COSMOGENIC ^{53}Mn IN ANTARCTIC METEORITES AND THEIR EXPOSURE HISTORY

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Abstract: Our previous studies on cosmic ray produced radio-nuclides in antarctic meteorites were extended to 13 additional antarctic meteorites in order to obtain information on the cosmic ray exposure history of these meteorites. The ^{53}Mn activities in these meteorites are all close to the saturation value which indicates exposure ages longer than 4 million years. The effect of weathering on the ^{53}Mn content was investigated in two oxidized meteorites, Yamato-7301 and Allan Hills-76008. These meteorites have been shown by our previous works to have experienced two-stage irradiations in space. The ^{53}Mn contents in the metal phases agreed within the error with those in the non-magnetic phases which contained high concentrations of oxidized Fe of metallic origin, and also with the values for bulk samples previously reported. The effect is clearly shown of minor significance. The data on spallogenic nuclides so far obtained for these two meteorites are summarized and discussed in light of a two-stage irradiation history.

1. Introduction

In our previous papers (NISHIIZUMI *et al.*, 1978, 1979a) we concluded, from the analyses of ^{53}Mn($t_{1/2}$=3.7×10^{6}y), ^{10}Be($t_{1/2}$=1.6×10^{6}y) and ^{26}Al($t_{1/2}$=7.2×10^{5}y) together with the reported data of rare gases, that at least two meteorites, out of 17 antarctic meteorites investigated, have unique cosmic ray exposure histories. These two meteorites, Yamato-7301 and Allan Hills-76008, have an order of magnitude younger exposure ages based on the radioactivity contents than those based on the spallogenic rare gas contents. This discrepancy has been explained by a two-stage irradiation model in which the meteorites were pre-irradiated under heavy shielding (within a few meters from the surface of the meteorite parent body) for a long time ($\sim 10^8$ years or longer) as a part of a large parent body before the recent brief exposure (~ 1.3 and $\sim 0.24 \times 10^6$ years for Yamato-7301 and Allan Hills-76008, respectively) as a much smaller body. This model is supported by the recent determination of ^{36}Cl with a Van de Graaff accelerator which provided information on the terrestrial ages of these antarctic meteorites (NISHIIZUMI *et al.*, 1979b).

A two-stage irradiation is closely related to the formation mechanism of the meteorites. Meteorites are supposed to be derived as either a part of catastrophic products or a part of cratoring ejecta from a meteorite parent body. According to MCDONNEL and ASHWORTH (1972), a body of >1 m size has a long lifetime ($\gtrsim 10^9$ y at 1 A.U.) against the impact destruction and a mean life against the impact abrasion is much shorter. Therefore meteorites are more likely to be a part of cratoring ejecta from the parent body than to be a part of catastrophic products from a larger meteorite. These parent bodies may be near earth asteroids like Apollo-Amor objects. Therefore, the pre-irradiation observed in Yamato-7301 and Allan Hills-76008 had possibly occurred within a few meters from the surface of the asteroid. Our model may also be discussed in relation to the evidence of pre-irradiation found in some brecciated ordinary chondrites, which was first reported by PELLAS *et al.* (1969) and LAL and RAJAN (1969). PELLAS (1973) has extensively studied both clast and matrix using track and rare gas methods. He concluded that pre-irradiation must have occurred in the accretional stage of the meteorite parent body. Recently SCHULTZ and SIGNER (1977) found evidence that the St. Mesmin chondrite, which is one of the chondrites studied by Pellas and his co-workers, was compacted relatively late in the history of the meteorite, not more than 1.3 b.y. ago. Our observations as well as those in the brecciated chondrites are related to the surface processes which occurred in asteroids during their evolutional history.

It is, therefore, quite interesting to know how frequently the pre-irradiation is observed among the meteorites of young exposure ages, and to determine the time scale and the condition in which the pre-irradiation occurred. Such information may offer a firm evidence of direct delivery of meteorites from asteroidal surface.

In this report, we describe the measurements of ^{53}Mn in 13 other antarctic meteorites and the results of weathering effect on the ^{53}Mn content in Yamato-7301 and Allan Hills-76008. Weathering effects were studied by analyzing the ^{53}Mn in the metallic phase *v.s.* the non-magnetic fraction enriched in oxidized iron. The radionuclide data and those of rare gases so far obtained are summarized and discussed in light of a two-stage irradiation in space.

2. Experimental and Results

Duplicate samples (0.5–0.9 g) of meteorites for ^{53}Mn determination were prepared from the crushed stone. For the iron meteorite, Allan Hills-76002, a small piece (0.1 g) was cut from the block and the surface was cleaned with dilute HCl. The metallic fractions of Yamato-7301 and Allan Hills-76008, which had each been separated from about 10 g crushed meteorite using a hand magnet, were refined by repeated grinding in an agate mortar. Meanwhile the non-magnetic fraction enriched in oxidized iron was recovered as the residue in the magnetic separation. The metallic fraction was further purified by repeated treatments with 0.2 N HCl in an ultrasonic bath followed by additional grinding and magnetic separation. The sieved fraction of >300 μm was used for the determination of ^{53}Mn. The recoil of spallation products is estimated to be of no significance for this size range (NYQUIST *et al.*, 1973). 150–300 μm fraction was used for ^{36}Cl (NISHIIZUMI *et al.*, 1979b) and rare gas analyses (TAKAOKA and NAGAO, 1979). For the mass spectroscopic analysis of cosmogenic ^{40}K($t_{1/2}=1.3\times10^9$y), this fraction was further treated with HF in an ultrasonic bath.

