Meteoritics & Planetary Science 41, Nr 4, 529–540 (2006)
Abstract available online at http://meteoritics.org

An improved extraction system to measure carbon-14 terrestrial ages of meteorites and pairing of the Antarctic Yamato-75097 group chondrites

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(Received 3 July 2005; revision accepted 11 November 2005)

Abstract–We examined an improved system for extraction of carbon from meteorites, using a vacuum-tight RF melting method. Meteorite samples mixed with an iron combustion accelerator, including a specific amount of carbon (0.052%), were combusted in a RF furnace (LECO HF-10). $^{14}CO_2$ extracted from the meteorite was diluted with a known amount of nearly ^{14}C-free CO_2 ,evolved from the iron accelerator on combustion. The ^{14}C activities of the recently fallen Holbrook (L6) and Mt. Tazerzait (L5) meteorites were measured by this method. The mean value was 56.5 ± 3.0 dpm/kg, which is similar to the values reported for recently fallen L6 chondrites. Furthermore, terrestrial ages were measured for four Antarctic meteorites: 1.8 ± 0.5 kyr for Yamato (Y-) 75097 (L6), 1.8 ± 0.5 kyr for Y-75108 (L6), and 0.1 ± 0.1 kyr for Y-74192 (H5). For Y-74190 (L6), an apparent age of 0.8 ± 0.5 kyr was calculated. After consideration of the shielding effect by using $^{22}Ne/^{21}Ne$ values, we obtained about 1.8 kyr for the terrestrial age of this chondrite. The five samples Y-74190, Y-75097, and Y-75108, together with Y-75102 (L6) and Y-75271 (L6), have been reported to be paired and fragments of an L-chondrite shower (Honda 1981; Takaoka 1987). The result of this work and literature data for the latter two samples confirmed that they are paired. More discussion and experimental work are needed for other recently fallen meteorites, both for L and H chondrites, and a correction for the shielding effect should be done to determine a more reliable terrestrial age.

INTRODUCTION

Terrestrial ages of Antarctic meteorites give us important information for estimating the terrestrial history of the meteorites, such as accumulation mechanisms and glacial movements. An Antarctic meteorite has a terrestrial age, which is the sum of the period of travel within a glacier from the point at which it fell to to the Earth to the period between its arrival in the area of accumulation until collection. The time required for burial within the glacier from its surface is short and negligible. Antarctic meteorites have old terrestrial ages (e.g., Jull 2001; Jull et al. 1998) relative to the other meteorites and have experienced little weathering and meteorite destruction because of the cold and dry conditions of Antarctica.

Carbon-14 has a short half-life of 5730 yr and is useful for determining the relatively young ages, up to 50–60 kyr, that are often observed in the Yamato ice field region of Antarctica (e.g., Beukens et al. 1988; Jull et al. 1993a; Nishiizumi et al. 1989, 1999). Carbon-14 in meteorites is produced primarily by spallation reactions of galactic cosmic-ray particles on oxygen and silicon. Exposure ages of most meteorites in space range from a few million to 50 million years, so their ^{14}C content is in equilibrium with production and decay. After the meteorites fall on Earth, their ^{14}C content only decreases, except for incorporation of atmospheric ^{14}C through carbonate weathering products (e.g., Jull et al. 1993b, 1998).

The first measurement of ^{14}C activities for meteorites was made by using the ^{14}C decay counting method (e.g., Goel and Kohman 1962; Suess and Wänke 1962) four decades ago. Fireman (1983), Fireman and Norris (1981), and Kigoshi and Matsuda (1986) also made some ^{14}C measurements on Antarctic meteorites using similar β-counting methods. The decay counting method needs a large sample (10–100 g), and it was difficult to use such a large sample size for meteorites. On the other hand, the accelerator mass spectrometry (AMS) method, which directly detects the number of ^{14}C atoms in a sample relative to the number of ^{13}C or ^{12}C atoms, can be used with a smaller sample of less than 1 g. Therefore, with the advent of AMS, many ^{14}C measurements have been performed by the Toronto and Arizona AMS groups, and others (e.g., Brown et al. 1984; Jull et al. 1984, 1993a, 1993b, 1994, 1998; Beukens et al. 1988; Cresswell et al. 1993).

Minami and Nakamura (2001) have constructed a system to extract carbon from meteorites using a vacuum-tight RF melting method, similar to Jull et al. (1993b). With this method, we measured terrestrial ^{14}C ages of two Antarctic meteorites, Yamato (Y-) 75102 (L6) and Allan Hills (ALH) 77294 (H5), from the Yamato and Allan Hills ice fields, and obtained results for terrestrial ages of 4.0 ± 1.0 kyr and 19.5 ± 1.2 kyr, respectively, roughly agreeing with the values reported by Fireman (1983) and Fireman and Norris (1981). However, there remain problems for our method: 1) One is a fixed loss of sample CO_2 during combustion and/or CO_2 separation from O_2 in the glassline system connected to a RF furnace, and the effect is large when the carbon extracted is low (Minami and Nakamura 2001). The loss of CO_2 gas could be caused on removing O_2 from the mixed gas of CO_2 and O_2 collected in liquid nitrogen traps. 2) Another is the existence of experimental blank, derived from O_2 flow gas, an iron combustion accelerator, an alumina crucible, and so on when carbon is extracted from a meteorite, and the effect is also large when the carbon extracted is low. The other is the remains of terrestrial weathering in meteorites. In order to solve the above analytical problems, and to obtain a more accurate ^{14}C terrestrial age of a meteorite, we developed an improved method for a meteorite sample in this study.

