COSMIC RAY INDUCED ^{53}Mn IN YAMATO-7301(j), -7305(k) AND -7304(m) METEORITES

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Abstract: Cosmic ray-induced long-lived nuclide, ^{53}Mn ($t_{1/2}=3.7\times10^6$y) has been determined by the neutron activation analysis in Yamato-7301(j), -7305(k), and -7304(m) meteorites, and the contents were found to be 102 ± 6, 357 ± 19 and 417 ± 22 dpm ^{53}Mn/kg Fe respectively. The content found in Yamato-7304 is in good agreement with other chondrites, and the slightly lower value in Yamato-7305, which is also common in others, may be due to some minor effects. In contrast, the content of ^{53}Mn in Yamato-7301 is remarkably low, lower than that of any other stone meteorites of relatively long exposure age so far studied. Several possible mechanisms are discussed to explain the puzzling ^{53}Mn activity in Yamato-7301: 1) Long terrestrial age of about 7 m.y., and the decay of ^{53}Mn. 2) Heavy preatomospheric shielding of more than 70 cm, and low production rate of ^{53}Mn. 3) Multistage irradiation history, and undersaturation of ^{53}Mn. 4) A combined mechanism of the above two or three factors.

1. Introduction

Many cosmic ray-induced radioactive and stable nuclides, occasionally called "cosmogenic", have been studied in meteorites and lunar samples. The cosmogenic nuclides in lunar samples permit us to study the time-variation of solar and galactic cosmic rays at known location (1 A.U.) in the solar system, the exposure age and surface erosion of lunar rocks, and mixing in the lunar regolith. Studies in meteorites, on the other hand, primarily inform us of meteorite formation in space and their preatmospheric size, in addition to the time variation of cosmic rays. Among long-lived nuclides, ^{59}Ni ($t_{1/2}=8\times10^4$ y), ^{81}Kr (2.1×10^5 y), ^{36}Cl (3.0×10^5 y), ^{26}Al (7.2×10^5 y), ^{10}Be (1.6×10^6 y) and ^{53}Mn ($3.7\times$

10^6 y) have been measured extensively in these extraterrestrial materials. In particular, ^{53}Mn is a useful nuclide for such purposes because of its long half-life—the longest next to ^{40}K, the high production yield from abundant iron target by cosmic ray interaction such as $^{56}Fe(p, \alpha)$, $Fe(p, x)$, $Fe(n, x)$ and $^{54}Fe(n, 2n)$ reactions (FURUKAWA and SHIZURI, 1972; GENSHO *et al.*, 1972), and also its high sensitivity to the detection as described below.

In addition to the above-mentioned topics, there is particular significance in detecting the cosmic ray-produced long-lived activities in the Yamato meteorites from the Antarctic. These meteorites are expected to be confined in the Antarctic ice for a long period of time before they appeared to the surface and were collected recently. The terrestrial ages, *i.e.*, the time elapsed after the fall of Yamato meteorites, can give significant clues to the accumulation mechanism and geological history of the Antarctic ice. Furthermore, measurements of cosmogenic nuclides in each of the collected meteorites can distinguish the individual fall and clarify the possibility of meteorite shower.

SUESS and WÄNKE (1962), and GOEL and KOHMAN (1962) reported the terrestrial ages of stony meteorites inferred from cosmogenic ^{14}C(5736 y) measurements. It is expected that some of the Yamato meteorites in the Antarctic ice may be older in terrestrial age than the mean life of ^{14}C. Hence, the longer-lived radioisotopes such as ^{26}Al, ^{53}Mn and some others might be better candidates for the study. The radioactivity of ^{26}Al in stony meteorites can be easily determined by non-destructive $\gamma-\gamma$ coincidence counting, but this method requires a large amount of sample, usually 10 g at least. On the other hand, a very sensitive method exists for ^{53}Mn based on the conversion of ^{53}Mn to $^{54}Mn(t_{1/2}=312$ d, 834.8 keV γ-ray emitter) through $^{53}Mn(n, \gamma)$ ^{54}Mn by neutron activation (MILLARD, 1965). The thermal neutron cross-section of $\sigma=82\pm7$ b has been obtained on the basis of the half-life of ^{53}Mn, 3.7×10^6 y, determined by HONDA and IMAMURA (1971). By this technique, less than 10^{-4} dpm of ^{53}Mn can be determined by irradiation with a total flux of more than 3×10^{19} neutrons/cm^2 in a reactor. The convenient sample size is usually less than 1 g for chondrites. This method, however, requires chemical and radiochemical separations of Mn and ^{54}Mn from the samples before and after neutron irradiation respectively, and a long irradiation time at a well-thermalized position in a reactor. In the present study, this method was applied for determination of ^{53}Mn in gram size samples of Yamato-7301, -7305 and -7304 chondrites.