^{53}Mn was determined by the neutron activation method (MILLARD, 1965). The chemical procedures were essentially the same as in our previous works (*e.g.* IMAMURA *et al.*, 1973). After the dissolution of the sample, an aliquot was taken for the chemical analysis. For the metallic samples, 212 μg Mn was added as carrier.

The neutron irradiations were performed in the VG-7–6 hole of the JRR-3 reactor of the Japan Atomic Energy Institute, Tokai, Ibaraki, for 266 hours (Irradiation A) and for 267.5 hours (Irradiation B). The total thermal neutron fluences were 2.3×10^{18} n/cm^2 for Irradiation A and 2.5×10^{18} n/cm^2 for Irradiation B, based on the Co monitor.

^{54}Mn was counted using three Ge(Li) detectors. A well-type (5 mmϕ) Ge(Li) detector from Princeton Gamma-Tech (st UCSD) was used for the samples of "Irradiation A". The counting efficiency (ε) was 4.6% for the ^{54}Mn photopeak and the B.G. was 0.014 cpm (834.8±3.5 keV). For the other samples, the counting was carried out by two Ge(Li) detectors, one (at ISSP) from Princeton Gamma-Tech (ε=2.3%; B.G.=0.04 cpm for 834.8±2.0 keV) and the other from Horiba (ε=1.9%; B.G.=0.07 cpm for 834.8±2.5 keV). The detailed data for the neutron activation of ^{53}Mn are presented in Table 1.

The concentrations of Al, Mn, Fe, Co and Ni in the meteorites were determined by atomic absorption spectroscopy and are given in Table 2. The errors for the determination are estimated to be 2% for Mn and 3% for Al, Fe, Co and Ni. The errors for Co and Ni in diogenites may be much larger. The weighted average for the duplicate ^{53}Mn analyses are shown in the last two columns of Table 2. In the

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

Sample		Sample description				Irradiation[1]	Sample counted after irradiation				After (*n*, 2*n*) correction	
		wt. (g)	Fe (%)	Mn (ppm)	Ni (%)		Fe (μg)	Mn (μg)	^{54}Mn[2] (cpm)	^{54}Mn[3] (cpm/mg Mn)	^{54}Mn[4] (cpm/mg Mn)	^{53}Mn[5] (dpm/kg Fe)
Yamato												
-692	(1)	0.496	13.5	3910	0.006	A	0.81	1637	25.98±0.65	15.83±0.50	12.78±0.51	461±26
	(2)	0.473	14.7	4300	0.006	A	0.34	1660	24.65±0.52	14.83±0.39	11.78±0.40	429±22
-74014	(1)	0.615	24.3	2630	1.49	A	0.42	1354	44.57±1.00	32.89±0.98	29.84±0.98	402±21
	(2)	0.629	26.6	2540	1.70	A	0.34	1239	46.17±0.98	37.24±1.09	34.19±1.09	407±21
-74037	(1)	0.467	13.3	4140	0.003	A	0.30	1676	24.96±0.43	14.87±0.39	11.82±0.40	458±24
	(2)	0.516	13.2	4060	0.006	A	0.34	1774	25.54±0.43	14.39±0.37	11.34±0.38	434±23
-74118	(1)	0.542	23.8	2660	1.44	A	0.49	1309	37.89±0.78	28.91±0.83	25.86±0.83	360±19
	(2)	0.488	21.3	2790	1.36	A	0.40	1241	31.76±0.76	25.57±0.80	22.52±0.80	367±20
-74136	(1)	0.532	12.6	4180	0.002	A	0.32	1982	28.87±0.72	14.57±0.46	11.52±0.47	476±27
	(2)	0.543	12.6	4180	0.002	A	0.56	1985	27.93±0.72	14.04±0.46	10.99±0.47	454±26
-74190	(1)	0.839	22.9	2740	1.24	A	0.38	2070	68.54±0.96	33.11±0.80	30.06±0.80	448±22
	(2)	0.843	22.5	2760	1.25	A	0.32	2105	68.41±1.48	32.50±0.96	29.45±0.96	450±23
-74354	(1)	0.910	22.1	2750	1.19	A	0.36	2121	75.52±1.02	35.59±0.87	32.54±0.87	504±24
	(2)	0.625	21.5	2780	1.11	A	0.37	1581	53.50±0.89	33.83±0.87	30.78±0.87	496±24
-74362	(1)	0.893	25.3	2660	1.45	A	0.47	2126	60.43±1.00	28.41±0.74	25.36±0.75	332±16
	(2)	0.822	23.9	2680	1.35	A	0.42	2013	55.85±1.26	27.74±0.83	24.69±0.83	345±18
-74371	(1)	0.737	28.3	2480	1.75	A	0.38	1655	51.85±1.00	31.30±0.87	28.25±0.87	308±16
	(2)	0.867	29.4	2480	1.78	A	0.48	1964	66.26±1.50	33.72±1.02	30.67±1.02	322±17
-74445	(1)	0.756	22.2	2710	1.15	A	0.39	1798	45.80±0.96	25.46±0.74	22.41±0.75	341±18
	(2)	0.758	22.1	2680	1.11	A	0.48	1569	38.63±0.76	24.61±0.70	21.56±0.71	326±17

Table 1 (continued-2).