Another purpose of this study is the identification of paired meteorites. Antarctic meteorites are composed of overlapping falls, which are superimposed mixtures of meteorite showers, and multiple or single meteorite falls (Honda 1981). Pairing means identifying collected meteorite specimens as fragments of a single fall. Lindström and Score (1995) suggested that the frequent number of Antarctic meteorites could be paired. For identifying paired meteorites, the occurrences and mechanical matching of recovered objects, chemical, and petrological characterization are used. Terrestrial ages determined by cosmogenic nuclides can be used to confirm pairing as well (e.g., Kigoshi 1986; Takaoka 1987). Many "Yamato meteorites" were recovered from the Yamato ice field during annual Japanese Antarctic expeditions. Most Yamato meteorites are small stones of 1–1000 g, and small pieces were recovered in small and narrow regions on the order of 10^3–10^4 m^2. Samples might be fragments of a large object or members of a shower, and they seem to belong to a single meteorite fall. Y-75102 has been reported to be paired with Y-74190 (L6), Y-75097 (L6), Y-75108 (L6), and Y-75271 (L6) (Honda 1981; Takaoka et al. 1981; Takaoka 1987). Honda (1981) also reported that the samples from Y-75108 to Y-75257, which were recovered from very small areas of 10 m × 50 m, seem to have originated from a meteorite. These samples were found at 71°50′S, 35°30′E in the Yamato Mountains, eastern Queen Maud Land, Antarctica. The group belongs to an L chondrite shower that deposited more than 20 kg in total and the collection field of these stones is extended more than 10 km. The summarized data for cosmogenic and for elemental compositions, noble gas, and cosmogenic nuclides, for these Yamato meteorites are summarized in Table 1. The meteorites are characterized by very low contents of radiogenic ^{4}He and ^{40}Ar. Their K-Ar ages are about 0.6 Gyr, a typical age for an L chondrite with a thermal history of collisional metamorphism (Takaoka et al. 1981). The irradiation history estimated from the ^{53}Mn radioactivity (Nishiizumi et al. 1980) as well as cosmogenic rare gas isotopes is compatible with their interpretation as paired meteorites.

Minami and Nakamura (2002) measured ^{14}C contents of Y-74190, Y-75097, and Y-75108 in addition to Y-75102, and reported that the three chondrites except for Y-74190 might have fallen at the same time. In this study, we analyze the Yamato chondrite samples using our improved method to clarify whether the samples have the same terrestrial ages after correction for the shielding effect by using $^{22}Ne/^{21}Ne$ values.

SAMPLES

Two recently fallen meteorites, Holbrook (L6) and Mt. Tazerzait (L5); three Yamato L6 chondrites, Y-74190, Y-75097, and Y-75108; and one H5 chondrite, Y-74192, were analyzed for ^{14}C concentrations. Y-74192 was found and collected at the same point as the L6 chondrites named above. The Holbrook meteorite fell in Navajo County, Arizona, USA (34°54′N, 110°11′W) on July 19, 1912, and the Mt. Tazerzait meteorite fell on Mount Tazerzait Tahoua, Niger (18°42′N, 04°48′E), on August 21, 1991. Total known weights are 218 kg for the Holbrook and 110 kg for the Mt. Tazerzait. The Holbrook and the Mt. Tazerzait meteorite samples were obtained from the Hori mineralogy and the Iwamoto mineral Co., Ltd., respectively. The Antarctic samples were provided by the National Institute of Polar Research (Tokyo).

EXPERIMENTAL PROCEDURE

Combustion

In Minami and Nakamura (2001, 2002), CO_2 extracted from meteorite samples was diluted with ^{14}C-free CO_2 after combustion, while in this study, samples were treated with H_3PO_4 before combustion to move weathering carbonates, and then mixed with the iron combustion accelerator containing a known amount of carbon, and then combusted. In this improved method, CO_2 extracted from a meteorite is diluted with a known amount of nearly ^{14}C-free CO_2 evolved from the iron accelerator at the same time on the combustion,

Table 1. Data compiled of chemical composition, major element composition (%), concentration of cosmogenic and radiogenic isotopes, and cosmic-ray and gas retension ages of the Antarctic meteorites of the Y-75097 group. Data of chemical composition and major element composition are from Yanai et al.(1995). Olivine composition is in mole percent Fe_2SiO_4 (Fa). Pyroxene composition is in mole percent $FeSiO_3$ (Fs). Rare gas data are from Takaoka et al. (1981) and Honda (1981). ^{53}Mn data are from Nishiizumi et al. (1980).

	Y-74190	Y-75097	Y-75102	Y-75108	Y-75110	Y-75271	Y-74192
	L6	L6	L6	L6	L6	L6	L6
Weight (kg)	3.236	2.570	11.000	0.591	0.707	1.799	0.420
%Fa in olivine	24.5	24.2	24.3	24.4	24.3	24.3	18.2
%Fs in pyroxene	20.6	20.1	20.9	20.8	20.7	20.5	15.8
SiO_2	39.24	39.71	39.33	–	39.34	–	35.62
TiO_2	0.10	0.21	0.07	–	0.06	–	0.11
Al_2O_3	2.83	2.60	2.61	–	2.77	–	2.87
Fe_2O_3	–	–	0	–	0.00	–	–
FeO	14.05	15.77	14.01	–	13.99	–	12.12
MnO	0.32	0.34	0.34	–	0.36	–	0.31
MgO	25.86	26.03	25.37	–	25.62	–	23.77
CaO	1.91	1.82	1.63	–	1.61	–	1.66
Na_2O	0.97	0.95	0.92	–	0.92	–	0.78
K_2O	0.12	0.08	0.07	–	0.08	–	0.07
$H_2O(-)$	0.00	0.02	0.03	–	0.00	–	0.21
$H_2O(+)$	0.1	0.0	0.0	–	0.00	–	0.80
P_2O_5	0.25	0.26	0.32	–	0.28	–	0.23
Cr_2O_3	0.51	0.58	0.53	–	0.61	–	0.53
NiO	–	0.590	–	–	–	–	0.74
FeS	6.13	5.94	6.12	–	6.11	–	5.32
Fe	6.55	4.88	7.39	–	7.09	–	13.60
Ni	1.11	0.73	1.12	–	1.05	–	1.10
Co	0.04	0.008	0.042	–	0.04	–	0.01
Total	100.09	100.51	99.90	–	99.93	–	99.84
Total Fe	21.36	20.91	22.17	–	21.84	–	26.40
$(^{3}He/^{21}Ne)c$	2.2	4.6	4.8	4.0	–	4.7	–
T_{21} (Myr)	22	18	17	19	–	19	–
$(^{22}Ne/^{21}Ne)c$	1.053	1.087	1.079	1.089	–	1.095	–
^{40}Ar	159	227	220	241	–	240	–
^{53}Mn (dpm/kg Fe)	441	424	452	407	–	424	–

and the total combined CO_2 of a meteorite and the combustion accelerator are analyzed.

