Bull. Natn. Sci. Mus., Tokyo, Ser. E, **12**, December 22, 1989

Cosmogenic ^{26}Al and ^{53}Mn in Japanese Chondrites

By

Sadayo YABUKI

The Institute of Physical and Chemical Research,
Wako-shi, Saitama

and

Masako SHIMA

Department of Science and Engineering,
National Science Museum, Tokyo

Abstract Cosmic-ray-produced long-lived radioactive nuclides, ^{26}Al and ^{53}Mn, were determined in eighteen chondrites which fell and recovered in Japan, by non-destructive γ-ray spectrometry and neutron activation analyses, respectively.

Data were compared with those of published spallogenic neon data obtained from the same chondrite specimens as well as published ^{53}Mn data. Generally, the correlation of ^{26}Al and ^{53}Mn with ^{22}Ne/^{21}Ne show normal trends in chondritic meteorites, except the Aomori, Kamiomi, Numakai and the gas-rich chondrites Nio and Fukutomi.

Cosmic-ray exposure ages of those chondrites were calculated from data of ^{53}Mn activities and ^{53}Mn/^{26}Al pairs and were compared with conventional ^{21}Ne ages. Obtained ages by the present methods are generally in good agreement with ^{21}Ne ages.

Discrepancies between ^{26}Al data and data of ^{53}Mn and apallogenic neon isotopic ratio of Kamiomi and between present ^{53}Mn data and published one of Satsuma (Kyushu) are due to the different sampling positions. High ^{21}Ne age compared with the present data in the Aomori could be explained by multi-irradiation model while low ^{21}Ne age observed in the Numakai are probably result of the gas loss by heavy shock of this meteorite in space.

1. Introduction

Cosmic-ray-produced radioactive nuclides in chondrites have been determined in time to time since 1950's (EHMANN and KOHMAN, 1958; ANDERS, 1960 and 1962; HONDA *et al.*, 1961; ROWE *et al.*, 1963; HONDA and ARNOLD, 1964). Especially ^{26}Al and ^{53}Mn have been measured in great numbers of chondrites (e.g., NISHIIZUMI, 1987). These nuclides have relatively long half-lives such as $t_{1/2}=7.2\times10^5$ y and 3.7×10^6 y respectively. ^{26}Al emits high energy γ-ray and ^{53}Mn converts efficiently to highly sensitive nuclide ^{54}Mn through the reaction with slow neutron. Results were compared with rare gas data and were used for discussing their cosmic ray exposure ages, preatmospheric size of respective meteorites, depth effects for the production of spallogenic nuclides and constansy of cosmic-ray intensity.

In this report, we present the analytical results of ^{26}Al and ^{53}Mn determined on fifteen- for each, altogether eighteen chondrites which had fallen in Japan.

2. Experimental

2.1. *Chondrite Sample*

Chondrites used in this study are listed in Table 1.

2.2. *Standard Materials*

2.2.1. *Standard for ^{26}Al*

The standard of ^{26}Al was prepared as follows: an aliquot of the calibrated source of ^{26}Al solution from New England Nuclear Co. Ltd., was deposited onto small aluminum plate, dried and wrapped with polyethylene sheets. For the standard of ^{40}K, chemically pure KCl was accurately weighed and employed by assuming the isotopic composition of ^{40}K as normal terrestrial abundance, 0.0117 atom %.

2.2.2. *Standard for ^{53}Mn*

The ^{53}Mn standard was provided by M. Imamura, originally extracted from the iron meteorite Grant by M. Honda (Tokyo standard). The original 5.24 dpm ^{53}Mn/

Table 1. Chondrites studied in this work.

Chondrite	Class	Date of fall	Recovered weight (kg)	Ref.
Nogata	L6	861/ 5/19	0.472	[1]
Ogi	H6	1741/ 7/ 8	14.4	[2]
Kesen	H4	1850/ 6/12	135	[3]
Sone	H	1866/ 6/ 7	17.1	[4, 5]
Takenouchi	H	1880/ 2/18	0.719	[6]
Fukutomi	L4	1882/ 3/19	16.77	[7]
Satsuma (Kyushu)	L6	1886/10/26	>46.5	[8]
Nio	H3–4	1897/ 8/ 8	0.465	[9, 10]
Kamiomi	H5	1915	0.448	[11]
Tane	L5	1918/ 1/25	0.906	[9]
Nagai	L6	1921/ 5/30	1.81	[12]
Numakai	H4	1925/ 9/ 5	0.356	[13, 14]
Duwun	L6	1943/11/23	2.117	[15]
Okabe	H	1958/11/26	0.194	[9]
Shibayama	L6	found 1969	0.235	[16]
Aomori	L6	1984/ 6/30	0.320	[17]
Tomiya	H5–6	1984/ 8/22	0.0275	[17]
Kokubunji	L6	1986/ 7/29	11	[18]