Sample	Sample description wt. (g)	Fe (%)	Mn (ppm)	Ni (%)	Irrad-iation[1]	Sample counted after irradiation Fe (μg)	Mn (μg)	^{54}Mn[2] (cpm)	^{54}Mn[3] (cpm/mg Mn)	After (*n*, 2*n*) correction ^{54}Mn[4] (cpm/mg Mn)	^{53}Mn[5] (dpm/kg Fe)
Yamato											
-74640 (1)	0.695	27.3	2410	1.66	A	0.27	1485	71.83±1.39	48.35±1.35	45.30±1.35	498±25
(2)	0.594	26.4	2400	1.70	A	0.10	1294	63.48±1.28	49.04±1.39	45.99±1.39	521±26
-74646 (1)	0.552	18.9	2830	0.84	A	0.49	1413	27.07±0.70	19.13±0.63	16.08±0.64	300±17
(2)	0.583	19.0	2830	0.88	A	1.03	1521	29.24±0.63	19.17±0.57	16.12±0.58	299±16
-7301 metal[6]	0.0735	87.9	46	6.49	B	0.2	183[8]	5.93±0.25	32.32±1.50	29.14±1.50	112± 7
n.m.[7]	0.0996	21.5	2820	1.63	B	0.1	212	2.07±0.18	9.72±0.87	6.54±0.87	99±13
Allan Hills											
-76002	0.0754	90.1	—	7.23	B	0.2	180[8]	29.18±0.61	162.0±4.7	158.8±4.7	572±22
-76008 metal[6]	0.1007	90.7	30	6.50	B	0.1	184[8]	2.16±0.17	11.70±0.95	8.52±0.95	23± 3
n.m.-1[7]	0.246	23.0	2250	1.40	B	0.2	402	2.22±0.19	5.48±0.49	2.30±0.50	26± 6
n.m.-2[7]	0.415	18.4	2685	0.77	B	0.1	768	3.69±0.18	4.80±0.25	1.62±0.27	27± 5
^{53}Mn standard	Solution (g)								dpm^{54}Mn/dpm^{53}Mn		
^{53}Mn-A (1)	0.239	0.781 dpm^{53}Mn /mg Mn			A	0.13	283	175.9±3.0	621±17	618±17	803±15
^{53}Mn-A (2)	0.327				A	0.25	380	242.4±3.5	638±16	635±16	
^{53}Mn-B (1)	0.362	0.445 dpm^{53}Mn /mg Mn			B	0.1	124	49.4±0.7	398±10	395±10	866±22
^{53}Mn-B (2)	0.425				B	0.1	144	54.5±1.0	379±10	376±10	
^{55}Mn standard	Mn metal (mg)								dpm^{54}Mn/mg Mn		
^{55}Mn-A (1)	6.9				A	0.03	6710	20.04±0.83	2.99±0.14	3.05±0.09	
^{55}Mn-A (2)	10.2				A	0.09	9450	29.33±0.85	3.10±0.11		
^{55}Mn-B (1)	2.98				B	0.1	2907	9.12±0.30	3.14±0.12	3.18±0.09	
^{55}Mn-B (2)	4.98				B	0.1	4911	15.90±0.67	3.24±0.15		

Table 1 (continued-3).

Sample	Sample description				Irrad-iation[1)]	Sample counted after irradiation				After $(n, 2n)$ correction	
	wt. (g)	Fe (%)	Mn (ppm)	Ni (%)		Fe (μg)	Mn (μg)	^{54}Mn[2)] (cpm)	^{54}Mn[3)] (cpm/mg Mn)	^{54}Mn[4)] (cpm/mg Mn)	^{53}Mn[5)] (dpm/kg Fe)
Fe standard	Fe metal (mg)					Yield (%)				dpm^{54}Mn/μg Fe	
Fe-A (1)	0.86				A	95.1		56.9±1.8	0.069±0.002	0.069±0.002	
Fe-A (2)	0.62				A	96.3		40.8±1.5	0.069±0.002		
Fe-B (1)	0.96				B	98.3		73.4±1.9	0.076±0.003	0.077±0.002	
Fe-B (2)	0.72				B	98.6		53.8±0.7	0.076±0.002		

1) Samples were irradiated in VG-7-6 of the JRR-3 reactor for the periods of May 15–26, 1978 (266 hours; Irradiation A) and December 4–15, 1978 (267.5 hours; Irradiation B). The total neutron fluence was 2.3×10^{18} n/cm^2 and 2.5×10^{18} n/cm^2, respectively, as estimated from the reaction ^{59}Co (n, γ) ^{60}Co, $\sigma=37b$.