2. Experimental

1.22, 1.02 and 1.08 g of Yamato-7301, -7305 and -7304 respectively were analyzed for the ^{53}Mn contents. The chemical and counting procedures used in this work have been described elsewhere (NISHIIZUMI *et al.*, 1977). The samples were irradiated in the VG-7-6 hole of JRR-3 reactor of the Japan Atomic Energy Research Institute, Tokai, Ibaraki, for 528 h 16 m with a total thermal neutron flux of 4.5×10^{18} n/cm^2, determined on the basis of monitor reaction $^{59}Co(n, \gamma)$

^{60}Co, $\sigma=37$ b. The ^{54}Mn countings were performed by two Ge(Li) detectors. The counting efficiency for ^{54}Mn photo peak was 2.33% and the B.G. was 0.031 cpm (834.8 ± 1.4 keV) by Princeton Gamma-Tech Ge(Li) detector, and 2.24% and 0.059 cpm (834.8 ± 2.5 keV) by Horiba Ge(Li) detector.

3. Results and Discussion

The chemical compositions of Yamato-7301, -7305 and -7304 are given in Table 1. Al, Mn, Fe and Ni were determined by atomic absorption spectroscopy. Fe, Co and Ni were also determined by neutron activation analysis using the TRIGA Mark II reactor of Rikkyo University. The results for ^{53}Mn in the Yamato meteorites are presented in Table 2 together with the rare gas exposure ages. Some typical data of ^{53}Mn in 6 other meteorites, Kesen (H4), Holbrook (L6), Abee (E4), Crab Orchard (mesosiderite) and Carbo (iron) are also given in the same table for a comparison with the Yamato meteorites. The overall uncertainties listed in terms of dpm ^{53}Mn/kg Fe were estimated by quadratically adding the uncertainties of 2% for Mn concentration, 3% for Fe

Table 1. Chemical composition of Yamato-7301, -7305 and -7304 meteorites.

Name of sample	Class	Al (%)	Mn (ppm)	Fe (%)	Co (ppm)	Ni (%)
Yamato-7301 (j)	H4	1.09	2310	25.4	700	1.56
Yamato-7305 (k)	L5	1.25	2630	22.9	640	1.47
Yamato-7304 (m)	L5	1.17	2700	22.5	590	1.30

Table 2. ^{53}Mn in meteorites.

Name of sample	Class	Recoverd mass (kg)	dpm ^{53}Mn /kg Fe obs.	dpm ^{53}Mn /kg Fe after Ni correction	Exposure age ^{3}He	^{21}Ne (my)	^{38}Ar	dpm ^{53}Mn /kgFe saturated
Yamato-7301 (j)	H4	0.65	104 ± 6	102	8.2[a]	13[a]	13[a]	112
Yamato-7305 (k)	L5	0.90	364 ± 19	357	17[a]	22[a]	28[a]	363
Yamato-7304 (m)	L5	0.50	425 ± 22	417	13[a]	18[a]	17[a]	432
Kesen	H4	135	358 ± 23	347	6.1[b]			510
Holbrook	L6	235	429 ± 25	421	12[c]	18[c]		436
Abee	E4	107	338 ± 18	329	7[c]	7[d], 8[c]	7[d]	451
Crab Orchard	meso-siderite	49	541 ± 29	521	64[e] (^{36}Cl-^{36}Ar)			521
Carbo	iron	454	403 ± 25	390	850[f] ($^{41}K/^{40}K$)			390

a) TAKAOKA and NAGAO (1977).
b) EBERHARDT *et al.* (1966).
c) HINTENBERGER *et al.* (1964).
d) SPANNAGEL and HEUSSER (1969).
e) BEGEMANN *et al.* (1976).
f) VOSHAGE (1967).

