COSMIC RAY PRODUCED RADIONUCLIDES IN ANTARCTIC METEORITES

Kunihiko NISHIIZUMI*,

Department of Chemistry, College of Science, Rikkyo University,
Nishi-ikebukuro 3-chome, Toshima-ku, Tokyo 171

Mineo IMAMURA

Institute for Nuclear Study, University of Tokyo,
Midori-cho 3-chome, Tanashi-shi, Tokyo 188

and

Masatake HONDA

The Institute for Solid State Physics, University of Tokyo,
Roppongi 7-chome, Minato-ku, Tokyo 106

Abstract: Cosmic ray produced ^{53}Mn ($t_{1/2}=3.7\times10^6$y) has been determined in seventeen antarctic meteorites, among which, two have extremely low contents of ^{53}Mn: 101 ± 6 dpm/kg Fe in Yamato-7301 and 22 ± 3 in Allan Hills No. 8. Even after corrections are made for undersaturation based on published values of rare gas ages, saturation specific activity of ^{53}Mn in these two meteorites is abnormally low compared with the average value of 470 ± 100 dpm/kg Fe in other antarctic meteorites. ^{26}Al ($t_{1/2}=7.2\times10^5$y) and ^{10}Be ($t_{1/2}=1.6\times10^6$y) were also measured in these two meteorites and in several others. These results as well as those of rare gas isotopes are discussed in terms of terrestrial ages of the meteorites, shielding effects in the parent bodies, undersaturation of the radioactivities and multi-stage irradiation histories. These two meteorites apparently experienced a two-stage exposure history with a long preirradiation ($>10^8$y) under heavy shielding in a large body before the recent brief exposure ($\lesssim2\times10^6$y) in a much smaller body. These facts seem to illustrate the complex nature of the exposure histories of most meteorites. The consequence of these multi-stage exposures is also discussed. Terrestrial ages, estimated for a few antarctic meteorites from the ^{26}Al/^{53}Mn ratio, seem to be younger than 3×10^5 years.

1. Introduction

Cosmic ray produced nuclides can provide information on cosmic rays themselves and also on the exposure histories of meteorites. Time-space-variation

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[*] Present address: Department of Chemistry, University of California, San Diego, La Jolla, California 92093, U.S.A.

of cosmic rays, meteorite formation in space and pre-atmospheric size of meteorites have been the major concern of many investigators. In addition to the above areas of study, measurements of terrestrial ages of antarctic meteorites, *i.e.*, the time elapsed after the fall of the objects, would offer information about the accumulation mechanism and geological history of the ice sheet in the regions where they were found. Furthermore, measurements of cosmogenic nuclides in each of the collected meteorites can distinguish individual falls and clarify the possibility of meteorite showers.

Radioactivity which can be usefully investigated may, however, be limited to long-lived radioisotopes such as ^{81}Kr($t_{1/2}=2.1\times10^5$y), ^{36}Cl(3.1×10^5y), ^{26}Al(7.2×10^5y), ^{10}Be(1.6×10^6y), ^{53}Mn(3.7×10^6y) and others. Recently FIREMAN (1978) tried to detect ^{14}C(5736y) in the antarctic meteorites, Allan Hills Nos. 5, 6 and 8, but obtained only the upper limits. Radioactivity with half-life less than $\sim10^4$ years may have decayed away to an undetectable level. On the other hand, to understand the antarctic meteorites, more systematic investigation of various cosmogenic nuclides is needed because of the complication induced by their possibly long and variable terrestrial ages. Among the above nuclides, ^{53}Mn is the most sensitive, it can be detected in samples less than 1 gram, and it has the longest half-life. From our previous study (NISHIIZUMI *et al.*, 1978) we have learned that the terrestrial age of some antarctic meteorites is not long enough to decrease the ^{53}Mn content appreciably. Therefore, useful data are expected from the systematic investigation of ^{53}Mn and other nuclides, including cosmogenic rare gas isotopes. Activity of ^{36}Cl or other shorter lived radionuclides, relative to ^{53}Mn values, may give the most definite values for terrestrial ages. If any deviation is found from the theoretical correlation between ^{53}Mn content and exposure age it could indicate a time variation of cosmic ray intensity during the last several million years. The ^{53}Mn content lower than the usual saturation level can be explained either by heavy shielding from cosmic rays or by a short exposure age. If low ^{53}Mn activity is found, data on at least two other radioisotopes will be necessary to clarify the situation, because of the possibility of a long terrestrial age. Concentrations of cosmic ray produced stable nuclides, such as rare gas isotopes, also have to be known, since there is a possibility of a two or multi-stage irradiation history for the meteorite (FUSE and ANDERS, 1969; NISHIIZUMI *et al.*, 1978; NISHIIZUMI, 1978). Such a complex exposure has been predicted by the model of ARNOLD (1964, 1965a, 1965b) for the formation of stone meteorites.

In the previous work we reported the determination of ^{53}Mn in three Yamato meteorites. In this research, we have extended the ^{53}Mn analysis to fourteen other antarctic meteorites: 5 Yamato, 2 Mt. Baldr and 7 Allan Hills meteorites. Also, ^{10}Be and ^{26}Al were determined in several meteorites including Yamato-7301(j) and Allan Hills No. 8, which were found to have very low ^{53}Mn contents. Data, including those of rare gas isotopes by TAKAOKA and NAGAO (1978), NAGAO and TAKAOKA (1979) and SCHULTZ (1978) and those of ^{26}Al by FRUCHTER and EVANS (1977), are discussed in terms of terrestrial age, shielding effects in the meteorite body, undersaturation of radioactivity, and multi-stage irradiation history.