[1] SHIMA *et al.*, 1983. [2] YABUKI *et al.*, 1981. [3] MIYASHIRO, 1962. [4] MIYASHIRO *et al.*, 1963. [5] KANDA, 1956. [6] MIYASHIRO *et al.*, 1973. [7] SHIMA *et al.*, 1979a. [8] MASON and WIIK, 1961. [9] MIYASHIRO and MURAYAMA, 1967. [10] SHIMA *et al.*, 1984. [11] OKADA *et al.*, 1979. [12] MURAYAMA *et al.*, 1978. [13] SHIMA, 1974. [14] YAGI *et al.*, 1976. [15] YABUKI and SHIMA, 1980. [16] SHIMA *et al.*, 1979b. [17] SHIMA *et al.*, 1986. [18] SHIMA and MURAYAMA, 1987.

mg-Mn solution was diluted to 1.56 dpm ^{53}Mn/mg-Mn with 200.0 ppm-Mn carrier solution by Imamura. An aliquot of the solution was deposited onto pure aluminum foil and dried. The standard was wrapped with pure aluminum foil, then irradiated by neutron together with samples.

In order to estimate efficiency of $(n, 2n)$ and $(n, p2n)$ side reactions occurred in meteorite samples by fast neutron in the reacter, several mg of 99.9% manganese metal powder and 99.99% of iron metal wire were taken, weighed and wrapped with aluminum foil, and irradiated together with samples.

2.3. *Determination of* ^{26}*Al*

For γ-ray spectrometry of ^{26}Al, whole chondrite specimens were used without any pretreatment. The determination of radioactivities of specimens except Kokubunji were performed by using coaxial type Ge(Li) γ-ray detector, 95 cm^3 in active volume, with 4096 multichannel analyzer. The guaranteed system resolution is 2.3 keV for 1332 keV ^{60}Co γ-ray and the counting efficiency is 15.2% for 511 keV positron annihilation peak and 2.5% for 1809 keV ^{26}Al photopeak. Typical background were obtained as 0.49 cpm (511 ± 3.5 keV) and 0.01 cpm (1809 ± 2.5 keV). The chondrite Kokubunji was measured by coaxial type pure Ge γ-ray detector, which will be described elsewhere.

For the determination of ^{26}Al and ^{40}K, 1809 and 1462 keV photopeaks were used respectively. The ^{40}K data were used as internal standard for approximate estimation of ^{26}Al activity by computing from activities of ^{40}K and potassium content.

After counting the radioactivity of each chondrite, the measurement was also performed on the mockup having the same geometry as the chondrite's in order to convert the obtained relative data to the absolute values.

A mockup of each chondrite was made up as follows: the shell was fabricated by covering each sample with aluminum foil, being pressed tightly against the surface to conform closely to its shape. A molding compound, araldite resin, was then coated on the surface of aluminum foil, about half of the area. After the compound hardened, the shell was removed from the meteorite and the another half area of the shell was fabricated in the same way. The two parts were then glued together using more amount of molding compound to form complete dummy shell. Olivine sand and iron powder were mixed thoroughly with the standards, and the mixture was stuffed into mockups for building the same specific density as the chondrite had.

For freshly fallen chondrites, Aomori, Tomiya and Kokubunji, mockups were made together with standard nuclides of short-lives, such as ^{22}Na, ^{51}Cr and ^{54}Mn.

Details of counting and calculating process are tabulated in Table 2.

2.4. *Determination of* ^{53}*Mn*

About 0.3 g specimens of each chondrite sample were powdered in the agate morter at liquid nitrogen temperature. The magnetic fraction was separated from stony phase by hand magnet and cleaned ultrasonically in aceton. The procedure

Table 2. Details for ^{26}Al determination.