2) Weighted averages of at least two counting periods. All activities have been corrected to the end of irradiation. The uncertainties are 1σ counting statistics.

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

4) Uncertainty calculated by quadratically adding the uncertainty in the ^{55}Mn $(n, 2n)$ ^{54}Mn correction to that for the ^{54}Mn (dpm/mg Mn).

5) The following uncertainties have been added quadratically: 2% for Mn concentration; 3% for Fe concentration and the percentage uncertainty from the previous column. The 5% uncertainty in the original standardization of the ^{53}Mn standard has not been included.

6) Metallic fraction (>300 μm).

7) Non-magnetic fraction enriched in oxidized Fe of metallic origin.

8) 212 μg of Mn carrier was added in the pre-irradiation chemistry.

Table 2. Chemical composition and ^{53}Mn content of antarctic meteorites.

Meteorite	Class	Al (%)	Mn (ppm)	Fe (%)	Co (ppm)	Ni (%)	Fe/Mn	dpm^{53}Mn/kg Fe	dpm^{53}Mn/kg (Mn+Fe+1/3 Ni)
Yamato-692	Diogenite	0.39	4100	14.1	35	0.006	34	442±17	429±17
-74014	H6-5	1.15	2590	25.5	770	1.60	98	405±15	393±15
-74037	Diogenite	0.50	4100	13.2	36	0.005	32	445±17	432±16
-74118	L5-6	1.19	2720	22.6	800	1.40	83	363±14	352±14
-74136	Diogenite	0.40	4180	12.6	32	0.002	30	464±19	449±18
-74190	L5-6	1.28	2750	22.7	600	1.25	83	449±17	436±16
-74354	L6-5	1.23	2760	21.9	540	1.16	79	500±17	485±17
-74362	L6	1.22	2670	24.6	780	1.40	92	338±12	328±12
-74371	H5-6	1.17	2480	28.9	890	1.77	116	315±12	306±12
-74445	L4-5	1.25	2700	22.2	620	1.13	82	333±12	324±12
-74640	H6-5	1.12	2400	26.9	850	1.68	112	509±18	494±17
-74646	LL5-6	1.29	2830	19.0	440	0.86	67	299±12	290±12
Allan Hills -76002	Octahedrite	—	—	90.1	4730	7.23	—	572±22	556±21

last column, the specific activities of ^{53}Mn in Fe have been corrected for a small contribution from Mn and Ni. The production rates of ^{53}Mn in Mn and Ni relative to that in Fe are estimated to be ~1 and one third (NISHIIZUMI, 1978), respectively.

3. Discussion

3.1. ^{53}Mn in 13 antarctic meteorites and their exposure history

Among the 13 meteorites studied in this work, only 3 meteorites, Yamato-692 (SHIMA *et al.*, 1973), Yamato-74190 and Yamato-74640 (KAMAGUCHI and OKANO, 1979) have been investigated for rare gases. The ^{21}Ne exposure ages for these three meteorites are reported as 31, 25 and 11 million years. The production rates of ^{53}Mn are calculated to be 432, 440 and 556 atoms/min/kg Fe, respectively. Except for the high value for Yamato-74640, the ^{53}Mn production rate in the other two meteorites are very close to the average saturation activity of 423±54 dpm/kg Fe found for 9 antarctic meteorites with exposure ages of $>10^7$y (NISHIIZUMI *et al.*, 1979a), and to the value 450±63 dpm/kg Fe compiled by ENGLERT and HERR (1978) for 16 meteorites with exposure ages of $>1.4\times10^7$y. These values can be explained by normal exposure histories. The ^{53}Mn values for the other stone meteorites measured in this work range from 290 to 500 dpm/kg Fe, which suggest exposure ages older than 4 million years.

So far we have analyzed 5 diogenites for ^{53}Mn: Yamato-692, -74013, -74037, -74097 and -74136. The ^{53}Mn activities in these meteorites are surprisingly clustered in the range of 425±25 dpm/kg Fe. According to YANAI (1978), Yamato-74013, -74097 and -74136 were originally one achondrite body, although they do not make a complete specimen. As far as the ^{53}Mn content and the major elemental abundance are concerned, these five meteorites are likely to be samples from a single meteorite.