Another improvement point is that we used the decreased amount of a sample (~0.5 g) and the increased amount of a combustion accelerator (~3 g) to combust a sample completely in the new method though about 1 g of a sample was mixed with 2 g of a combustion accelerator in the old method. These improvements can produce some advantages: Smaller sample size, complete combustion of meteorite samples, removal of terrestrial weathering from samples, and reduction in effect of fixed loss of CO_2 gas extracted from samples because the total amount of gas extracted from a sample and 0.052% C steel is larger.

For a combustion accelerator, carbon steel for minor element determination (catalog no. JSS 170-7) supplied by the Iron and Steel Institute of Japan was used. The carbon content in the ISI-certified carbon steel is 0.0519 ± 0.0012 wt%. The reason for the selection of steel is its carbon content. About 1 mg of carbon is obtained on combustion of about 3 g of the steel, and the amount of carbon is suitable for measurements of ^{14}C concentration and δ^{13}C.

The experimental procedure is as follows: a meteorite sample is crushed and then treated with H_3PO_4 (85%) to remove carbonates. The sample is then washed with distilled water and dried before about 0.5 g of the sample and about 3 g of the combustion accelerator are mixed and placed in an alumina crucible which had been preheated at 1000 °C for 10 h previously. In order to remove terrestrial contaminants and atmospheric ^{14}C, the sample and combustion accelerator in the crucible are preheated at 500 °C for 1 hr in air in a muffle furnace (Jull et al. 1989; Lifton et al. 2001). The preheated sample and combustion accelerator in the covered crucible are then placed in an RF induction furnace (LECO HF-10), and heated in a flow of purified oxygen for up to 4 min. A gas flow meter was introduced to make the total flow of oxygen on combustion constant, and hence to try to keep the C/O ratio constant. The sample gas is passed through MnO_2 at room temperature and through Pt/CuO at

450 °C. The former removes SO_2 and the latter ensures conversion of any CO to CO_2 in the gas. Then the CO_2 gas is collected in three liquid nitrogen traps, and the O_2 is pumped off slowly. The CO_2 extracted from the sample is then graphitized by reduction with hydrogen in a Fe-powder catalyst at 650 °C in a sealed Vycor tube, and the graphite produced is measured of its ^{14}C concentration using a Tandetron accelerator mass spectrometer at the Center for Chronological Research, Nagoya University. Standards used were graphites made from NIST standard oxalic acid (SRM-4990C). The $\delta^{13}C$ value for the sample gas was obtained on a Finnigan MAT-252 mass spectrometer using part of the CO_2 gas before graphitization.

Calculation of Terrestrial Ages

The total amount of carbon extracted from a meteorite sample (abbreviated as "sam") and a combustion accelerator of 0.052% C steel (abbreviated as "com"), $C_{sam+com}$, can be measured after combustion. The amount of carbon extracted from the combustion accelerator alone, C_{com}, is determined by using its weight and carbon content. The amount of carbon in a sample, C_{sam}, can be obtained by subtracting the amount in the combustion accelerator from the amount in the mixture of the sample and the steel. The $^{14}C/^{12}C$ ratio of a meteorite divided by $^{14}C/^{12}C$ of the oxalic acid standard, $(^{14}C/^{12}C)_{sam}/(^{14}C/^{12}C)_{std}$ ratio, is calculated by the following equation:

$$\begin{aligned}&(^{14}C/^{12}C)_{com}/(^{14}C/^{12}C)_{std} \times C_{com}/C_{sam+com} + \\ &(^{14}C/^{12}C)_{sam}/(^{14}C/^{12}C)_{std} \times C_{sam}/C_{sam+com} = \\ &(^{14}C^{12}C)_{sam+com}/(^{14}C/^{12}C)_{std}\end{aligned} \tag{1}$$

The activity of a sample, A_{sam} (dpm/kg), is calculated by using the following equation:

$$(^{14}C/^{12}C)_{sam}/(^{14}C/^{12}C)_{std} = A_{sam} \times W_{sam}/V_{CO_2}/D \tag{2}$$

where W_{sam} is the weight of the sample in kg, V_{CO2} is the volume of CO_2 at STP extracted from a sample, and D is the abundance of ^{14}C in the oxalic acid standard: 9.7432×10^{-3} ^{14}C min^{-1} cm^{-3} CO_2. A terrestrial age is calculated by the following equation:

$$\text{Terrestrial age} = -T_{1/2} / \ln 2 \times \ln (A_{sam}/A_{sat}) \tag{3}$$

where $T_{1/2}$ is the half-life for ^{14}C, which is 5730 yr, and A_{sam} is saturated activity.

In this study, four different experiments were made: 1) combustion of a combustion accelerator alone to determine the mean value of CO_2 extracted from the accelerator; 2) combustion of a combustion accelerator together with quartz to examine if the amount of CO_2 extracted from a combustion accelerator is changed by addition of materials other than the combustion accelerator; 3) analysis of recently fallen meteorites to obtain the saturated activity for L chondrites; and 4) analysis of three Y-75097 group chondrites to determine their terrestrial ages and to investigate their pairing together with Y-75102.

RESULTS AND DISCUSSION

Blank Correction and Dilution of Gas

About 3 g of the combustion accelerator of 0.052% C steel was placed into a crucible and combusted in a RF furnace to determine the carbon amount extracted from all materials other than the meteorite sample. The combustion experiments were performed 20 times to examine the reproductively of the data. The results are shown in Table 2. The average value of carbon yield was 84.9 ± 2.6 % (mean ± 1 standard deviation). There was a little scatter of the yield data. The carbon content of the steel is not completely homogeneous, and the scatter might be due in part to the heterogeneous carbon contained. The deviation of the carbon yield, however, is slightly larger than that of the carbon content, and it might be caused by analytical errors, such as an error derived from experimental blank from a crucible and O_2 flow gas, in addition to the heterogeneity in the steel.