Chondrite	Specimen used (g)	Time to measure (sec)	^{26}Al Activity obtained (counts)	Mean* (counts)	Counting Efficiency (10^{-3})	Obtained ^{26}Al (dpm/kg)** 1	2
Nogata	461	400000 400000	273±17 272±17	272±12	1.61 ±0.09	55± 4	259
Kesen	175	800000	978±66	489±33	6.98 ±0.38	60± 5	322
Sone	17000	400000 400000	1414±58 1339±64	1377±43	0.224±0.013	54± 3	276
Takenouchi	36	800000	306±58	153±29	11.4 ±0.6	56±10	292
Fukutomi	2140	400000 400000	830±36 725±37	778±26	0.880±0.046	62± 4	300
Satsuma (Kyushu)	600	400000 768209	440±26 816±36	430±15	1.92 ±0.10	56± 3	272
Nio	182	400000 599101	197±20 258±19	180±11	2.63 ±0.13	56± 4	292
Kamiomi	432	800000	452±32	226±16	1.43 ±0.07	55± 4	278
Tane	270	800000 400000	566±34 270±25	279±14	2.42 ±0.13	64± 5	310
Duwun	2080	248524	575±28	925±45	1.06 ±0.06	63± 5	313
Okabe	119	800000	258±28	129±14	2.75 ±0.15	59± 7	302
Shibayama	199	800000 757892 800000	374±30 381±28 408±36	197± 9	2.42 ±0.12	61± 4	303
Aomori	154	800000 800000 800000 800000	346±26 326±26 256±30 232±24	168± 9 121± 9	4.62 ±0.32 3.19 ±0.22	37± 2	177
Tomiya	19	1600000 1600000	178±29 216±29	50± 5	6.44 ±0.62	60± 8	313
Kokubunji	157	1010000 2080000	554±35 1027±48	205± 8	3.19 ±0.12	61± 3	298

* Mean value were calculated per 400,000 sec.

** Columns 1 and 2 of that in "Obtained ^{26}Al" were given by dpm per kg of whole sample and of target which was calculated to equivalent amount of Si from chemical data, dpm/kgSi$_{\mathrm{equ.}}$, (HERPERS and ENGLERT, 1983), respectively.

was repeated several times until no stony phase was visible in the sight area of the microscope. The purified magnetic fraction was weighed and dissolved in dil. HNO_3.

The separation of Mn by anion and cation exchange resins and ^{53}Mn determination via ^{54}Mn by radiochemical neutron activation analysis was performed by the method by IMAMURA *et al.* (1973). The irradiation by thermal neutron was made in the VT-5 core of the JRR-2 reactor of the Japan Atomic Energy Research Institute, Tokai-Mura. Irradiation (a) on June 16–27, 1985, and irradiation (b) on January 19–30, 1987, were carried out for one cycle, approximately 268 hours. The position in the reactor was selected where the fast neutron flux is as low as possible. The ^{53}Mn standard, Fe- and Mn-metal were also irradiated together with samples. Irradiation characteristics of each experiment are shown in Table 3.

^{54}Mn activities of the samples of irradiations (a) and (b) were counted using

Table 3. Irradiation characteristics for ^{53}Mn determined from standards.

Standard used	Recovery (%)	Obtained ^{54}Mn Activity (cph)	Determined Quantity per unit	Determined Quantity Weighted Mean
Irradiation (a)				
^{53}Mn (dpm)			cph ^{54}Mn/dpm ^{53}Mn	
1 0.0270	85.3	116.6±2.1	4118±226	3914±152
2 0.0256	88.3	100.0±2.2	3707±205	
Mn (mg)			cph ^{54}Mn/mg Mn	
1 6.49	90.0	2130± 2	328± 4	317± 9
2 4.60	83.7	1400±17	304± 4	
Fe (mg)			cph ^{54}Mn/mg Fe	
1 0.71	100	1846±59	2600±83	2637±43
2 0.95	88.8	2519±52	2650±50	
Irradiation (b)				
^{53}Mn (dpm)			cph ^{54}Mn/dpm ^{53}Mn	
1 0.0228	95.9	26.8±1.6	1150±70	1102± 9
2 0.0207	97.0	23.4±1.0	1107±48	
3 0.0191	97.5	19.4±1.3	993±68	
4 0.0193	96.0	22.2±1.2	1127±62	
Mn (mg)			cph ^{54}Mn/mg Mn	
1 16.43	96.0	635± 6	38.6±0.4	35.7±0.2
2 14.44	96.1	485± 5	33.6±0.3	
Fe (mg)			cph ^{54}Mn/mg Fe	
1 1.23	85.0	226± 2	184± 2	229±1.4
2 1.04	90.0	272± 2	262± 2	

Irradiation (a): May 16–27, 1985.
Irradiation (b): Jan 19–30, 1987.

Ge(Li) detector and pure Ge detector of Ortec Co. Ltd., respectively. The counting efficiency of Ge(Li) detector was 8.0% for ^{54}Mn photopeak, and the back ground was 0.05 cpm (834±2.5 keV). The counting efficiency of pure Ge detector was 2.4%, and the back ground was 0.03 cpm. The counting process of ^{54}Mn and determination of ^{53}Mn are presented in Table 4.