2. Experimental and Results

Antarctic meteorite samples from 0.5 to 1.0 g were used for the determination of ^{53}Mn by the neutron activation method. The chemical procedures were essentially the same as in the previous work with lunar samples and meteorites (IMAMURA *et al.*, 1973, 1974; NISHIIZUMI *et al.*, 1976, 1978; NISHIIZUMI, 1978). The Mn extracts were irradiated in the VG-7–6 hole of JRR3 reactor of the Japan Atomic Energy Research Institute, Tokai, Ibaraki, for 267.5 hours for a total thermal neutron fluence of 2.5×10^{18} n/cm^2, determined on the basis of the monitor reaction ^{59}Co(n, γ)^{60}Co, $\sigma = 37$b. The ^{54}Mn counting was performed using two Ge(Li) detectors the same as those described by NISHIIZUMI *et al.* (1978). Detailed data on the determination of ^{53}Mn are presented in Table 1.

^{10}Be was determined in several meteorites: (Yamato-7301, -7304, -7305 and -74155) using the fraction of Be separated during the manganese chemistry (the 3–4 column volume fraction of 1N HCl eluted during the cation exchange separation; see IMAMURA *et al.*, 1973). A total of 100 μg Be carrier was added before dissolution to each sample. Be fractions of duplicate samples were combined. The purification steps for Be were essentially the same as the procedure described in MERRIL *et al.* (1960), TANAKA *et al.* (1977) and INOUE and TANAKA (1978). In addition to these samples, 10.2 g and 10.7 g of Yamato-7301 and -7304 were ground to the size finer than $\sim$50 mesh and separated into non-magnetic and magnetic fractions. ^{10}Be was determined in 8.73 g and 9.06 g, respectively, of the non-magnetic fractions. These fractions were also used for the determination of ^{26}Al. The magnetic fractions were kept for the analysis of rare gas isotopes. ^{10}Be activity was determined employing low background needle GM counters (FUJITA *et al.*, 1975) and a Si(Li) spectrometer located in the Low Background Cell at Institute for Nuclear Study, University of Tokyo. The detailed counting data are shown in Table 2.

The data of ^{26}Al activity in the Yamato-7301 and -7304 meteorites were obtained from the non-magnetic fractions by a low background β-γ counting method using the detector system at the University of California, San Diego (SHEDLOVSKY *et al.*, 1970; FINKEL *et al.*, 1971).

The concentrations of Al, Mn, Fe, Co and Ni in the meteorites are given in Table 3. Al, Mn, Fe and Ni were determined by atomic absorption spectroscopy. Co was determined by neutron activation analysis (NAA) using the Triga Mark II reactor at Rikkyo University. Mn, Fe and Ni were also determined by NAA and

Table 1. Results on neutron activation analysis of ^{53}Mn.

Sample	Sample description				Sample counted after irradiation			^{54}Mn[#2)] (dpm/mg Mn)	After (*n*, 2*n*) correction ^{54}Mn (dpm/mg Mn)	^{53}Mn (dpm/kg Fe)	After Ni and Mn correction ^{53}Mn[#3)] (dpm/kg Fe)
	wt. (g)	Fe (%)	Mn (ppm)	Ni (%)	Fe (μg)	Mn (mg)	^{54}Mn[#1)] (dpm)				
Yamato-											
74013	0.949	13.9	4040	<0.005	0.7	3.38	53.7±3.2	15.87±1.00	12.62±1.00	413±38	401±37
74097	1.498	12.7	4130	<0.005	1.0	5.37	81.3±4.5	15.13±0.89	11.88±0.89	435±39	421±38
74155 (1)	1.056	29.8	2360	2.02	1.2	2.07	66.6±3.7	32.12±1.90	28.87±1.90	257±21	249±20
(2)	0.799	24.4	2520	1.62	2.2	1.80	47.6±2.7	26.34±1.59	23.09±1.59	268±22	260±21
74191 (1)	1.285	18.9	2910	1.00	0.9	3.32	103.3±4.8	31.09±1.57	27.84±1.57	482±35	467±34
(2)	1.126	19.7	2810	1.11	0.8	2.78	86.5±3.8	31.10±1.50	27.85±1.50	447±32	433±31
74647 (1)	0.894	24.4	2600	1.70	1.1	2.06	64.7±2.9	31.37±1.54	28.12±1.54	337±24	326±23
(2)	0.918	25.7	2600	1.86	1.3	2.12	71.1±3.2	33.49±1.65	30.24±1.65	344±25	333±24
Mt. Baldr											
No. 1	1.119	25.3	2550	1.63	0.9	2.52	98.6±6.2	39.10±2.58	35.85±2.58	406±35	394±34
No. 2	1.000	26.4	2460	1.82	0.9	2.21	84.4±4.0	38.16±1.96	34.91±1.96	366±27	355±26
Allan Hills											
No. 1	1.143	21.4	2640	1.33	0.8	2.55	92.5±4.3	36.25±1.84	33.00±1.84	458±34	443±33
No. 3 (1)	0.775	21.1	2700	1.26	0.6	1.86	63.4±3.3	34.06±1.90	30.81±1.90	443±34	429±33
(2)	0.803	20.9	2720	1.20	1.0	1.95	66.1±4.4	33.86±2.36	30.61±2.36	448±41	434±40
No. 5	0.982	15.4	4660	<0.005	0.5	3.95	59.5±3.5	15.05±0.94	11.80±0.94	402±37	390±36
No. 6	1.062	27.7	2270	1.76	0.5	2.14	115.2±5.7	53.81±2.87	50.56±2.87	466±34	453±33
No. 7	1.241	21.4	2740	1.14	0.6	3.03	81.9±4.1	27.01±1.46	23.76±1.46	342±27	332±26
No. 8 (1)	1.177	25.5	2410	1.68	0.7	2.56	13.91±1.00	5.41±0.41	2.16±0.42	23±4.6	22±4.5
(2)	1.166	25.6	2380	1.68	0.5	2.45	13.45±0.97	5.47±0.41	2.22±0.42	23±4.6	22±4.5
No. 9	1.002	21.2	2750	1.22	1.9	2.47	91.5±3.9	36.98±1.74	33.73±1.74	492±34	477±33