3.2. *Weathering effect on cosmic ray produced ^{53}Mn in antarctic meteorites*

Many antarctic meteorites are observed to have suffered from severe oxidation. The extent of oxidation from sample to sample is quite variable and depends on the meteorite type and the existence of fractures as well as the terrestrial age of the meteorite. In some meteorites, significant oxidation is observable only along the fracture lines. In meteorites such as Yamato-7301 and Allan Hills-76008, homogeneous oxidation is seen to extend over most of the meteorite. Yet these meteorites are quite solid. It is of interest to know the relation of such extensive oxidation with the terrestrial age of the meteorite, the hydrostatic pressure experienced by the meteorite while buried deep in the ice sheet, and the length of time the meteorite was exposed on the surface of the ice flow. Actually an interesting case has been found. Allan Hills-77002, L5 chondrite, contains only 1% metal (NISHIIZUMI *et al.*, 1979b). This meteorite was found to have exceptionally old terrestrial age estimated to be 0.7–0.8 million years from ^{26}Al (EVANS and RANCITELLI, 1979) and ^{36}Cl (NISHIIZUMI *et al.*, 1979b) measurements. Oxidation seems to be correlated with long terrestrial age.

The effect of weathering on cosmic ray produced radionuclides was raised by the discussion of FIREMAN *et al.* (1979). They prefer the model of a long terrestrial age to explain the data of ^{14}C ($t_{1/2}=5.7\times10^3$y; <1.7 dpm/kg meteorite) and ^{26}Al in Allan Hills-76008 as opposed to a two-stage irradiation history. They have tried to explain the low content of ^{53}Mn, which was inconsistently low compared to ^{26}Al in their model, by a leaching mechanism, that is, the loss of cosmic ray produced ^{53}Mn from the oxidized phase of metal in antarctic meteorites. Their model, however, inconsistent with the ^{36}Cl result for the metallic phase recently obtained using an accelerator (NISHIIZUMI *et al.*, 1979b).

It is reasonable to consider the transport of the soluble elements by the action of interstitial water, however, Mn does not seem to be soluble as it is oxidized to Mn^{3+} or Mn^{4+} in the open air. The corrosion of iron meteorites have been investigated by HEIMANN (1974) for the chemical composition of major and minor elements. Mn was found to be enriched in the oxidized phase of the Ider iron meteorite, consistent with the expected behavior of Mn (of terrestrial origin) as Mn^{3+}/Mn^{4+} scavenged by hydrated ferric iron oxides. Therefore ^{53}Mn is estimated to remain in the iron oxide phase unless the meteorites are exposed to water of high Mn concentration, where ^{53}Mn may be lost due to the isotopic exchange between

Table 3. ^{53}*Mn in metal and oxidized phases of Yamato-7301 and Allan Hills-76008.*

Sample	Fe (%)	Co (%)	Ni (%)	$Fe^0/\Sigma Fe$[c)]	$Fe(0)/\Sigma Fe$[d)]	Fe^{II} (silicate +troilite)/Σ Fe	dpm^{53}Mn/kg (Mn+Fe+1/3 Ni)
Yamato-7301							
metal[a)]	89.7	0.47	9.3	1.0	0	0	109±7
bulk	25.4	0.070	1.56	0.30	0.21	0.49	101±6[e)]
n.m.[b)]	21.5	0.067	1.63	0	0.58	0.42	95±12
Allan Hills -76008							
metal[a)]	89.8	0.48	9.0	1.0	0	0	23±3
bulk	25.5	0.074	1.68	0.29	0.24	0.47	22±3[f)]
n.m. (1)[b)]	23.0	0.072	1.40	0	0.57	0.43	25±6
n.m. (2)[b)]	18.4	0.039	0.77	0	0.38	0.62	26±5

a) Magnetic fraction of >300 μm. The chemical compositions are given for the bulk metallic fraction after the correction for the silicate contribution.
b) Non-magnetic fraction, enriched in oxidized iron, Fe(0), of metallic origin.
c) The fraction of metallic iron to total Fe.
d) The fraction of oxidized iron, Fe(0), of metallic origin to total Fe, estimated from Co content. Co in the silicate phase was assumed to be 20 ppm. The Co in the troilite phase was neglected in the estimation.
e) NISHIIZUMI *et al.*, 1978.
f) NISHIIZUMI *et al.*, 1979a.

water and the oxide phases. Conditions in Antarctica do not favor the loss of ^{53}Mn.

To confirm the above considerations, we have performed an experiment to measure the ^{53}Mn content in the metal and oxide phases of Yamato-7301 and Allan Hills-76008. Because contact of the oxidized phase with solvents such as water or alcohol might cause the re-distribution of some elements, we did not try to extract pure oxidized phase by (heavy) liquid separation. Since the oxidized phases adhere closely to the metal grains, the non-magnetic fraction recovered as the residue during the magnetic separation becomes increasingly enriched in the oxidized phase of iron as the purification proceeds. Thus we could prepare the samples enriched in oxidized iron of metallic origin by repeated grinding. Table 3 shows the results of the experiment.

The content of oxidized iron relative to total iron was estimated from the Co content assuming the oxidation proceeded equally on α- and γ-Fe-Ni. The Co in the silicate phase was assumed to be 20 ppm (IMAMURA and HONDA, 1976) and the contribution of troilite was neglected. Ni also can be an indicator, but it seems to be contained in significant amount in the troilite phase of Yamato-7301 (YAGI *et al.*, 1978). The results show the effect of weathering on ^{53}Mn content is of minor importance for the interpretation of the data. The depletion of ^{53}Mn in the oxidized phase is estimated to be less than 30%.