The result of combustion of the steel together with terrestrial quartz is also shown (Table 2). Compared with combustion of the steel alone, almost the same amount of carbon was extracted upon combustion of 0.25 g of quartz, while less carbon was extracted on combustion of 0.5 g of quartz. Nearly the same amount of carbon should be extracted from the mixture of steel and quartz because quartz contains no carbon. Therefore, the decreased amount of CO_2 upon combustion of quartz was due to a decreased amount of carbon extracted from the steel. On combustion of the steel, an electric conductor, mixed with quartz, an insulator, the electric current produced by electromagnetic induction in a RF furnace might decrease and the absorption efficiency of RF into a sample could be reduced lower. The same phenomenon could occur on combustion of carbon from a meteorite. Since less carbon was extracted on combustion of 0.5 g of quartz, which is the same as the weight of meteorite sample used, we will use the carbon yield of 80.3 ± 3.8% on combustion of a crucible with the steel and quartz as the total amount of carbon extracted from the steel and the crucible. This allows an estimate of the dilution factor and the blank to be made from the quartz results.

The $^{14}C/^{12}C$ ratio was unchanged during combustion of the steel alone or on combustion of the steel together with quartz, regardless of changes of carbon extraction yield (Fig. 1), and also did not change systematically with the change of $\delta^{13}C$ (Fig. 2), thus carbon isotope in extracted gas was not different with different efficiencies of steel combustion. The means of the $(^{14}C/^{12}C)_{com}/(^{14}C/^{12}C)_{std}$ ratio and $\delta^{13}C$ on combustion of quartz were 0.0199 ± 0.0010 ($n = 5$), and the mean of $\delta^{13}C$ is -25.3 ± 0.3 ‰ ($n = 5$), respectively, similar to those on the combustion of the steel alone, within the errors.

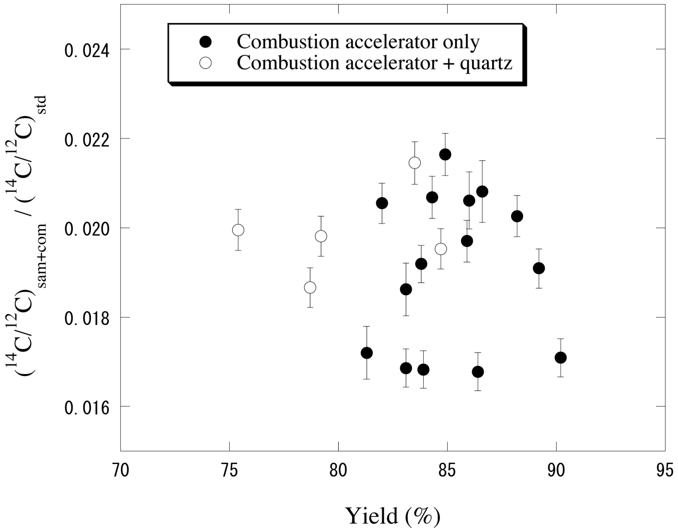

Fig. 1. A plot of $(^{14}C/^{12}C)_{sam + com}/(^{14}C/^{12}C)_{std}$ versus carbon yield (wt%) on combustion of 3 g of 0.052% C steel alone and of a mixture of the steel and 1 g of quartz.

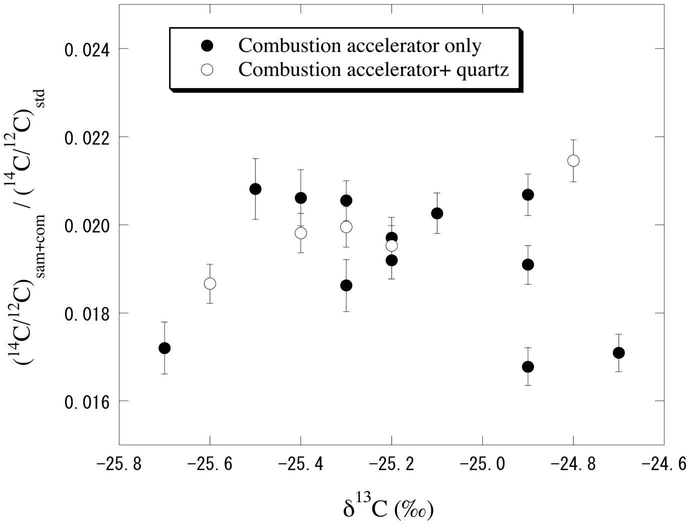

Fig. 2. A plot of $(^{14}C/^{12}C)_{sam + com}/(^{14}C/^{12}C)_{std}$ versus $\delta^{13}C$ (‰) on combustion of 3 g of 0.052% C steel alone and of a mixture of the steel and 1 g of quartz.

Recently Fallen Meteorites

The ^{14}C activity of a recently fallen meteorite is regarded as the saturated activity of the meteorite in space, and we can estimate the terrestrial age of the other meteorite by comparing its ^{14}C activity with its saturated ^{14}C activity. Because ^{14}C in a meteorite is produced by a spallation reaction of cosmic-ray particles on the O of silicate (Jull et al. 1989; Sisterson et al. 1994), the ^{14}C content depends on the O content of the meteorite. Every meteorite has a different chemical composition, size, and location in the meteoroid, and the irradiation conditions at the location of the sample in the meteoroid are also different (Born and Begemann 1975). Each meteorite, therefore, has an individual saturated ^{14}C activity, but it is impossible to know the actual saturated activity of a meteorite. On the other hand, meteorites of the same classes generally have similar saturated activity, summarized by Jull et al. (1998). We use the mean value of saturated activity for the same class of recently fallen meteorites.

In this study, we analyzed saturated ^{14}C activities of two recently fallen meteorites, Holbrook and Mt. Tazerzait, which are completely saturated for ^{14}C. The ^{14}C activities were slightly different between Holbrook and Mt. Tazerzait (Table 3). The Holbrook meteorite was reported to be 58.9 ± 0.5 dpm/kg in Jull et al. (1989) and 44 ± 1 dpm/kg in Jull et al. (1998). The latter data is obtained by combustion after phosphoric acid treatment of the meteorite.