concentration and 2% for the conversion factor by neutron activation, dpm ^{54}Mn/dpm ^{53}Mn, to the statistical counting error ($\pm 2\ \sigma$). But their absolute values do not include the uncertainty of ^{53}Mn standard extracted from the Bogou meteorite, which may be estimated at $\pm 5\%$ ($\pm 1\ \sigma$). In the fourth column of Table 2, the specific activity of ^{53}Mn in Fe is not corrected for a small contribution from Ni target. From the data of ^{53}Mn measured in high and low Ni/(Fe+Ni) phases in the St. Séverin meteorite, the ratio of ^{53}Mn production rate in Ni to that in Fe has been estimated at 0.3 $\pm$0.1 (NISHIIZUMI *et al.*, 1977). The fifth column of Table 2 gives the corrected specific activities based on the Ni content of each sample. The specific activity is reduced by 2% in the chondrites, and about 3% in the mesosiderite and iron meteorites. The shorter ^{3}He exposure age of the Yamato meteorites, as compared with ^{21}Ne and ^{38}Ar exposure ages, may be explained by He loss (TAKAOKA and NAGAO, 1977). In the last column of Table 2, the saturation specific activity has been calculated from the respective exposure ages and values after Ni correction. The saturation activity is equal to the production rate at equilibrium for long irradiation, sufficiently longer than the mean life of ^{53}Mn, by cosmic ray. The content of ^{53}Mn in Yamato-7304 has a normal level of 417 dpm ^{53}Mn/kg Fe, comparable with other meteorites. Yamato-7305 has a slightly lower activity level of 357 dpm ^{53}Mn/kg Fe. Yamato-7301, on the other hand, has an extremely low value, 102 $\pm$6 dpm ^{53}Mn/kg Fe. It may be mentioned that such a low ^{53}Mn activity has never been obtained before in stones excepting the Brenham pallasite (HONDA and ARNOLD, 1964) and the Bondoc mesosiderite (CRESSY and SHEDLOVSKY, 1965) which perhaps had extremely large preatmospheric size. Because of this unusual observation we tried to obtain some preliminary data of ^{10}Be activity in Yamato-7301 and -7305 meteorites using their Be fractions and employing a low background (BG = 0.004 cpm) needle GM counter (FUJITA *et al.*, 1975) established in the Institute for Nuclear Study, The University of Tokyo, Low Background Cell (TANAKA *et al.*, 1977). Yamato-7301 seemed to have a lower ^{10}Be content, 9$\pm$3 and Yamato-7305 15$\pm$5 dpm ^{10}Be/kg meteorite ($\pm 1\ \sigma$). Unfortunately, no simple explanation for the low ^{53}Mn activity determined in Yamato-7301 meteorite correlated to rare gases and ^{10}Be data can be offered at the present time. Although the data in Yamato-7305 is at a rather normal level among stony meteorites, because of the smallness of sample size, larger errors might interfere any direct comparison with the ^{53}Mn. In the following sections, we discuss several possible mechanisms to explain the low ^{53}Mn in Yamato-7301. Incidentally, the effects described in 3–1 through 3–3 are known among iron meteorites but not for stone meteorites.