3. Results and Discussions

3.1. *Results*

The detailed results for γ-ray spectrometry and neutron activation analyses of ^{26}Al and ^{53}Mn, respectively, are given in Table 5 together with ^{53}Mn data by NISHIIZUMI (1978 and 1987), and spallogenic ^{21}Ne contents and ^{22}Ne/^{21}Ne ratios (TAKAOKA *et al.*, 1989) in the respective chondrites. Errors quoted with ^{26}Al data are sum of counting statistics of ^{26}Al γ-ray measurements of chondrites, of mock-ups and of back grounds. The uncertainty of standard ^{26}Al activities were estimated as ±5%. Errors for ^{53}Mn activities include those of counting statistics for ^{54}Mn activities, errors in the Mn and Fe determination by atomic absorption spectrometry, and the errors in the

Table 4. Details for ^{53}Mn determination.

Chondrite		Magnetic Fraction				Irradiated Sample			
Name	taken (g)	Total (mg)	Fe (mg)	Ni (mg)	Mn (μg)	Mn (μg)	Fe (μg)	Gross ^{54}Mn (cph)	Net ^{54}Mn (cph)
Irradiation (a)									
Nogata	0.555	24.5	13.6	0.76	12.4	10.5	1.21	22.3±1.1	15.8±0.8
Ogi	0.518	73.7	52.0	4.7	32.8	26.6	3.81	73.3±2.9	54.9±2.9
Fukutomi	0.716	49.5	33.6	1.85	37.0	29.2	5.69	59.9±2.9	35.7±3.0
Nio	0.343	64.2	46.2	3.8	54.2	38.3	2.50	80.4±4.4	61.7±3.6
Duwun	0.899	85.8	54.0	3.3	70.9	57.8	12	122.0±5.6	71.7±6.0
Aomori	0.572	38.4	27.3	2.1	16.6	12.4	3.37	36.3±1.1	23.5±1.5
Tomiya	0.308	62.5	47.7	3.3	27.9	27.7	2.5	84.2±2.0	68.9±2.0
Irradiation (b)									
Kesen	0.793	143	106.9	8.8	117.0	72.1	0.20	28.9±1.6	26.1±1.4
Sone	0.205	71.3	33.9	1.9	72.9	35.5	0.93	9.2±0.5	6.9±0.4
Satsuma (Kyushu)	0.392	27.3	16.6	2.9	21.6	20.1	0.22	7.9±0.5	7.0±0.4
Kamiomi	0.755	176	109.1	8.7	164.4	118.2	0.18	23.8±1.3	19.3±1.1
Nagai	0.495	40.7	28.0	4.8	22.3	16.8	0.16	9.5±0.5	8.7±0.5
Numakai	0.589	162	60.8	6.9	135.4	75.1	0.12	17.0±1.7	14.2±1.4
Okabe	—	108	59.5	6.3	17.2	14.0	0.07	18.8±1.1	18.2±1.0
Kokubunji	0.570	45.0	30.0	4.9	22.7	18.4	0.07	10.5±0.6	9.8±0.6

Table 5. Results of the ^{26}Al and ^{53}Mn determination.

Chondrite	^{26}Al (dpm/kg)***	^{53}Mn (dpm/kg)*		^{21}Ne** (10^{-8})	^{22}Ne/^{21}Ne
		This work	Publ.****		
Nogata	55± 4	344±25	271±11	15.8	1.168
Ogi	51± 3*****	323±18	270±11	7.60	1.179
Kesen	60± 5	350±33	347±23	3.20	1.087
Sone	54± 3	372±36	413±15	5.24	1.087
Takenouchi	56±10	—	487±17	4.36	1.068
Fukutomi	62± 4	338±29	294±10 316±12	3.82	1.316
Satsuma (Kyushu)	56± 3	388±39	468±22	14.1	1.068
Nio	56± 4	470±29	403±16	12.4	1.460
Kamiomi	55± 4	217±20	213± 9	1.39	1.222
Tane	64± 5	—	409±18	15.1	1.124
Nagai	—	354±34	336±14	2.76	1.092
Numakai	—	368±46	—	1.64	1.111
Duwun	63± 5	408±34	380±15	5.86	1.085
Okabe	59± 7	329±31	314±13	2.19	1.109
Shibayama	61± 4	—	322±16	4.47	1.144
Aomori	37± 2	287±54	—	14.3	1.133
Tomiya	60± 8	363±15	—	2.25	1.144
Kokubunji	61± 3	347±32	—	5.44	1.157

* unit: dpm/kg {Fe+(1/3)×Ni}.