Table 2. Result of CO_2 extraction experiment for determination of blank value.

No.	Sample	Weight of sample (g)	Weight of combustion accelerator (g)	C_{com} (mg)	Yield (wt%)	$\frac{(^{14}C/^{12}C)_{sam + com}}{(^{14}C/^{12}C)_{std}}$	Lab. code # (NUTA2-)	$\delta^{13}C$ (‰)
1	–	–	3.008	1.320	84.3	0.0207 ± 0.0005	4283	–24.9
2	–	–	3.003	1.280	82.0	0.0206 ± 0.0005	4284	–25.3
3	–	–	3.007	1.310	83.8	0.0192 ± 0.0004	4285	–25.2
4	–	–	3.000	1.300	83.1	0.0186 ± 0.0006	4286	–25.3
5	–	–	3.003	1.350	86.6	0.0208 ± 0.0007	4290	–25.5
6	–	–	3.010	1.350	86.0	0.0206 ± 0.0006	4291	–25.4
7	–	–	3.001	1.270	81.3	0.0172 ± 0.0006	4292	–25.7
8	–	–	3.005	1.330	84.9	0.0216 ± 0.0005	4293	–
9	–	–	3.016	1.350	85.9	0.0197 ± 0.0005	4455	–25.2
10	–	–	3.003	1.298	83.1	0.0169 ± 0.0004	4456	–
11	–	–	3.001	1.393	89.2	0.0191 ± 0.0004	4457	–24.9
12	–	–	3.003	1.281	82.0	–	–	–
13	–	–	3.006	1.311	83.9	0.0168 ± 0.0004	4458	–
14	–	–	3.009	1.334	85.2	–	–	–
15	–	–	3.004	1.378	88.2	0.0203 ± 0.0005	4463	–25.1
16	–	–	3.020	1.285	81.8	–	–	–
17	–	–	3.002	1.348	86.4	0.0168 ± 0.0004	4465	–24.9
18	–	–	3.004	1.409	90.2	0.0171 ± 0.0004	4466	–24.7
				Average ± 1σ	84.9 ± 2.6	0.0193 ± 0.0021		–25.2 ± 0.3
1	Quartz	0.250	3.001	1.341	85.9	–	–	–
2	Quartz	0.250	3.003	1.309	83.8	–	–	–
3	Quartz	0.250	3.000	1.328	85.1	–	–	–
				Average ± 1σ	84.9 ± 1.0			
1	Quartz	0.501	3.000	1.302	83.5	0.0215 ± 0.0005	4467	–24.8
2	Quartz	0.500	3.003	1.178	75.4	0.0200 ± 0.0005	4470	–25.3
3	Quartz	0.501	3.012	1.326	84.7	0.0195 ± 0.0005	4471	–25.2
4	Quartz	0.502	3.001	1.229	78.7	0.0187 ± 0.0004	4472	–25.6
5	Quartz	0.502	3.003	1.238	79.2	0.0198 ± 0.0005	4473	–25.4
				Average ± 1σ	80.3 ± 3.8	0.0199 ± 0.0010		–25.3 ± 0.3

Table 3. ^{14}C activities of two recently fallen meteorites.

Sample (class)	Weight of sample (g)	Weight of combustion accelerator (g)	CO_2 (cm^3STP)	CO_2 (cm^3STP)	$\frac{(^{14}C/^{12}C)_{sam}}{(^{14}C/^{12}C)_{std}}$	Lab. code # (NUTA2-)	^{14}C (dpm/kg)
Holbrook	0.502	3.000	0.092	2.639	29.9 ± 0.4	4673	53.6 ± 0.7
(L6)	0.501	3.000	0.147	2.694	19.0 ± 0.4	4674	54.4 ± 0.8
Mt.Tazerzait	0.501	3.000	0.186	2.733	16.0 ± 0.4	4675	57.8 ± 0.8
(L5)	0.501	3.000	0.283	2.830	10.9 ± 0.4	4680	60.1 ± 0.9

Because saturated activity does not appear to differ greatly among L chondrites, the mean activity value of the Holbrook and Mt. Tazerzait chondrites was used as the saturated activity for Y-74190, Y-75108, and Y-75097. Bruderheim, an L6 chondrite that fell in 1960, has been often mentioned as a representative example of an L chondrite in previous reports as listed in Table 4; the activity is 48–55 dpm/kg. The other L6 chondrites show ^{14}C activities of 50–58 dpm/kg (see Table 4). The scatter in the saturated values is presumably due to uncertainty according to the different depth in the meteoroid (Jull et al. 1994; Leya et al. 2000; Schultz et al. 2005). The ^{14}C concentrations of the Mt. Tazerzait and Holbrook chondrites obtained in this study were 54–60 dpm/kg (the mean: 56.5 ± 3.0 dpm/kg), similar to the reported values within the errors. The activity is considered to be reliable and can be used in the dating of the three Antarctic L6 chondrites because they were measured by the same experimental system.

The saturated activity of an H chondrite has been reported to be 42 ± 2 dpm/kg for Torino, an H6 chondrite that

Table 4. Literature data for saturated ^{14}C activities measured on some recently fallen meteorites.

Meteorite	Class	Fall	^{14}C (dpm/kg)	Reference
Holbrook	L6	1912	58.9 ± 0.5	Jull et al. (1989)
			44 ± 1	Jull et al. (1998)
Bruderheim	L6	1960	57 ± 3	Fireman (1983)
			49.8 ± 1.8	Brown et al. (1984)
			51 ± 2	Jull et al. (1993b)
			54.6 ± 0.5	Cresswell et al. (1993)
			47.6 ± 2.0	Knauer et al. (1995)
Leedey	L6	1943	50.0 ± 1.7	Jull et al. (1989)
Mbale	L6	1996	58.1 ± 0.4	Jull et al. (1998)
Peace River	L6	1963	55.1 ± 1.0	Cresswell et al. (1993)
Peekskill	L6	1992	51.1 ± 0.4	Graf et al. (1997)
Torino	H6	1992	42 ± 2	Wieler et al. (1996)
Richardton	H5	1918	34.6 ± 0.8	Jull et al. (1989)
Nuevo Mercurio	H5	1978	35.3 ± 0.4	Jull et al. (1989)

fell in 1992, by Wieler et al. (1996). Low saturated activities were also reported for the Richardton (H5) and Neuvo Mercurio (H5) chondrites by Jull et al. (1989), but these values might be lower than the true activity due to the shielding effects (Born and Begemann 1975). Jull et al. (1998) estimated an average value of the saturated activity for H chondrites by normalization of the mean value of the ^{14}C content of the Bruderheim (51.1 dpm/kg) to the oxygen contents of the H chondrites (Mason 1979). The resulting value is 46.5 dpm/kg for H chondrites. We will use this saturated activity to estimate the terrestrial age of Y-74192.