Table 1 (Continued).

^{53}Mn standard	Solution (g)#4)						dpm^{54}Mn/dpm^{55}Mn#5)
^{53}Mn-1	0.2994	0.3	0.338	233.1± 8.9	690±30	687±30	889±28#6)
^{53}Mn-2	0.3253	0.6	0.365	257.6±10.8	706±33	703±33	
^{55}Mn standard	Mn metal (mg)						
^{55}Mn-1	8.73	0.1	8.49	29.19±1.65	3.44±0.21	3.25±0.09#6)	
^{55}Mn-2	11.39	0.1	11.00	35.18±1.79	3.20±0.10		
Fe standard	Fe metal (mg)	Chemical yield (%)			dpm^{54}Mn/μg Fe		
Fe-1	3.53	96.3		273±18	0.080±0.006	0.078±0.005#6)	
Fe-2	3.56	98.4		262±24	0.075±0.007		

#1) Weighted averages. All activities have been corrected to September 23, 1977, the end of irradiation. The uncertainties include 2 σ counting statistics plus a 3 % uncertainty in the geometrical reproducibilities of the counting sample. The uncertainty in the absolute counting efficiency is not included.

#2) Corrected for ^{54}Fe(n, p)^{54}Mn. A 2 % uncertainty for the Mn yield has been added quadratically to the counting error.

#3) The following uncertainties have been added quadratically: 2.5 % each for the Mn and Fe concentration and the percentage uncertainty from the previous column.

#4) Standard solution, "B-II"; 0.781 dpm ^{53}Mn/mg Mn and 1200 ppm Mn.

#5) The 5 % uncertainty in the original standardization of the ^{53}Mn has not been included.

#6) Weighted average.

Table 2. Low level counting of ^{10}Be.

Meteorite	Sample wt. (g)	Counter***	Counting time (min.)	Net count rate (cpm)	^{10}Be (dpm/kg)	Remarks
Yamato-7301	1.218	Needle-GM	6966	0.0048±14	9±3	b.g. ~~=0.0040~~ cpm
		Needle-GM	5775	0.0043±18		
	8.73*	Needle-GM	2736	0.0341±39	9±1**	
		Needle-GM	4320	0.0400±37		b.g. =0.0118 cpm
Yamato-7304	1.084	Needle-GM	7245	0.0083±18	18±4	
	9.06*	Si(Li)	2736	0.0347±49	20±2**	160–555 keV
		Si(Li)	4200	0.0469±47		80–555 keV
Yamato-7305	1.020	Needle-GM	6966	0.0059±17	15±5	
Yamato-74155	1.855	Needle-GM	2736	0.0063±23	9±3	

*Non-magnetic fraction.

**4 dpm/kg Fe (saturated value) was assumed for ^{10}Be production from Fe.

***Two sets of needle-type GM counters and one Si(Li) spectrometer were used for the counting. Background and absolute counting efficiency were 0.0072–0.0090 cpm and 47±3 % for needle-type GM counters, and 0.039 cpm and 35±2% (80–555 keV) for Si(Li), respectively, unless otherwise stated. Final data include the uncertainties in absolute counting efficiency as well as 1 σ counting statistics.

Table 3. Chemical composition of antarctic meteorites.*

Meteorite		Class	Al (%)	Mn (ppm)	Fe (%)	Co (ppm)	Ni (%)	Remarks
Yamato	-7301(j)	H4	1.09	2310	25.4	700	1.56	Nishiizumi *et al.*, 1978
	-7304(m)	L5	1.17	2700	22.5	590	1.30	〃
	-7305(k)	L5	1.25	2630	22.9	640	1.47	〃
	-74013	Diogenite	0.53	4040	13.9	37	<0.005	
	-74097	Diogenite	0.34	4130	12.7	-	<0.005	
	-74155	H4	1.18	2430	27.5	810	1.85	
	-74191	L3	1.33	2860	19.3	450	1.05	
	-74647	H5	1.20	2600	25.1	710	1.78	
Mt. Baldr	No. 1	H	1.20	2550	25.3	730	1.63	
	No. 2	H	1.14	2460	26.4	780	1.82	
Allan Hills	No. 1	L	1.22	2640	21.4	540	1.33	
	No. 3	L	1.22	2710	21.0	530	1.23	
	No. 5	Eucrite	5.47	4660	15.4	6±3	<0.005	
	No. 6	H	1.12	2270	27.7	880	1.76	
	No. 7	L	1.22	2740	21.4	580	1.14	
	No. 8	H	1.17	2400	25.6	740	1.68	
	No. 9	L	1.27	2750	21.2	590	1.22	

* Errors are estimatied to be 2.5 % for Mn and Fe, 3 % for Al and Ni and 5 % for Co.