** Neon data: after TAKAOKA *et al.* (1989). Unit of ^{21}Ne concentrations: 10^{-8} cm^{3}STP/g sample.

*** unit: dpm/kg sample.

**** after NISHIIZUMI (1978 and 1987).

***** after BHANDARI *et al.* (1979).

measurements of standards listed in Table 3. The present ^{53}Mn data for seven chondrites out of eleven agree with the published data well. The Nogata and Ogi seem to be better in the present value (see also Fig. 3 and Table 6). On the Satsuma (Kyushu), discrepancy could be due to different sampling position, and on the Nio, the reason is still unknown. As seen in Table 5, ^{26}Al in most chondrites measured are reached apparently secular equilibrium except that in the Aomori. Data for the Sone and Satsuma show somehow lower than the saturation values. In the case of ^{53}Mn, about a half of chondrites measured were not saturated.

3.2. *Comparison with rare gas data*

As is well known, spallogenic ^{22}Ne/^{21}Ne ratios of chondrites are the indicator of the hardness of cosmic-ray energy in the position where samples were located. Therefore, the shielding conditions could be estimated from the neon isotope ratio. Present data of ^{26}Al in L- and H-chondrites and ^{53}Mn in all chondrites against the ratios were plotted in Figs. 1, 2 and 3, respectively. The figures are proposed by NISHIIZUMI *et al.* (1980) for rather small chondrite, recovered mass of which is less than 200 kg.

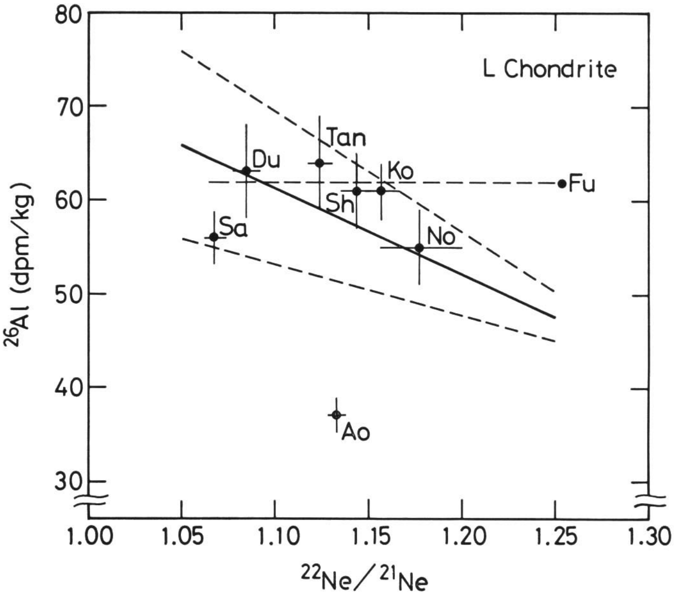

Fig. 1. The relationship between saturation ^{26}Al activities and ^{22}Ne and ^{21}Ne ratios in L and LL chondrites. No: Nogata, Fu: Fukutomi, Sa: Satsuma, Tan: Tane, Du: Duwun, Sh: Shibayama, Ao: Aomori, Ko: Kokubunji.

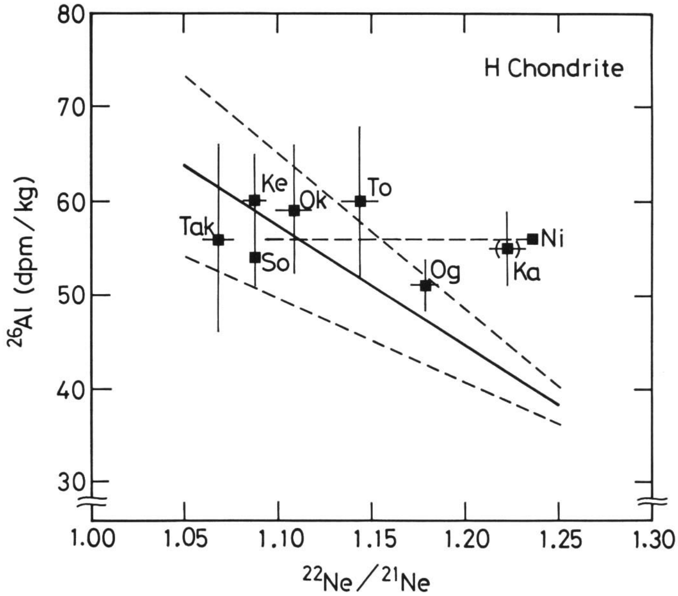

Fig. 2. The relationship between saturation ^{26}Al activities and ^{22}Ne and ^{21}Ne ratios in H chondrites. Og: Ogi, Ke: Kesen, So: Sone, Tak: Takenouchi, Ni: Nio, Ka: Kamiomi, Ok: Okabe, To: Tomiya.