For noble gases, GIBSON and BOGARD (1978) have observed the decrease of noble gas content with increasing terrestrial ages in their investigation of Holbrook meteorites. They ascribed the effect mainly to the corrosion of the metal phase followed by the degassing of spallogenic noble gases. The effect was prominent for ^{3}He and ^{38}Ar, which are contributed in a large proportion from metallic iron, but not very significant for ^{21}Ne. In support of this, the ^{21}Ne exposure ages for antarctic meteorites have been found to be longer than the ^{3}He and ^{38}Ar ages (WEBER and SCHULTZ, 1978). In light of this, noble gas data in antarctic meteorites need to be carefully interpreted.

3.3. *Exposure histories of Yamato-7301 and Allan Hills-76008 and their implication to the formation mechanism of meteorites*

As pointed out by us in the previous papers (NISHIIZUMI *et al.*, 1978, 1979a), the contents of cosmic ray produced radionuclides and rare gases in Yamato-7301 and Allan Hills-76008 require a two-stage irradiation model (a simplified version of a multi-stage irradiation model). Table 4 summarizes the data of ^{53}Mn, ^{10}Be, ^{26}Al, ^{36}Cl and pertinent rare gas data. The ^{36}Cl-^{36}Ar exposure ages are calculated assuming the production ratio of $P(^{36}Cl)/P(^{36}Cl+^{36}Ar)=0.83$ (SCHAEFFER and HEYMANN, 1965; SHIMA *et al.*, 1969) and the branching coefficient for ^{36}Cl β^--decay to ^{36}Ar as 0.981. The values are not corrected for the decay of ^{36}Cl due to terrestrial age. The data for Yamato-7304 and Bruderheim are also presented for comparison.

In the two-stage irradiation model (NISHIIZUMI *et al.*, 1978, 1979a), the observed activity is expressed in the equation:

$$A = A_0 f_0[1-\exp(-\lambda T_2)]\exp(-\lambda\tau) + A_0 f_p[1-\exp(-\lambda T_1)]\exp[-\lambda(\tau+T_2)]$$

where T_1 is the duration of cosmic ray exposure in a heavily shielded (~meters depth) position of the parent body (1st stage irradiation), T_2 is the time spent as a small pre-atmospheric object ejected from the parent body (2nd stage irradiation), A_0 is the saturated activity for a normal sized meteorite, f_0 is the shielding factor during the second stage irradiation, f_p is the shielding factor for the irradiation in the parent body, τ is the terrestrial age and λ is the decay constant ($\ln 2/t_{1/2}$). The saturation factor, $A/A_0 f_0$, is a function of 4 parameters, f_p, f_0, τ, T_2. T_1 can be calculated from the relation: $T_1 \approx (T_{\text{apparent}} - T_2)/f_p$ where T_{apparent} is calculated from rare gas data (*see footnote* 3) in Table 4). When $f_p/f_0 \ll 1$ or $f_0 \sim 1$, $A/A_0 f_0$ can be approximated as

$$A/A_0 f_0 \approx [1-\exp(-\lambda T_2)\{1-f_p+f_p\exp(-\lambda T_1)\}]\exp(-\lambda\tau). \quad (1)$$

In Fig. 1, $A/A_0 f_0$ is graphically illustrated with the measured data as a function

Table 4. Cosmic ray produced nuclides in two particular antarctic meteorites, Allan Hills-76008 and Yamato-7301, in comparison with Yamato-7304 and Bruderheim.

Meteorite (Class)	Allan Hills-76008 (H6)	Yamato-7301 (H4)	Yamato-7304 (L5)	Bruderheim (L6)
^{53}Mn (dpm/kg Fe) ($t_{1/2}=3.7\times10^6$ y)	22 ± 3 [b] 23 ± 3 [1]	101 ± 6 [a] 109 ± 7 [1]	412 ± 21 [a]	419 ± 38 [i]
^{10}Be (dpm/kg) (1.6×10^6 y)	—	9 ± 1 [b]	19 ± 2 [b]	19 ± 2 [j]
^{26}Al (dpm/kg) (7.2×10^5 y)	11.2 ± 0.4 [d]	29 ± 2 [2]	62 ± 3 [2]	60 ± 6 [j]
^{36}Cl (dpm/kg metal) (3.0×10^5 y)	9.4 ± 1.0 [c]	17.8 ± 1.9 [c]	24.2 ± 2.5 [c]	28.8 ± 3.0 [c] 27.0 ± 1 [l]
^{21}Ne$_c$ (10^{-8}ccSTP/g)[3]	0.61 [f] 0.81 [h] 0.77 [n]	5.7 ± 1.6 [e]	8.2 ± 1.0 [e]	8.9 [f] 10.1 [m]
^{36}Cl-^{36}Ar age (my)[4]	—	17	18	22
$(^{22}Ne/^{21}Ne)_{bulk}$	1.06 [f] 1.09 [h] 1.05 [n]	1.12 [e]	1.09 [e]	1.10 [f, m]
$^{21}Ne_{bulk}/^{38}Ar_{metal}$[5]	—	5.8 [g]	5.4 [g]	4.7 [k]