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The terrestrial ages calculated by using the above equations and the average of saturated activity are shown in Table 5, together with the literature data. The average of saturated activity shows an error of about 5% in this study, but a saturated activity of ^{14}C generally has an error of ±15% due to shielding effects (Jull et al. 1998). Therefore, the errors in the terrestrial ages become larger if shielding effects are considered.

The terrestrial ages in this study are about 2.0 kyr younger in three samples of Y-74190, Y-75108, and Y-75097, compared with the data reported by Minami and Nakamura (2002) shown in Fig. 3. We believe the carbon extraction procedure is an improvement over the experimental procedure used by Minami and Nakamura (2001, 2002). The differences are a) acid treatment of meteorite samples and b) dilution of ^{14}C extracted from meteorites with nearly ^{14}C-free CO_2 on combustion. Therefore, several causes are considered: Effective removal of weathering products from a meteorite by acid treatment, a decrease in ^{14}C contamination upon dilution, and an increase in extraction yield on combustion. A little cosmogenic ^{14}C is lost by acid treatment, but the loss is kept to a minimum. The terrestrial ages of Y-75097 and Y-75108 were 1.8 kyr, similar to the results for Y-75102 obtained previously by Jull et al. (1984) and for Y-75271 obtained by Jull et al. (1993a) and Beukens et al. (1988) (Fig. 3). The data by Fireman (1983), where the ß-counting method is used for ^{14}C measurements, are older than these ages. This might be due to the counting method, different from the AMS method, and/or due to non acid-treatment of the meteorites. Our measurements in this study generally show agreement with the literature results.

On the basis of the ^{14}C terrestrial ages in this study and the literature data, Y-75097, Y-75102, Y-75108, and Y-75271 might be paired, while Y-74190, which has almost the same major element compositions as the other meteorites (Table 1), has a slightly younger terrestrial age, and so its pairing with them is kept ambiguous, judging from these terrestrial ages only. Wieler et al. (1996) studied ^{14}C production rates with different specimens of the Torino H6 chondrite, and reported that the saturated activity in the meteorites of precursor radii from 20 to 45 cm varies from about 38 to 52 dpm/kg. Therefore, a lower saturated activity should be used for the meteorites from apparently smaller meteoroids. Rare gas and other radioisotope data, such as $^{22}Ne/^{21}Ne$ ratios, can be used to estimate the shielding depth of the sample in the meteoroid. Eberhardt et al. (1966) first noted a correlation, named the "Bern line," between $^{3}He/^{21}Ne$ and $^{22}Ne/^{21}Ne$ ratios of bulk samples from different chondrites. This correlation could be attributed to the depth dependence of the two ratios.

Figure 4 shows a correlation plot between $^{3}He/^{21}Ne$ and $^{22}Ne/^{21}Ne$ ratios for the pairing group of meteorites that we name the Y-75097 group. The data of Y-75097, Y-75102, Y-75108, and Y-75271 plot along the Bern line, while Y-74190 is plotted below the line. One of the reasons for this could be cosmogenic ^{3}He (and/or tritium) loss. Because retentivity for He is low, a loss of ^{3}He is recognized in the Bern line, while the cosmogenic $^{22}Ne/^{21}Ne$ ratio is pertinent to discussion on shielding depth. $(^{22}Ne/^{21}Ne)c$ for four specimens fitting the

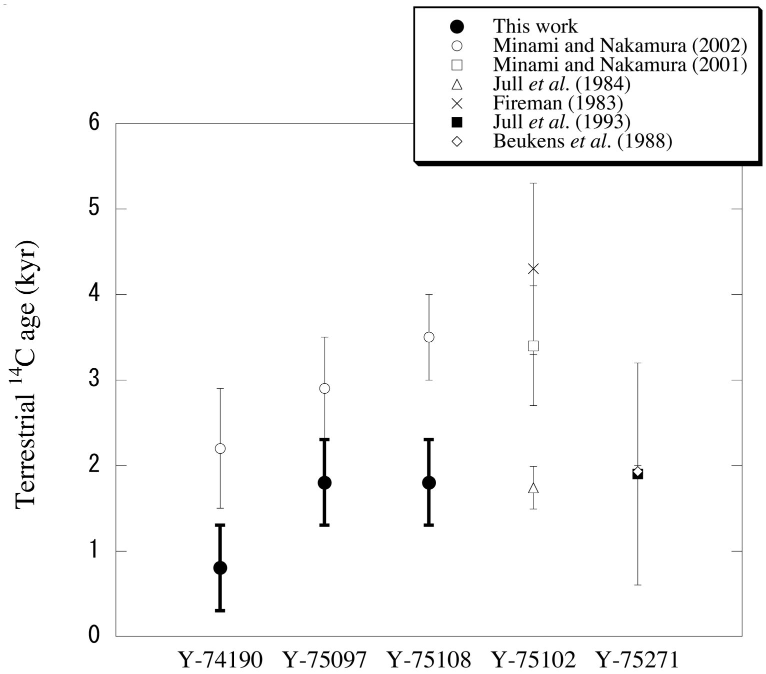

Fig. 3. Carbon-14 terrestrial ages of Y-75097 group chondrites together with some literature data.

Bern line is around 1.09 while that for Y-74190 is 1.053, distinctly lower than the other three samples. This suggests that Y-74190 was irradiated at greater depth than those three specimens.