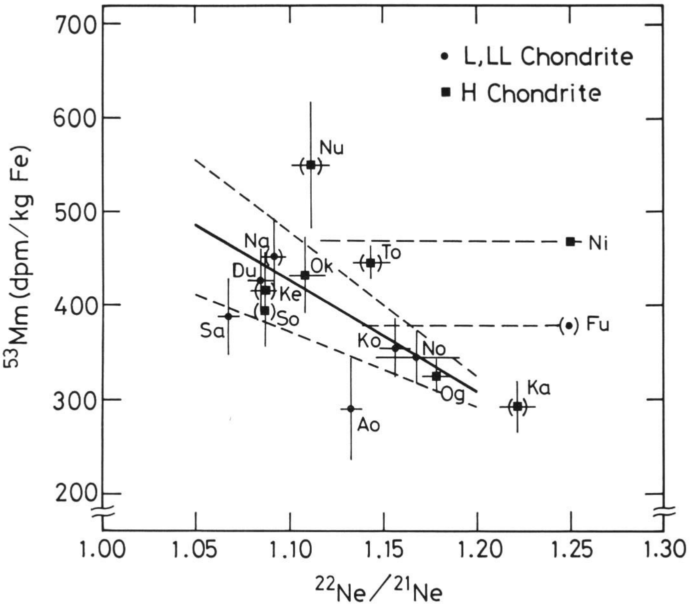

Fig. 3. The relationship between saturation ^{53}Mn activities and ^{22}Ne/^{21}Ne ratios. Na: Nagai, Nu: Numakai, Others are the same as Figs. 1 and 2.

and the cosmic-ray exposure age of which is long, cosmogenic ^{21}Ne content being higher than 1.5 and 5.5×10^{8} cm^3/g for ^{26}Al and ^{53}Mn, respectively. According to them, the data obtained from such chondrites fall within the areas of dotted lines. The gas-rich chondrites, Fukutomi and Nio, are exceptional, because it is hard to obtain accurate cosmogenic ^{22}Ne/^{21}Ne ratio. In Figs. 1 and 2, both the Aomori and Kamiomi deviate largely from the anti-correlation areas. The Kamiomi is only a chondrite with ^{21}Ne content less than 1.5×10^{8} cm^3/g, while the Aomori is just in the range. For ^{53}Mn, only Numakai, Tomiya and Aomori are not on the area, though about a half of chodrites have somewhat lower cosmogenic ^{21}Ne contents than that by their proposal. In the Fig. 3, chondrites whose cosmogenic ^{21}Ne contents are lower than 5.5×10^{8} cm^3/g are shown by square points with bracket.

3.3. *Cosmic-ray exposure ages*

As was described in the former section, ^{26}Al activities in chondrites used in this study, reached secular equilibrium except Aomori. Therefore cosmic-ray exposure ages of those chondrites must be longer than 5 million years. This is true from the ages obtained from spallogenic ^{21}Ne concentration of each chondrite (TAKAOKA *et al.*, 1989). The present ^{53}Mn activity data were compared with saturated ^{53}Mn values estimated from the ^{22}Ne/^{21}Ne ratios (NISHIIZUMI *et al.*, 1980) of each chondrite in order to compute cosmic-ray exposure ages. For the gas-rich condrites, Fukutomi and Nio, ^{22}Ne/^{21}Ne ratios are estimated as values of average shielding, 1.11 (NISHI-

Table 6. Exposure ages of chondrites studied.

Chondrite	^{21}Ne Age* (myr)	^{53}Mn Age (Myr)		^{53}Mn/^{26}Al (Myr)	
		Present Work	Published**	Present Work	Published**
Nogata	64	>20	9	>12	6.6
Ogi	34	$\geqq 20$	$\geqq 10$	$\geqq 12$	7.2
Kesen	9.9	8.4	8.3	7.1	7.0
Sone	16	10	15	13	>12
Fukutomi	12	9.1	6.6 7.7	7.7	5.8 6.6
Satsuma (Kyusyu)	37	9.7	>20	>12	>12
Nio	54	>20	>20	>12	>12
Kamiomi	7.2	7.8	7.5	4.0	3.9
Nagai	8.2	9.0	7.9	—	—
Numakai	5.9	12	—	—	—
Duwun	17	14	10	11	9.2
Okabe	7.6	8.4	7.5	7.0	6.5
Aomori	51	7.2	—	>12	—
Tomiya	9.0	$\geqq 10$	—	8.2	—
Kokubunji	21	$10\sim19$	—	$\geqq 9$	—

* TAKAOKA *et al.* (1989).