Table 4. ^{53}Mn, ^{10}Be and ^{26}Al in antarctic meteorites.

Meteorite	Class	Recovered mass (kg)	dpm ^{53}Mn* /kg Fe	dpm ^{10}Be /kg	dpm ^{26}Al /kg	Apparent exposure age (my)			dpm ^{53}Mn /kg Fe saturated
						^{3}He	^{21}Ne	^{38}Ar	
Yamato -7301 (j)	H4	0.65	101 ± 6^{d}	9 ± 1	30 ± 3	8.2^{a}	13^{a}	11^{a}	111
-7304 (m)	L5	0.50	412 ± 21^{d}	19 ± 2	61 ± 4	13^{a}	18^{a}	14^{a}	427
-7305 (k)	L5	0.90	352 ± 18^{d}	15 ± 5		17^{a}	22^{a}	23^{a}	358
-74013	Diogenite	2.06	401 ± 37			28^{b}	32^{b}	39^{b}	402
-74097	Diogenite	2.19	421 ± 38			29^{b}	35^{b}	35^{b}	422
-74155	H4	3.07	255 ± 15	9 ± 3					
-74191	L3	1.09	449 ± 23			4.3^{b}	6.4^{b}	5.3^{b}	643
-74647	H5	2.32	329 ± 17						
Mt. Baldr No. 1	H	4.11	394 ± 34			3.1^{b}	4.9^{b}, 5.8^{c}	4.1^{b}	623
No. 2	H	13.78	355 ± 26				4.8^{c}		600
Allan Hills No. 1	L	20.15	443 ± 33			23^{b}	28^{b}, 29^{c}	23^{b}	445
No. 3	L	10.50	431 ± 25				30^{c}		432
No. 5	Eucrite	1.43	390 ± 36		88.9 ± 1.5^{e}				
No. 6	H	1.14	453 ± 33		50.6 ± 0.8^{e}		16^{c}		477
No. 7	L	0.41	332 ± 26		45.2 ± 1.0^{e}		21^{c}		339
No. 8	H	1.15	22 ± 3		11.4 ± 0.4^{e}	0.98^{b}	1.4^{b}, 1.8^{c}	1.1^{b}	89
No. 9	L	407.04	477 ± 34				15^{c}		508

* After Ni and Mn correction.

a) TAKAOKA and NAGAO (1978), b) NAGAO and TAKAOKA (1979), c) SCHULTZ (1978), d) NISHIIZUMI *et al.* (1978), e) FRUCHTER and EVANS (1977).

the concentrations were in good agreement with those measured by atomic absorption spectroscopy. However, the NAA values had to be corrected for the relatively large neutron flux gradient observed inside the sample stack (~2 cm thick), which ranged from ~0% for Fe to ~15% for Ni, and therefore the data were not included in the table.

The final results for ^{53}Mn in 17 antarctic meteorites including 3 Yamato meteorites studied during the previous work (NISHIIZUMI *et al.*, 1978; NISHIIZUMI, 1978) are presented in Table 4. Data of duplicate analyses for 5 meteorites, which all showed excellent agreement as seen in the Table 1, were calculated as weighed averages and the values in the table include the corrections for the Ni and Mn contribution to the ^{53}Mn production. ^{53}Mn produced from Ni has been determined to be 1/3 of that from Fe (NISHIIZUMI, 1978) and that from Mn was estimated to be nearly equal to that from Fe. Data for ^{10}Be, which again showed excellent agreement in duplicate analyses, and for ^{26}Al are also presented in Table 4, together with other data for ^{26}Al determined by another group and the rare gas exposure ages given in literature. Errors attached to the data for ^{10}Be and ^{26}Al represent 1 σ counting statistics plus uncertainty in calibration of absolute counting efficiency. Errors for ^{53}Mn are those in the last column of the Table 1.

3. Discussion

In the last column of Table 4, the saturation specific activity of ^{53}Mn has been calculated from the respective exposure ages and the measured activities after Ni and Mn corrections. The ^{21}Ne exposure age was used in all calculations because the ^{3}He exposure age may represent an incorrect shorter age due to He loss. The saturation activity is equal to the production rate at equilibrium for a long cosmic rays irradiation, sufficiently longer than the mean life of ^{53}Mn, 5.3×10^{6}y. The variation in saturation activity may be due to the fact that the rare gas exposure ages do not necessarily reflect the actual exposure age. The production rates of cosmogenic nuclides vary with depth in a pre-atmospheric body but in the calculation of rare gas ages the same production rates are used for the same types of chondrites with no consideration for sample depth. The frequency distribution of saturated ^{53}Mn activity (calculated) in antarctic meteorites is shown in Fig. 1, together with those for other chondrites. In this figure meteorites that required a saturation correction greater than a factor of 2 (rare gas age $<3\times10^{6}$y) have not been included due to the uncertainty in the rare gas ages as mentioned above. In the present work it was found that the average specific saturation activity of ^{53}Mn in 12 antarctic meteorites, excluding the low activity of Yamato-7301 and Allan Hills No. 8, is 470±100 dpm/kg Fe. This value agrees fairly well with 450±90 dpm/kg Fe for 17 chondrites, reported by NISHIIZUMI (1978), and 520±80 dpm^{53}Mn/kg Fe by HEIMANN *et al.* (1974).