Leya et al. (2000) presented depth- and size-dependent production rates of cosmogenic nuclides from a physical model with normalization to measured saturation activities. Figure 5 shows the correlation between ^{14}C activity and the cosmogenic shielding indicator $^{22}Ne/^{21}Ne$ ratio according to their data. The dependence of ^{14}C on cosmogenic $^{22}Ne/^{21}Ne$ is observed for L chondrites of preatmospheric radii of 15, 50, and 85 cm (Schultz et al. 2005). The data of the Y-75097 group chondrites and the ones of an L5 chondrite of Knyahinya (Jull et al. 1994) are also plotted in Fig. 5, where a good correlation of ^{14}C with $^{22}Ne/^{21}Ne$ is observed for the Y-75097 group chondrites and the Knyahinya, respectively. According to the data, the samples of Y-75097, Y-75102, Y-75108, and Y-75271 might be inferred to have been exposed to cosmic-ray irradiation at depths of around 20 cm in the meteoroid, while Y-74190 might be derived from a greater depth, that might be >~40 cm, in the meteoroid. The saturated activities, estimated from of terrestrial age of 1.8 kyr, of the Y-75097 group chondrites are also plotted in Fig. 5. It is interesting to note that the plots of the Y-75097 group chondrites fit the trend for the correlation line calculated by Leya et al. (2000). This indicates that Y-74190 is confirmed to be paired with Y-75097, Y-75102, Y-75108, and Y-75271, and that the Y-75097 group chondrites might be fragments of a large object as suggested by Honda (1981), Takaoka et al. (1981) and Takaoka (1987).

The terrestrial age of Y-74192 (H5) is calculated to be 0.1 kyr by using the saturated activity of 46.4 dpm/kg. Nishiizumi et al. (1989) reported the terrestrial age of 20 ± 20 kyr for Y-74192 by ^{36}Cl measurement (Table 5). It is possible, therefore, that Y-74192 is a recently fallen meteorite with activity near to saturated activity. The Yamato site is known to have younger meteorite falls than the other areas in Antarctica, and most of the Yamato meteorites have younger terrestrial ages (e.g., Honda 1981; Beukens et al. 1988; Nishiizumi et al. 1989; Jull et al. 1999). The obtained terrestrial age might not be unusual.

CONCLUSIONS

To measure ^{14}C terrestrial ages of Antarctic meteorites, we have improved the carbon extraction system from meteorites. Total O_2-gas flow was kept constant when meteorite samples were combusted to extract carbon from the samples, and the $^{14}CO_2$ gas extracted was diluted with nearly ^{14}C-free CO_2 extracted from an iron combustion accelerator on combustion in an RF furnace. The terrestrial ages of L6 chondrites Y-74190, Y-75108, and Y-75097 were measured by using the saturated activity of 56.5 ± 3.0 dpm/kg for recently fallen Holbrook and Mt. Tazerzait meteorites. When the results obtained in this study are compared with the literature values, the terrestrial ages were similar to those reported by Jull 17 et al. (1984, 1993a). The age of Y-74192 (H5) was calculated by using a saturated ^{14}C activity of 46.4 dpm/kg as average for H chondrites. The obtained terrestrial age suggests that Y-74192 could be a recently fallen

Table 5. ^{14}C terrestrial ages of some Antarctic Yamato meteorites.

Sample (class)	Weight of sample (g)	Weight of combustion accelerator (g)	CO_2 (cm^3STP)	Diluted CO^2 (cm^3STP)	$\frac{(^{14}C/^{12}C)_{sam}}{(^{14}C/^{12}C)_{std}}$	Lab.code # (NUTA2-)	^{14}C (dpm/kg)	Saturated activity used (dpm/kg)	Terrestrial age (kyr)	Reference
Y-74190	0.500	3.001	0.122	2.669	21.4 ± 0.4	4681	51.0 ± 0.4	56.5 ± 3.0	0.8 ± 0.5	This work
(L6)	1.056	2	–	–	–	–	40.8 ± 1.2	53	2.2 ± 0.7	Minami and Nakamura (2002)
Y-75097	0.500	3.000	0.246	2.793	9.5 ± 0.3	4683	45.6 ± 0.5	56.5 ± 3.0	1.8 ± 0.5	This work
(L6)	1.001	2	–	–	–	–	37.5 ± 1.2	53	2.9 ± 0.6	Minami and Nakamura (2002)
Y-75108	0.500	3.002	0.103	2.650	22.6 ± 0.3	4686	45.2 ± 0.4	56.5 ± 3.0	1.8 ± 0.5	This work
(L6)	0.733	2	–	–	–	–	34.7 ± 1.0	53	3.5 ± 0.5	Minami and Nakamura (2002)
Y-75102 (L6)	0.982	0.015	–	–	–	–	5.3 ± 1.1	53	3.4 ± 0.7	Minami and Nakamura (2001)
	5.0	7.6	–	–	–	–	46.3 ± 1.4	57	1.74 ± 0.25	Jull et al. (1984)
	–	–	–	–	–	–	34.1 ± 2.7	57	4.3 ± 1.0	Fireman (1983)
Y-75271	0.177	3–5	–	–	–	–	40.5 ± 0.6	51.1	1.9 ± 1.3	Jull et al. (1993a)
(L6)	1.123	–	–	–	–	–	39.63 ± 0.25	50.1	1.93 ± 0.07	Beukens et al. (1988)
Y-74192	0.500	3.000	0.288	2.835	10.5 ± 0.4	4682	45.6 ± 0.5	46.4	0.1 ± 0.1	This work
(H5)									20 ± 20	Nishiizumi et al. (1989)