** Ages calculated from ^{53}Mn data by NISHIIZUMI (1978 and 1987).

IZUMI *et al.*, 1980), because measured ratios in Table 5 may not indicate spallogenic neon ratios. The cosmic-ray exposure ages were also calculated by the ^{53}Mn/^{26}Al method proposed by HERPERS and ENGLART (1983). Results are listed in Table 6 together with ^{21}Ne ages. It is said that ^{53}Mn/^{26}Al age has the advantage that the irradiation age of meteorite can be determined independently of depth and size. However, it is only applicable to chondrites having exposure ages $(1\sim12)\times10^{6}$ years. The ^{53}Mn and ^{53}Mn/^{26}Al ages obtained in this work, are generally agree well except the Kamiomi. Compared with ^{21}Ne ages, only the Fukutomi, Numakai and Aomori show some descrepancies. Discussion on these chondrites will be given in the following section.

3.4. *Discussion for individual chodrites*

3.4.1. *Fukutomi*

All ^{53}Mn- and ^{53}Mn/^{26}Al ages calculated based on the published data (NISHIIZUMI, 1978 and 1987) and the present data agree each other, but are about 60% lower than ^{21}Ne age. Since the Fukutomi is gas-rich chondrite, it may say that the present ^{53}Mn and ^{53}Mn/^{26}Al ages give more reliable values than rare gas ages.

3.4.2. *Satsuma* (*Kyushu*)

The present ^{53}Mn data is a slightly lower than that expected from the neon isotope ratio (FIg. 3) and the data measured by NISHIIZUMI (1987) (Table 5). This could be explained by sampling position of the chondrite. The Satsuma fell as a big meteorite shower. The specimens used by TAKAOKA *et al.* (1989) and NISHIIZUMI (1987) were taken from a fragment named Oshima No. 3, while the sample for the present work was taken from a fragment called Hishikari. Oshima and Hishikari used to be the old names of one of the local village and prefecture where the respective Satsuma (Kyushu) chondrite specimens fell (Murayama, 1962). According to BHATTACHARYA *et al.* (1980) and ENGLERT and HERR (1980), production rate of ^{53}Mn is sensitive with preatmospheric size of chondrite. For example, in the chondrite which masses up to ~1400 kg, ^{53}Mn activities increase with depth ~300 to ~550 dpm.

The present different values for ^{53}Mn activity data in both works are attributed to the depth of the location 5~20 cm apart from each other according to Fig. 3 by BHATTACHARYA *et al.* (1980), which was calculated for the St. Séverin with the exposure age, 11.2×10^6 years. Such exposure age indicate that ^{53}Mn activities in chondrites are almost reach secular equilibrium. It is still unknown whether the specimens, Oshima No. 3 is real core sample or not. It could be concluded that a part of specimen of the Satsuma (Kyushu), Hishikari, from which the present sample was taken, was located $\geqq 5$ cm from the surface of preatmospheric body, while other samples taken from Oshima No. 3 were at rather deeper position. Preatmospheric radius and weight of the Satsuma (Kyushu) were estimated as $\geqq 15$ cm and $\geqq 50$ kg, respectively, with lowest limit is just the weight of the recovered mass.

3.4.3. *Kamiomi*

The high ^{22}Ne/^{21}Ne ratio, 1.22 of Kamiomi (Takaoka *et al.*, 1989) suggests that this chondrite was irradiated by cosmic rays near the surface of the parent body or as a part of small meteoroid. The low ^{53}Mn saturation activity agrees with this argument, but ^{26}Al activity does not correlate with the shielding factor of 1.22. It could be rather due to the fact that the sample used for ^{53}Mn and rare gas measurements of Kamiomi was taken from the place close to the preatmospheric surface but that the sample for ^{26}Al determination was not. The 437 g of whole rock was used for the ^{26}Al measurement. If it was placed on the opposit position from the site where specimens for rare gas and ^{53}Mn measurements was taken, obtained data may not represent the values at the surface. Because it is well known that intensity of γ-ray decrease with square of distance from the counter even in vacuum. Further, REEDY (1985) reported very steep gradient to increase spallogenic ^{26}Al production rates. According to his Fig. 4, L-chondrite with preatmospheric radii less than 300 cm, about 30% higher ^{26}Al activities could be obtained in about 20 g/cm^2 interior sample than that at the surface. By such reason, ^{22}Ne/^{21}Ne ratio, 1.22, may not be attribute ^{26}Al data of Kamiomi.