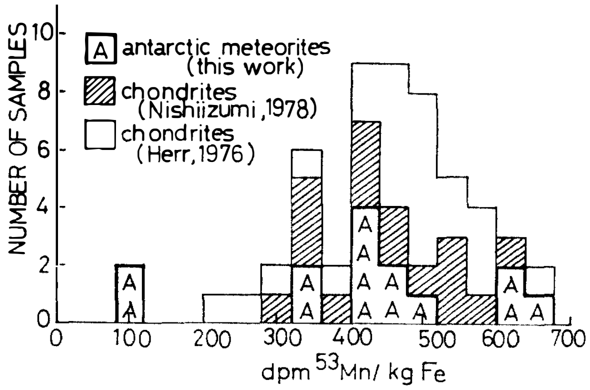

Fig. 1. Frequency distribution of saturation specific activity of ^{53}Mn in antarctic meteorites, compared with the distribution for other chondrites.

Three meteorites, Yamato-74191, Mt. Baldr No. 1 and No. 2, seem to have exceptionally high saturation activity. Although their values are within 2 standard deviations of the average specific saturation activity, the higher values are unlikely to be due to a chance event since all these meteorites have short exposure ages of 4–7 million years. This is more clearly shown by examining the average activity for nine antarctic meteorites with exposure ages of $>10^7$y, which is 423 ± 54 dpm/kg Fe, thus specific saturation activities of the above three meteorites are larger than the average for meteorites with exposure age of $>10^7$y by more than 3 standard deviations. The situation remains the same when the data of other six chondrites of long exposure age (NISHIIZUMI, 1978) are included. For all 15 stone meteorites of long exposure age, the average ^{53}Mn activity is 404 ± 56 dpm ^{53}Mn/kg Fe. Possible reasons for the high ^{53}Mn values in these meteorites are discussed later in this paper.

In contrast to the above three meteorites, two meteorites have extremely low values of ^{53}Mn, 101 ± 6 for Yamato-7301 and 22 ± 3 dpm/kg Fe for Allan Hills No. 8. It may be mentioned that Yamato-7301 and Allan Hills No. 8 have the lowest ^{53}Mn saturation activity contents reported so far among chondrites.

3.1. Terrestrial age

The low ^{53}Mn contents in Yamato-7301 and Allan Hills No. 8 cannot be explained by the long terrestrial ages of these objects because of the relatively high activities of ^{26}Al and ^{10}Be measured in them. The ratio of radioactivity content to the saturation activity should obey the function of $\exp(-\lambda\tau)$, where λ is the decay constant and τ the terrestrial age, so that the amount of decay should be larger for the nuclide of shorter half-life. ^{26}Al and ^{10}Be are not as low in these two meteorites as expected from the ^{53}Mn values. The average saturation specific activity of ^{26}Al is reported as 60.2 for H-group chondrites and 65.3 dpm/kg for the ones belonging to the L-group (FUSE and ANDERS, 1969). Saturation activity of

^{10}Be in ordinary chondrites is 20 dpm/kg (HONDA and ARNOLD, 1967).

The terrestrial ages of antarctic meteorites seem to be shorter than 10^6y because the average specific activity of ^{53}Mn for the meteorites with long exposure ages ($>10^7$y) is 423 ± 50 dpm/kg Fe while that for six chondrites, which were observed to fall, is 381 ± 50 dpm/kg (NISHIIZUMI, 1978).

A more stringent upper limit can be obtained from the ^{26}Al data. $[(^{26}\mathrm{Al})_{\mathrm{obs}}/(^{26}\mathrm{Al})_{\mathrm{cal}}]/[(^{53}\mathrm{Mn})_{\mathrm{obs}}/(^{53}\mathrm{Mn})_{\mathrm{cal}}]$ will give a value of $(^{26}\mathrm{Al})_{\mathrm{obs}}/(^{26}\mathrm{Al})_{\mathrm{cal}}$ corrected for shielding effects for meteorites with long exposure ages, excluding Yamato-7301, since ^{26}Al and ^{53}Mn are produced by cosmic rays in similar types of nuclear reactions. Taking $(^{53}\mathrm{Mn})_{\mathrm{cal}}$ as 423 dpm^{53}Mn/kg Fe and $(^{26}\mathrm{Al})_{\mathrm{cal}}$ as the values from FUSE and ANDERS (1969), we calculate 0.93, 0.75 and 0.86 for Yamato-7304 and Allan Hills Nos. 6 and 7, respectively. This suggests that the terrestrial ages of antarctic meteorites are less than $\sim 3\times10^5$y, although longer terrestrial ages cannot be excluded for unexamined meteorites. On the other hand, the absence of ^{14}C activity in three antarctic meteorites (FIREMAN, 1978) implies a terrestrial age $>2.5\times10^4$y for these objects. These values will provide boundary conditions on the mechanism of accumulation of antarctic meteorites proposed by NAGATA (1977).