3.1. Long terrestrial age

First possible explanation may be that Yamato-7301 has undergone a very long terrestrial age in the Antarctic ice to such an extent that most of ^{53}Mn has been decayed. Under this assumption a terrestrial age of 7$\pm$1 m.y. can be calculated by assuming 400$\pm$50 dpm ^{53}Mn/kg Fe for the saturation activity in chondrites. Even under the assumption that our sample came from very near the surface of a small meteorite, like the Harleton chondrite studied by HONDA and ARNOLD (1964), at least 4 m.y. will be resulted. No other stone meteorite of such a long terrestrial age has been reported up to date. A long age of *ca.* 2 m.y. (CHANG and WÄNKE, 1969), was reported for the Tamarugal iron meteorite. In the Tamarugal, 155 $\pm$20 dpm ^{53}Mn/kg meteorite (IMAMURA *et al.*, 1969), 163$\pm$8 dpm ^{53}Mn/kg meteorite (HERPERS *et al.*, 1969), and 182 $\pm$7 dpm ^{53}Mn/kg Fe (NISHIIZUMI *et al.*, 1977) were found. Based on the ^{53}Mn data a period of 4 m.y. may be more realistic for the Tamarugal. Assuming the saturation activity of ^{10}Be in ordinary stone meteorite as 20 dpm ^{10}Be/kg meteorite, from ^{10}Be data, 9 $\pm$3 dpm/kg met., the terrestrial age of Yamato-7301 may be estimated between 2 and 3 m.y. Viewed from another angle, Yamato-7301 is expected to have about 1 dpm ^{10}Be/kg meteorite and no detectable ^{26}Al activity if the terrestrial age is really in the vicinity of 7 m.y.

The accumulation mechanism of the Yamato meteorites has been tentatively visualized as follows: They were transferred with ice flow from the point of fall to the edge of the bare ice field and were elevated to the surface by evaporation and upward movement of the ice (NAGATA, 1977). The horizontal flow rate of ice at the surface was measured to be a few meters/y near Yamato Mountains where the Yamato meteorites were collected (SHIRAISHI *et al.*, 1976). If the flow rate of ice at deeper parts is of the same order as that of the surface, the long age requires $n \times 10^4$ km which is one order larger than the linear size of Antarctica. On the other hand, the evaporation rate is nearly equal to the uprising rate of ice which is a few cm/y in the bare ice area. Assuming that the transfer rate is the same as the uprising rate Yamato-7301 could have fallen within a distance of a few hundred km about 7 m.y. ago. Another pertinent point to be considered is the shape and the collection site of Yamato-7301 which is different from those of other Yamato meteorites. It was collected near the confluence of moraines about 1,850 m above sea level whereas other Yamato meteorites were collected at the elevations of 2,000 to 2,200 m (SHIRAISHI *et al.*, 1976). Also it is not covered with fusion crust and has a smooth brownish surface. These facts are suggestive of an extensive weathering effect and a longer terrestrial age. If, however, we take the ^{10}Be data seriously, low ^{53}Mn in Yamato-7301 could not be attributed to the terrestrial age alone.

3.2. *Low production rate in a large preatmospheric body*

The observed low activity of ^{53}Mn in Yamato-7301 meteorite could be attributed also to a depth effect or absorption of secondaries under a constant cosmic ray irradiation. As the galactic cosmic rays penetrate into a meteoroid, low energy secondary particles—mainly neutrons—are produced and become effective for nuclear reactions. HONDA (1962) measured the distribution of spallation products in a thick iron target bombarded by 3 GeV protons and calculated the production rate as a function of depth to be compared with the meteorite data. KOHMAN and BENDER (1967) and TRIVEDI and GOEL (1969)