1) Data for metal phase (>300 μm).
2) The values are slightly different from those in NISHIIZUMI *et al.* (1979a) due to the improvement in counting statistics.
3) Cosmogenic ^{21}Ne content. Apparent cosmic ray exposure ages based on ^{21}Ne are calculated assuming the production rate, P_{21}, from the relation: $T={}^{21}Ne_c/P_{21}$. According to HERZOG and ANDERS (1972), $P_{21}=0.433$ and 0.466×10^{-8} cc STP/g/my for H and L chondrites, respectively.
4) ^{36}Ar data from *g*) for Yamato-7301 and -7304, and from *k*) for Bruderheim. Calculated ages given here are not corrected for terrestrial ages.
5) $^{38}Ar_{metal}$: ^{38}Ar in metal phase of 150–300 μm.

References: *a*) NISHIIZUMI *et al.* (1978). *b*) NISHIIZUMI *et al.* (1979a). *c*) NISHIIZUMI *et al.* (1979b). *d*) EVANS and RANCITELLI (1979). *e*) TAKAOKA and NAGAO (1978). *f*) NAGAO and TAKAOKA (1979). *g*) TAKAOKA and NAGAO (1979). *h*) WEBER and SCHULTZ (1978). *i*) ENGLERT and HERR (1978). *j*) HONDA *et al.* (1961). *k*) NYQUIST *et al.* (1973). *l*) BEGEMANN and VILCSEK (1969). *m*) SCHULTZ and KRUSE (1978). *n*) FIREMAN *et al.* (1979).

of the half life of the radioactivity for the various sets of parameters which can best explain the data of Yamato-7304, -7301 and Allan Hills-76008. The measured values are plotted assuming A_0 values of 25 dpm ^{36}Cl, 58 and 62 dpm ^{26}Al for H and L chondrites, respectively, 19 dpm ^{10}Be and 430 dpm ^{53}Mn. For Yamato-7301, a rather small value of 0.8 was assumed for the shielding factor f_0 of the low energy products, ^{26}Al and ^{53}Mn, while f_0 for the other nuclides was taken as 1.0. This means that Yamato-7301 was a small pre-atmospheric meteorite and the build-up of secondary cosmic rays was inefficient. In the other cases, f_0 was

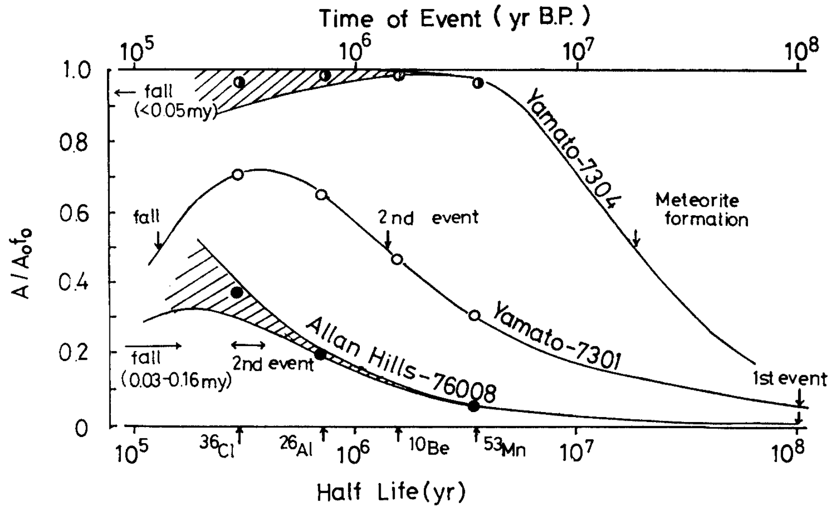

Fig. 1. The saturation factor, A/A_0f_0, of radioactivity is shown as a function of half life for the various sets of parameters which correspond to the exposure histories of Yamato-7304, Yamato-7301 and Allan Hills-76008. The parameters (f_p, T_2 and τ) in eq. (1) are assumed to be (0, 18 my and <0.05 my), (0.1, 1.3 my and 0.13 my) and (0.01–0.24 my and 0.03–0.16 my), respectively. T_1 for Yamato-7301 and Allan Hills-76008 is calculated to be $\sim 10^8$ years for both cases. Shaded area represents the range of uncertainty due to the uncertainty in the estimation of τ (NISHIIZUMI et al., 1979b) for cases of Yamato-7304 and Allan Hills-76008. Also shown is the time scale of events for the meteorites. The details are referred to in the text.

assumed to be 1.0 for all the nuclides. The estimation of terrestrial age depends largely on the ^{36}Cl data, and the estimated values of τ have rather large uncertainties (NISHIIZUMI *et al.*, 1979b). The range of uncertainty is shown in Fig. 1 as shaded area for cases of Allan Hills-76008 (τ=0.03–0.16 my) and Yamato-7304 ($\tau \lesssim 0.05$ my).

