, N., SAITO, K., OHBA, Y. and NAGAO, K. (1981): Rare gas studies of twenty-four Antarctic chondrites. Mem. Natl Inst. Polar Res., Spec. Issue, 20, 264–275.
- YANAI, K., comp. (1979): Catalog of Yamato Meteorites. 1st ed. Tokyo, Natl Inst. Polar Res., 188 p. with 10 pls.

(Received June 12, 1982; Revised manuscript received August 23, 1982)