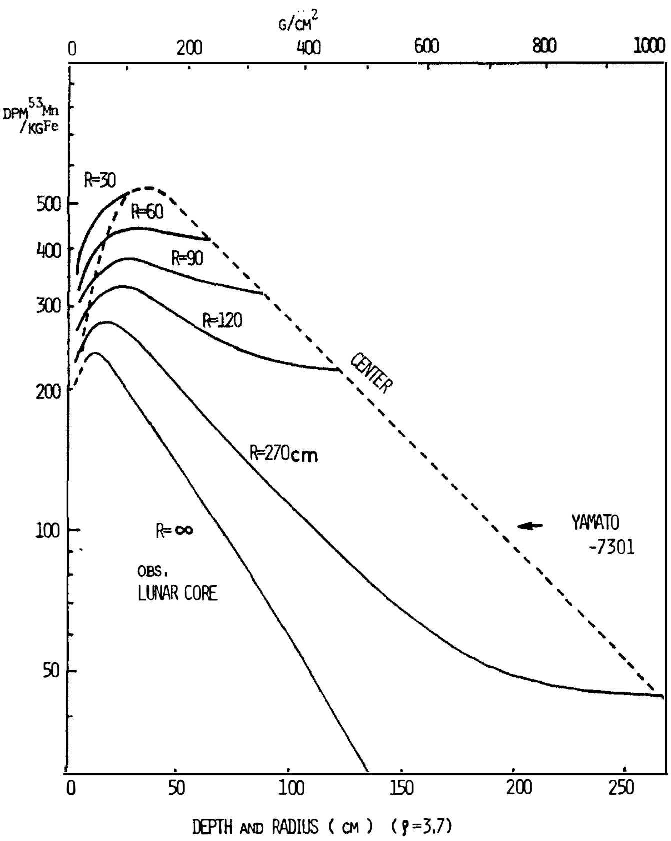

Fig. 1. Estimated ^{53}Mn contents in spherical pre-atmospheric meteoroids of various radii (R) and observed ^{53}Mn depth profiles in Apollo 15, 16 deep drill cores (IMAMURA et al., 1973, 1974). Observed activity level in Yamato-7301(j) is shown.

also extended the calculation using thick target bombardment data. REEDY and ARNOLD (1972) calculated the production rate of radionuclides produced by bombardments of galactic cosmic rays and solar cosmic rays in the case of the lunar surface. Their calculations were done for a semi-infinite slab with 2 π isotropic flux. Recently, ^{53}Mn in the lunar core, from surface to a depth of 400 g/cm^2, has been measured (IMAMURA *et al.*, 1973, 1974; NISHIIZUMI *et al.*, 1976). The level of ^{53}Mn activity in the lunar soil due to galactic cosmic rays is about 40% higher than the estimation by REEDY and ARNOLD (1972), but the shape of depth profile is in good agreement with the model. Therefore, we can estimate directly the variation of production rate of ^{53}Mn in spherical (pre-atmospheric) meteoroid based on our measurements of ^{53}Mn in Apollo 15, 16 deep drill cores. This estimation could be performed by making some modifications of the thick target result. The depth profiles of production of ^{53}Mn in spherical bodies of various radii are shown in Fig. 1, where absolute values of near maximum are reduced from depth profiles and published data of ^{53}Mn found in iron and stony iron meteorites. The values are variable to some extent, 20% or so, most probably due to orbital histories in space. For depths below about 100 g/cm^2, the production rate decreases exponentially with increasing depth with half attenuation length of about 150 g/cm^2. The low ^{53}Mn value of Yamato-7301 could suggest that it has been heavily shielded in large preatmospheric mass. With the assumption of a spherical preatmospheric body, the minimum radius, therefore the maximum possible depth, is calculated to be 190 cm corresponding to a mass of 106 tons, when the sample is located near the center of the body. On the other hand, a minimum depth of 67 cm from the surface is obtained by assuming the semi-infinite case. Among irons and stony-irons, shielding effects are common. For example, the Bondoc mesosiderite is well known for its large preatmospheric size. CRESSY and SHEDLOVSKY (1965) and BORN and BEGEMANN (1975) estimated a preatmospheric radius of more than 200 cm ($\rho=5$ g/cm^3) for this meteorite from the data of ^{10}Be–^{26}Al and ^{14}C–^{39}Ar correlations respectively. We also estimated a radius of more than 190 cm based on 48 ± 3 dpm ^{53}Mn/kg Fe in the Bondoc (NISHIIZUMI *et al.*, 1977). ^{10}Be in Yamato-7301 is expected to be about 3 dpm ^{10}Be/kg meteorite with the half attenuation length of ^{10}Be (106 g/cm^2) estimated from REEDY and ARNOLD (1972) and by assuming usual activity level of ^{10}Be in stone. The data of ^{10}Be in Yamato-7301 seem rather high compared with the values expected in this model. ^{26}Al in Yamato-7301 is expected to be 12 dpm ^{26}Al/kg meteorite using the normal activity level of 60 dpm/kg meteorite.