3.2. *Multi-stage irradiation history*

In our previous papers (NISHIIZUMI *et al.*, 1978; NISHIIZUMI, 1978), we discussed the low ^{53}Mn content in Yamato-7301 with reference to its long exposure age and tried to explain the discrepancy by a long terrestrial age, heavy shielding in the pre-atmospheric meteorite body and multi-stage irradiation history with a long exposure preceding a recent short exposure. We tentatively decided the best explanation was a two-stage irradiation history (a simplified version of a multi-stage irradiation history) from the preliminary result of ^{10}Be. The low ^{53}Mn content in Allan Hills No. 8 seems to be due to a similar exposure. ^{53}Mn, ^{26}Al and rare gases cannot be explained consistently by either long terrestrial age or heavy shielding. The low content of ^{53}Mn in Allan Hills No. 8 as well as in Yamato-7301 can be explained by a model of a two-stage cosmic ray exposure history. The model can be visualized as follows: In the first stage, the meteorites were heavily shielded in a large parent body for a long time (T_1). Most of the cosmogenic stable nuclides might have been produced during this period. In the second stage, they were a part of small size secondary bodies ejected from the parent bodies possibly by a cratering event. These small bodies were irradiated with galactic cosmic rays for a short time (T_2) so that the radioactivity was less than the saturation value when the meteorites fell in Antarctica. The relationship between several parameters in a two-stage irradiation can be written as follows:

$$A=\bar{A}_0 f_s\,[1-\exp(-\lambda T_2)]\cdot\exp(-\lambda\tau)$$
$$+A_p\,[1-\exp(-\lambda T_1)]\cdot\exp[-\lambda(\tau+T_2)] \qquad (1)$$

where A is the measured radioactivity, $\bar{A}_0$ is the average saturated specific activity, f_s is the shielding factor, *i.e.*, the correction factor for shielding relative to a normal size meteorite, τ is the terrestrial age, A_p is the expected saturation activity at a particular depth and radius in a large parent body and λ is the decay constant. Assuming $T_1 \gg 1/\lambda$ (mean life of the radioactivity), eq. (1) can be rewritten as follows;

$$A=[\bar{A}_0 f_s-(\bar{A}_0 f_s-A_p)\exp(-\lambda T_2)]\exp(-\lambda\tau) \tag{2}$$

$$A_p/\bar{A}_0 f_s=1-\exp(\lambda T_2)[1-(A/\bar{A}_0 f_s)\exp(\lambda\tau)]. \tag{3}$$

We can use eq. (3) for as many radionuclides as have been measured. Assuming various values of $\bar{A}_0$, τ and f_s, we can determine the constraints on $A_p/\bar{A}_0$ and T_2. T_1 is calculated from the relationship; $T_1=(T_s-T_2)/(A_p/\bar{A}_0)$, where T_s is the apparent exposure age from rare gases.

In Figs. 2a and 2b, the constraints on $A_p/\bar{A}_0$ and T_2 are graphically illustrated for Yamato-7301. Instead of the average saturation specific activity, the saturated specific activity of Yamato-7304 was substituted for $\bar{A}_0$ ($\bar{A}_0$ for ^{26}Al was corrected for the difference of meteorite type). The same value of f_s was assumed for every nuclide although this is not strictly valid. Fig. 2a is the case with $\Delta\tau=0.2\times10^6$y and $f_s=1$ (usual size), and Fig. 2b the case with $\Delta\tau=0$ and $f_s=0.6$ (large size), where $\Delta\tau$ represents the difference of terrestrial age between Yamato-7301 and Yamato-7304. When $\Delta\tau=0$ is assumed, the best fit is obtained for the f_s values, 0.73, 1 and 0.73 for ^{26}Al, ^{10}Be and ^{53}Mn, respectively. This case gives T_2 of 1.4×10^6y and T_1 of 2×10^8y. The above f_s values, which are for a very small pre-atmospheric size body, seem to be reasonable since the ^{22}Ne/^{21}Ne ratio has been reported as 1.12 (TAKAOKA and NAGAO, 1978a), larger than 1.08, the value expected from heavy shielding in the first stage. This may suggest irradiation in a very small body during the second stage. On the other hand, a positive $\Delta\tau$ value is indicated by the observation of weathering and oxidation in Yamato-7301 (YAGI *et al.*, 1978). If terrestrial age for Yamato-7304 is assumed to be 10^5y, an age of $3\text{–}4\times10^5$y for Yamato-7301 gives the realistic values of T_2 of $1.1\text{–}1.6\times10^6$y and T_1 of $>10^8$y for this meteorite. To determine T_2 and T_1 more definitely, it is important to measure shorter-lived radionuclides such as ^{81}Kr and ^{36}Cl.

The case for Allan Hills No. 8 is less certain, because data for only two nuclides, ^{53}Mn and ^{26}Al, are available. The ^{22}Ne/^{21}Ne ratio was found to be 1.08 (NAGAO and TAKAOKA, 1979), which is consistent with exposure under considerable shielding. From the recovered mass of 1.15 kg, an assumption of normal pre-atmospheric size may be valid so that $f_s=1$. Assuming a terrestrial age for Allan Hills No. 8 comparable to that for Yamato-7304, we can estimate T_2 from the following equation;

$$T_2=[A(^{26}\mathrm{Al})/\bar{A}_0(^{26}\mathrm{Al})-A(^{53}\mathrm{Mn})/\bar{A}_0(^{53}\mathrm{Mn})]/[\lambda(^{26}\mathrm{Al})-\lambda(^{53}\mathrm{Mn})]. \tag{4}$$