The two-stage irradiation model discussed above assumes the pre-irradiation occurred in the deep part of the parent body. If this is the case for Yamato-7301 and Allan Hills-76008, isotopic evidence of heavy shielding might be found in rare gas data. The $^{22}Ne/^{21}Ne$ ratio have widely been used as the index of irradiation hardness. The $^{22}Ne/^{21}Ne$ ratio corresponding to the 1st stage irradiation for Yamato-7301 is 1.10–1.11, corrected for the small contribution of hard irradiation in the 2nd stage. The ratio for Allan Hills-76008 is 1.05–1.09. These values are compared to the value, 1.10, for Yamato-7304 and Bruderheim which are estimated to be normal sized meteorites with normal exposure histories. The $^{21}Ne_{bulk}/^{38}Ar_{metal}$ ratio is also useful as an indicator of shielding (NYQUIST *et al.*, 1973) since the ^{21}Ne in the bulk sample is a low energy product produced mainly from Mg (mass difference: ΔA=3) while ^{38}Ar in the metallic phase is a high energy product from Fe ($\Delta A=18$). Spallogenic ^{38}Ar contents in the metallic phases of Yamato-7301 and -7304 (the 150–300 μm size fraction containing 1.4% and 0.5% silicates) has been determined as 0.99×10^{-8} ccSTP/g and 1.53×10^{-8} ccSTP/g, respectively, by TAKAOKA and NAGAO (1979). The $^{21}Ne_{bulk}/^{38}Ar_{metal}$ ratios are calculated to be 6.2±1.8 (normalized to L chondrite composition) for Yamato-7301 and 5.4±0.7 for Yamato-7304. The value for Yamato-7301 indicates heavy shielding, however, it is difficult to be sure of this due to the large errors involved.

It seems that the above isotopic ratios used as a measure of the shielding effect for cosmic radiation are sensitive only for a few tens of centimeters from the surface and more or less saturated for the more heavily shielded conditions. This is due to the equilibrated shape of the energy spectrum of low energy secondaries after a few interaction lengths.

So far we have assumed that the pre-irradiation of the meteorite immediately preceded the second stage exposure as a small body. This assumption depends largely on the interpretation of the ^{53}Mn data. Some ambiguities remain, however, in the estimation of f_0 and in the extent of the constancy of galactic cosmic ray intensity. A pre-irradiation in a period which does not immediately precede the 2nd stage is equally probable if we assume a somewhat larger production rate, for example 600 atoms/min/kg Fe, for ^{53}Mn. This is a case that assumes a significant variation of cosmic ray intensity in the past 10^7 years (YANAGITA and IMAMURA, 1979). With this higher production rate, all the ^{53}Mn could be produced during the second stage. There would be no need for ^{53}Mn production during the first stage. Rare gases would be produced mainly during a time period that did not immediately precede the 2nd stage. It could be the late accretional stage of an asteroid, during which there must have been a high probability that a large part of the asteroidal surface was exposed to galactic cosmic radiation due to extensive gardening by heavy bombardment with meteorites. PELLAS (1973) found that in several gas-rich chondrites some of the clasts had different exposure histories which suggests pre-irradiation before the compaction of the meteorite. He argued that the pre-irradiation found in the xenolithic chondrites must have occurred 3.5–4.5 billion years ago. Some of these meteorites, however, could have been brecciated relatively late in the history of asteroids as evidenced by the study of the St. Mesmin chondrite by SCHULTZ and SIGNER (1977). These observations suggest that there are meteorites which came from the near surface layer of a parent body that had experienced a pre-irradiation which occurred some time in the past.

Cosmic ray produced ^{40}K is useful to determine when such a pre-irradiation had occurred. We have undertaken the measurements of ^{40}K in the metallic phase for several antarctic meteorites including Yamato-7301 and Allan Hills-76008. Preliminary results show ^{40}K in Yamato-7301 is not significantly decayed. This supports our model of a two-stage irradiation that 1st stage irradiation immediately preceded the 2nd stage.

It is noteworthy to recall that the two antarctic meteorites found to have very young exposure age (<2 my) show evidence of pre-irradiation. We are not sure if this is significant or not. HEYMANN and ANDERS (1967) and FUSE and ANDERS (1969) in their comparative study of ^{26}Al and rare gas contents have found evidence for pre-irradiation in two meteorites out of 13 stony meteorites with exposure ages <2.5 my. Three L chondrites and one LL chondrite are included in the list of 13 meteorites, but none of ordinary chondrites were found to be pre-irradiated. These 13 meteorites would be worthwhile re-examining for pre-irradiation by measuring other radionuclides including ^{53}Mn and ^{40}K. The population of pre-irradiated meteorites among those with young exposure ages would provide information concerning the formation mechanism of meteorites, for example, the mean crator depth as noted by FUSE and ANDERS (1969). With the detailed study of cosmic ray produced radionuclides including ^{36}Cl, ^{26}Al, ^{10}Be, ^{53}Mn and ^{40}K, it may be possible to investigate the details of the surficial processes in the parent body as well.

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