We estimate that the production rate of ^{21}Ne in the deep part of a preatmospheric body may have decreased to about 1/5 of an ordinary level. With this estimate, the exposure age is calculated to be about 60 m.y. based on published value of the ^{21}Ne content (TAKAOKA and NAGAO, 1977). WRIGHT *et al.* (1973) measured the cosmogenic rare gas concentration and ratios of $^{3}He/^{21}Ne$ and $^{22}Ne/^{21}Ne$ as a function of depth in the Keyes chondrite. $^{22}Ne/^{21}Ne$ in Yamato-7301 by TAKAOKA and NAGAO (1977) seems equal to the value of ordinary small size chondrites. However, their values are not applicable for the test because none of stony meteorite data of deeper than 100 cm are available except for the Bondoc.

To resolve the problem the determination of ^{26}Al in Yamato-7301 must be useful. Besides, the track density of heavy primary cosmic rays has to be measured for a test of this model. If both ^{26}Al and ^{10}Be are higher and a significant density of tracks is observed, the ^{53}Mn in Yamato-7301 could not be explained by shielding.

3.3. *Undersaturation due to multi-stage irradiation history*

Not many but a few meteorites are supposed to have complex irradiation records. The Sikhote-Alin (VILCSEK and WÄNKE, 1961), the Serra de Magé eucrite (FUSE and ANDERS, 1969; CARVER and ANDERS, 1970), the Pitts octahedrite (BEGEMANN *et al.*, 1970) and the Weston chondrite (SCHULTZ *et al.*, 1972) are such examples. Low ^{53}Mn in Yamato-7301 can also be explained

Table 3. Comparison of the models to explain low ^{53}Mn in Yamato-7301(j).

Model	dpm ^{53}Mn/kg Fe		Terrestrial age (m.y.)	Radius and depth		Expected* dpm/kg met.		Exposure age (m.y.)	
	Present	At time of fall		(cm)	(cm)	^{10}Be	^{26}Al		
1. Long terrestrial age	102	365	7	normal		1	0	13	
2. Heavy shielding	102	102	0	>190	67~190	3	12	60	
		after first stage						first stage	second stage
3. Two-stage irradiation		102 (0)	0	very large		9	44	long	1.4
	102	102 (30)	0	>300	135~300	7	38	240	1.0
		102 (68)	0	>230	90~230	5	27	100	0.5

* Assuming the normal activity levels of 20 dpm ^{10}Be/kg meteorite and 60 dpm ^{26}Al/kg meteorite.

by a model of two-stage cosmic ray exposure history (a simplified version of multi-stage model), as follows. In the first stage, it has been heavily shielded in a large parent body for a period of long time as described above. Most of the cosmogenic stable nuclides might have been produced during this period. At the second stage, after the parent body was broken Yamato-7301 was a part of a small size secondary body. It was irradiated with the galactic cosmic rays for a shorter time so that ^{53}Mn was not saturated to the value for the present location. The irradiation time of the second stage may have ranged from 0 to 1.4 m.y. The activities expected ^{10}Be and ^{26}Al in such cases are shown in Table 3. The comparison suggests that in the first stage the meteorite was deeper than a few meters from the surface of the parent body during a period of more than 100 m.y. In the second stage, it was expelled from that location and irradiated at a normal level of cosmic-ray flux for the last ≤ 1.4 m.y. As a result, long-lived nuclides such as ^{53}Mn, ^{10}Be and ^{26}Al are not saturated.

A combination of the above three mechanisms may be also considered. At any rate it is desirable to determine quantitatively some of the ^{26}Al, ^{36}Cl, ^{81}Kr, ^{59}Ni and ^{14}C for solving the puzzle of low ^{53}Mn in Yamato-7301 in future.

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