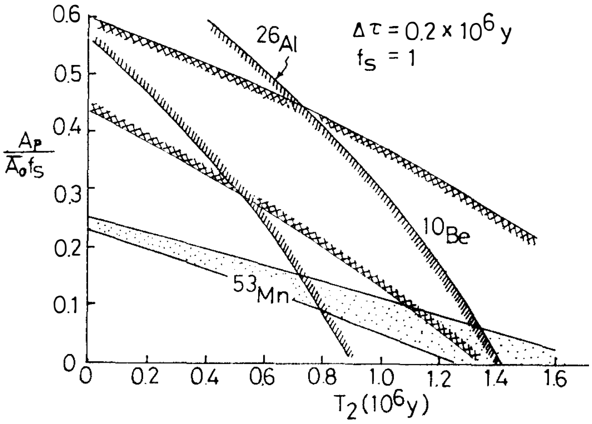

Fig. 2a. Constraints on T_2 and $A_p/\bar{A}_0$ for Yamato-7301 assuming $\Delta\tau=0.2$ million years and $f_s=1$ for a two-stage irradiation model. T_2 is the duration of the 2nd stage, A_p is the saturation activity in the first stage and $\bar{A}_0$ is the average saturation activity in usual size meteorites. T_1, the time span of pre-irradiation in the first stage, is calculated from the relation: $T_1=(T_s-T_2)/(A_p/\bar{A}_0)$, where T_s is the apparent exposure age from rare gases. The ranges given for the three radionuclides are due to the uncertainties in the measured data. The uncertainty ($\pm 10\%$) of ^{53}Mn half-life is also included.

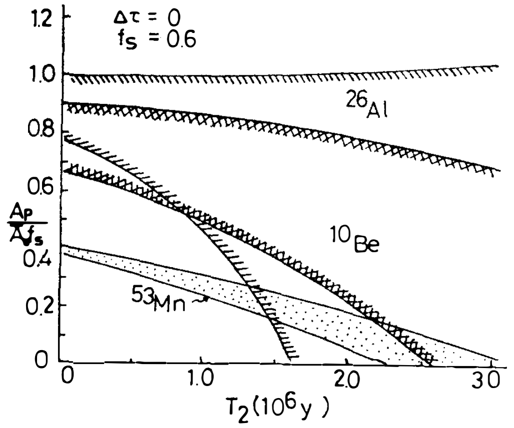

Fig. 2b. Constraints on T_2 and $A_p/\bar{A}_0$ for Yamato-7301 assuming a negligible terrestrial age ($\Delta\tau=0$) and a large pre-atmospheric size ($f_s=0.6$).

Eq. (4) is valid when $T_2 \ll t_{1/2}(^{26}\text{Al})$. Because of the very low activities measured for ^{53}Mn and ^{26}Al use of this equation seems reasonable. Substituting 61±4 dpm/kg and 427±11 dpm/kg Fe, the values of Yamato-7304, for $\bar{A}_0(^{26}\text{Al})$ and $\bar{A}_0(^{53}\text{Mn})$, respectively, we get 0.17±0.02 million years for T_2, which is smaller by one order of magnitude than the ^{21}Ne age, 1.6±0.2 million years. A_p is calculated as 7±4 dpm/kg Fe for ^{53}Mn, suggesting the pre-irradiation occurred at a depth of

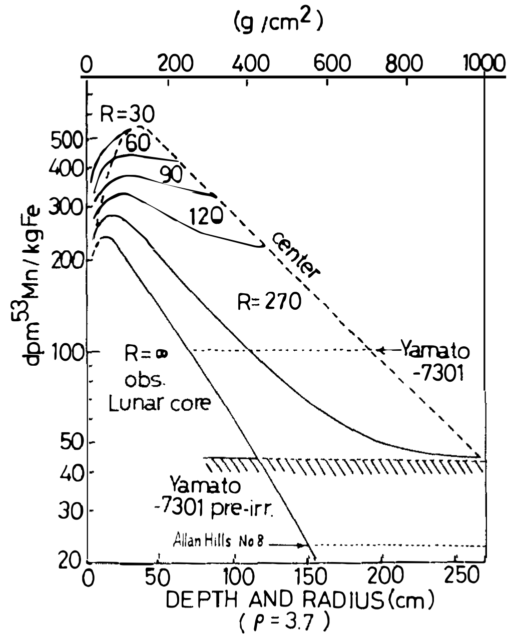

Fig. 3. Estimated ^{53}Mn contents in spherical pre-atmospheric meteoroids of various radii (R) and observed ^{53}Mn depth profiles in Apollo 15 and 16 deep drill cores (Imamura et al., 1973, 1974). Observed activity levels in Yamato-7301 and Allan Hills No. 8 is shown. The estimated upper limit of the activity level in the first stage of irradiation in Yamato-7301 is also shown.

2–4 m in a large parent body with a radius larger than 4 m (Fig. 3). T_1 is then estimated to be 90±50 million years. This might, however, give only the lower limit of T_1 because the assumption of a terrestrial age long compared to Yamato-7304 leads to a higher value of T_2, therefore lower value of A_p and higher T_1. Determination of ^{36}Cl or ^{81}Kr will help to obtain an upper limit of T_2.