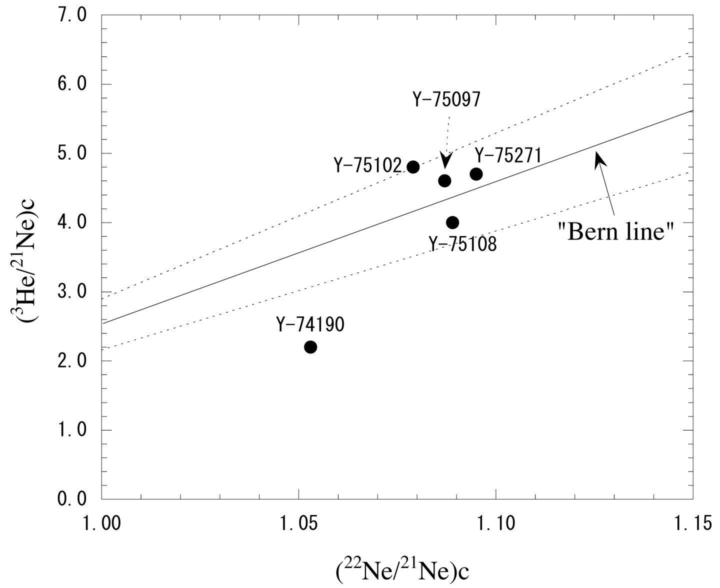

Fig. 4. Correlation between $(^3He/^{21}Ne)c$ and $(^{22}Ne/^{21}Ne)c$ of Y-75097 group chondrites. Data from Takaoka et al. (1981) and Honda (1981). The Bern line (Eberhardt et al. 1966) is a correlation line between $^3He/^{21}Ne$ and $^{22}Ne/^{21}Ne$ ratios of bulk samples from different chondrites. Dashed lines show a ±15% band covering the variation caused by different chemical composition and/or different shape.

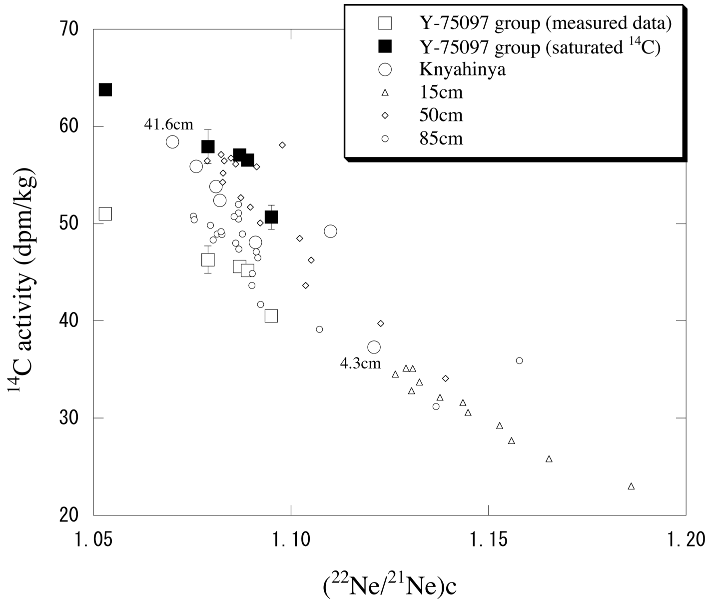

Fig. 5. Correlation between ^{14}C activities and $(^{22}Ne/^{21}Ne)c$ of Y-75097 group chondrites. Their decayed ^{14}C activities (measurement data) and saturated activities estimated by the terrestrial age of 1.8 kyr are plotted. Plots of Knyahinya, which fell in 1988, are also shown. Data of $(^{22}Ne/^{21}Ne)c$ for Knyahinya are from Graf et al. (1990), and ones for ^{14}C activity are from Jull et al. (1994). The depths in cm for each plot of two samples from deeper and shallower depths in Knyahinya are indicated in this figure. The ^{14}C production rates for L chondrites with preatmospheric radii of 15, 50, and 85 cm as a function of $(^{22}Ne/^{21}Ne)c$ are also given according to the data by Leya et al. (2000).

meteorite. It is doubtful, however, that the value of 46.4 dpm/kg is suitable for the saturated ^{14}C activity used in estimating the terrestrial age of Y-74192. In order to improve the accuracy of terrestrial age, better data for the saturated ^{14}C activity for H condrites is needed.

With respect to the pairing of Y-75097 group, the terrestrial ages of Y-75097 and Y-75108 agree with each other, with ages similar to those of Y-75102 and Y-75271 within experimental errors, and so they could be fragments of the same fall. The apparent age for the Y-74190 is younger than the other samples. However, the meteorite may have been irradiated at a deeper depth in the meteoroid than the other three samples on the basis of the $^{22}Ne/^{21}Ne$ and $^3He/^{21}Ne$ ratios, and it may have higher ^{14}C saturated activity.

With the saturated ^{14}C activity corrected for shielding depth, we have the same age as that for the other Y-75097 group specimens. Hence the Y-74190 is confirmed to be paired with Y-75097, Y-75102, Y-75108, and Y-75271. It is important to make shielding correction by $^{22}Ne/^{21}Ne$ ratio for determination of the terrestrial age of a meteorite. If a correlation line of ^{14}C on $^{22}Ne/^{21}Ne$ for a meteorite is fixed by its terrestrial age and shielding depth alone, the terrestrial age corrected for shielding depth could be estimated by measurement of its $^{22}Ne/^{21}Ne$ ratio. On the other hand, the ^{14}C-^{10}Be method can also be used to make shielding corrections because both of these radionuclides are produced by the same spallation reactions on oxygen and because their production rates in meteorites are almost constant (Jull et al. 2001; Kring et al. 2001; Neupert et al. 1997). Therefore, further studies, such as measurement of ^{10}Be, are needed for shielding or depth corrections for determining the ^{14}C terrestrial age of meteorite samples, and then more detailed pairing could be made by use of ^{10}Be for shielding correction.

Acknowledgments–We are grateful to the National Institute of Polar Research for provision of several Antarctic meteorite samples, to Mr. S. Yoshioka and Dr. E. Niu of the Center for Chronological Research, Nagoya University, for their technical assistance, and to Dr. K. Mimura of Graduate School of Environmental Studies, Nagoya University for discussion. We are also grateful to Dr. A. J. T. Jull of the University of Arizona for fruitful advice in starting this research and a constructive review of this manuscript. This paper was also benefited from a helpful review by Dr. M. Honda.

Editorial Handling—Dr. A. J. Timothy Jull

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