3.4.4. *Numakai*

In the anti-correlation figure of ^{53}Mn with ^{22}Ne/^{21}Ne, Fig. 3, Numakai deviates largely from other chondrites. Exposure ages from ^{21}Ne and ^{53}Mn are different about factor 2. This may be explained by the escape of light rare gases by shock effect from the chondrite. Although it is not described about shock effects on this chondrites (YAGI *et al.*, 1976), their observation indicate the chondrite was subjected to considerable shock in space. In the thin section, major silicate minerals such as olivin and pyroxene are fractured and show waving extinction under microscope. Kamacite grains show deformed Neumann bands due to shock effects.

3.4.5. *Aomori*

Concerning Aomori, we will discuss its irradiation history elsewhere (YABUKI and SHIMA to be published). It is summerized as follows: in spite of its longer ^{21}Ne exposure age, ^{26}Al and ^{53}Mn activities of Aomori are fairly low and the shielding indicators ^{22}Ne/^{21}Ne of 1.133 is not consistent with these low activities. Multi-stage irradiation model may interpret this inconsistency.

4. Conclusion

Cosmogenic long-lived radioactive nuclides, ^{26}Al and ^{53}Mn, in eighteen chondrites which fell and recovered in Japan, were determined by nondestructive γ-ray spectrometry and neutron activation analyses, respectively.

Data were compared with those of spallogenic component of neon (TAKAOKA *et al.*, 1989) obtained from the same chondrite specimens. Generally, the anti-correlation trends of ^{26}Al and ^{53}Mn with ^{22}Ne/^{21}Ne indicate that the chondrites determined were moderate sizes in space and were subjected normal abration processes except the Aomori, Kamiomi, Numakai and the gas-rich chondrites, Fukutomi and Nio.

The ^{26}Al activities of all chondrites except the Aomori, apparently reach secular equilibrium, although those ^{53}Mn activities of some chondrites do not. Thus, cosmic-ray exposure ages were calculated from ^{53}Mn activities and ^{53}Mn/^{26}Al pairs.

The ^{53}Mn and ^{53}Mn/^{26}Al pair ages are generally in good agreement except the Kamiomi. Compared with ^{21}Ne ages, the Fukutomi, Satsuma (Kyushu), Numakai and Aomori show some discrepancies.

For the Fukutomi, ^{53}Mn and ^{53}Mn/^{26}Al ages are about 60% lower than ^{21}Ne age. Since the Fukutomi is gas rich chondrite as mentioned above, the present exposure age is probably more reliable than ^{21}Ne age.

In the case of the Satsuma (Kyushu), the ^{53}Mn data are slightly lower than that expected from the neon isotope ratio and the published data. This is due to the different sampling position of both specimens. Preatmospheric radius and weight of the Satsuma (Kyushu) were estimated as $\geqq$15 cm and $\geqq$50 kg, respectively.

Low ^{53}Mn saturation activity of the Kamiomi agree with its high ^{22}Ne/^{21}Ne ratio, *i.e.* both indicate light shielding in space. On the other hand, ^{26}Al activity does not correlate with the shielding factor. The reason could be the result of the different sampling position, i.e. the samples used for ^{53}Mn and rare gas measurements were taken from the place close to the preatmospheric surface, while ^{26}Al was measured by using whole specimens.

For the Numakai, ^{21}Ne age is lower than ^{53}Mn age. This may be owing to the gas loss by heavy shock of this meteorite in space.

A large deviation from correlation of ^{26}Al and ^{53}Mn with ^{22}Ne/^{21}Ne ratio of the Aomori could be explained by the multi-irradiation model.

5. Acknowledgment

The most of chondirte samples used in this work were from collection of National Science Museum. However, the chondrites, Kamiomi, Nagai, and Shibayama were loaned out by respective posessors to the Museum for display, and the Nogata, Aomori, Tomiya and Kokubunji were lent us by owners for scientific work. They allowed us to take off small pieces for destructive work such as ^{53}Mn determination. We are very much grateful to them. The tiny specimens of the chondrite Ogi were from British Museum Collection exchanged with our collection by Mr. Murayama, the former curator in this Museum.

Thanks are also due to Dr. M. Imamura of the Institute for Nuclear Studies, the University of Tokyo, for providing us the ^{53}Mn standard and for useful advices and suggestions for ^{53}Mn determination of chondrites.

We would like to thank to Prof. Y. Ito, Mr. T. Takano and other personnel of Tokai Branch, Research Center for Nuclear Science and Technology, the University of Tokyo and reactor staff of the Japan Atomic Energy Research Institute.

A part of this work was supported by Grant-in-Aid for Scientific Research B, project No. 62470037 for 1987 granted to one of the authors, M. S., and National Universities' Program for the Commom Use of JAERI facilities 1985 and 1986, the Ministry of Education, Science and Culture.

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