In both cases of Yamato-7301 and Allan Hills No. 8, we have had evidence for a two-stage irradiation history for the meteorites. The duration of pre-irradiation was found to be of the order of 10^8y or longer compared to the recent exposure of $\sim 10^6$y. This seems to be quite consistent with the long argued scenario for the formation of meteorites from asteroid families with earth-crossing orbits (Arnold, 1965b; Heymann and Anders, 1967). Since no chondrites with an exposure age of $>10^8$y are known, the parent bodies of Allan Hills No. 8 and Yamato-7301 are, probably, not simply large meteorites but bodies like asteroids.

It should be noted that T_1 might be influenced by the removal rate of material from the surface of the parent body, as was once considered as an explanation for the exposure ages of meteorites (Whipple and Fireman, 1959). The upper limit of removal rates is easily obtained because of the simple relationship between the production rate of a nuclide and the depth in very deep locations, that is, an exponential decrease with increasing depth. In the case of ^{53}Mn production from

Fe, a half attenuation length of about 150 g/cm^2 has been found (IMAMURA *et al.*, 1973, 1974). If the same value is adopted for ^{21}Ne production, a removal rate is calculated as ≤ 6 mm/10^6y. This is quite similar to the erosion rates observed in the rocks on the lunar surface (FINKEL *et al.*, 1972; WAHLEN *et al.*, 1973). Because too small a parent body will lose all the materials when excavated, a low rate of material removal might suggest the existence of enough gravity to keep materials of low velocity from leaving the surface of the parent body.

The possibility of a two-stage irradiation history is not new to meteorites. A few meteorites are thought to have had complex irradiation records. The Sikhote-Alin (VILCSEK and WÄNKE, 1961), the Serra de Mage 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 cases. Probably many more meteorites have experienced a multi-irradiation history since it is usually very difficult to detect the evidence when the exposure age in the second, most recent, stage exceeds 3 million years. Accumulation of ^{53}Mn data together with those of rare gases is necessary to obtain statistically meaningful data on the general patterns of meteorite exposure histories.

3.3. *High ^{53}Mn content in meteorites of young exposure ages*

It has been pointed out above that three meteorites, Yamato-74191, Mt. Baldr No. 1 and No. 2, seem to have high saturation activities of ^{53}Mn and that these are all young in exposure age, in the range of 4–7 million years. This suggest possible variation in the production rate for the ratio $^{53}Mn/^{21}Ne$, if we exclude the possibility that cosmic rays were enhanced in the last 4–5 million years, but had been reduced by a factor in the preceding millions of years. Gas loss may be neglected because K-Ar ages (NAGAO and TAKAOKA, 1979) for these three meteorites seem to be old enough to support the absence of any recent violent event to cause degassing of cosmic ray produced rare gases. ^{53}Mn is produced mainly from ^{56}Fe through the $(n, p3n)$ and $(n, 4n)$ reactions. Whereas ^{21}Ne is produced by somewhat different types of reactions, mainly by ^{24}Mg (n, α), which is caused by low energy secondary neutrons, and ^{24}Mg $(n, 2p2n)$, ^{24}Mg $(n, p3n)$ and other reactions. The latter reactions resemble those for ^{53}Mn production. Furthermore, a considerable part of ^{21}Ne (*ca.* 30%) comes from Si through high energy spallation reaction. Therefore, the production rate of ^{21}Ne is theoretically lower than ^{53}Mn in both the surface and the deep places of a meteorite body in space. For a surface sample, the use of a conventional production rate for ^{21}Ne could cause an underestimation of the exposure age by ~30% (HERZOG and CRESSY, 1974). Actually Yamato-74191 does seem to be a small meteorite. It's $^{22}Ne/^{21}Ne$ ratio is 1.19 (NAGAO and TAKAOKA, 1979), suggesting irradiation at shallow depths (WRIGHT *et al.*, 1973). Recently FINKEL *et al.* (1978) measured rare gas exposure ages in the small San Juan Capistrano meteorite which has the ratio $^{22}Ne/^{21}Ne$ of

1.19. The ^{81}Kr-Kr age of the meteorite is longer than the ^{3}He and ^{21}Ne ages by 30–50%. The saturation activity calculated from the increased exposure age is 550 dpm^{53}Mn/kg Fe. This seems still too high a value because ^{53}Mn production is thought to be lower in the surface than inside a meteorite body of usual size.

On the other hand, ^{22}Ne/^{21}Ne in Mt. Baldr No. 1 is reported to be 1.05 (NAGAO and TAKAOKA, 1979), which seems to indicate a large pre-atmospheric size. This effect may increase the exposure age of Mt. Baldr No. 1 by some tens of percent, but not enough to decrease the saturation activity of ^{53}Mn to the normal range. In addition to the difficulties with the ^{21}Ne production rates discussed above, the high saturation specific activities might be due to the overestimation of rare gas production because of a multi-stage irradiation history. As discussed in the preceding section, such complex irradiations might be very common among meteorites. Excess rare gases from a pre-irradiation could bias the estimation of the production rate of rare gas isotopes from direct comparison between ^{26}Al and rare gas contents. If the parent body has been irradiated for a period of more than 3×10^{9}y, an excess of ^{21}Ne would be significant for the meteorite derived from a depth of <5 m. Thus the production rates given by HERZOG and ANDERS (1972) might be too high. The use of the previous values for rare gas production rates would decrease the saturation value of ^{53}Mn for the 3 meteorites to 530–580 dpm/kg Fe, which still seems high. Some combination of the two effects mentioned above might best explain the high saturation activity of ^{53}Mn in these objects.

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