COSMOGENIC K-40 IN ANTARCTIC METEORITES

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Abstract: The present work extends the measurements of cosmogenic K-40 by mass spectrometry in the metal phase separated from chondrites and others, especially antarctic meteorites. Potassium-40 ($t_{1/2}$: 1.28×10^9y) is regarded as a stable nuclide rather than radioactive, in the time scale of cosmic ray exposure to chondrites. According to our current sampling, chemical and mass-spectrometric techniques, starting from 10 mg size metal samples and at a contamination level of lower than 10^{-8} g K, we are able to measure the exposure ages as low as one million years.

We determined the production rate of ^{40}K in Fe+Ni relative to the rates of ^{36}Ar and ^{36}Cl, using samples separated from some mesosiderites and irons. Potassium-40 (irradiation) ages obtained in this work were compared with the ages based on the light rare gases contents. Both agreed with each other. Some samples are indicating their near-surface irradiation histories. In a few samples, Yamato-74116 and the Kirin chondrite, multi-stage irradiation histories were detected.

1. Introduction

We have been engaged in the measurements of cosmogenic nuclides, especially long-lived radioactive nuclides, in the antarctic meteorites (NISHIIZUMI *et al.*, 1978, 1979; IMAMURA *et al.*, 1978, 1979). In this report we describe some results of determination for cosmogenic ^{40}K in several chondrites and one iron meteorite recovered from two fields in the antarctic area. Besides, the determinations were also made with several known chondrites and some mesosiderites simultaneously.

As a cosmogenic nuclide, the content of ^{40}K was first determined in iron meteorites by β-ray counting of the radioactivity by HONDA (1959). On the other hand, the measurement for irradiation ages of iron meteorites has been developed, using ratios of a cosmogenic pair of ^{40}K and ^{41}K massspectrometrically by VOSHAGE and HINTENBERGER (1959, 1961). The measurement for concentrations of ^{40}K then has

been extended to chondrites by IMAMURA *et al.* (1975) in our laboratory. Potassium-40, $t_{1/2}=1.28\times10^9$ y, may actually be regarded as stable rather than radioactive, when chondrites have been irradiated not longer than 10^8 y as it is believed in general.

In our previous work, we have had some puzzling discrepancies between production rates of ^{40}K in metal phase and of ^{21}Ne in bulk of chondrites. We therefore started this work with a reexamination of our previous results and with a determination of the relative production rate of ^{40}K, directly comparing other rates in metal phase of chondrites.

In our earlier experiment, the experimental uncertainty reached or exceeded $\pm20\%$ with the meteorites irradiated for shorter than 10 my. Many meteorites, however, have records of 1–10 my. In this study, improvements in the determination had to be achieved. It has been expected that recent developments in the mass-spectrometry may increase the experimental reproducibility and reduce subjective factors and labors remarkably, as compared with the older technique. On the other hand, however, we still had to reduce the sample and laboratory contamination levels. The separation of metal phases free from potassium, or free from the silicates, must be performed at the level of lower than one ppmK in the sample. In the former work, more than 100 mg of metal fractions were dissolved. Recently 10 mg size metal samples are found to be useful to determine ^{53}Mn and ^{36}Cl. We therefore had to improve the method by reducing the contamination to the level of lower than 1 ppmK, starting with 10 mg size metal sample. This provides a good mass-spectrometric sample with some 10^{-9} g of potassium, and the measurement is possible for a few my samples with a reproducibility of $\pm10\%$ or so.

2. Experimental

2.1. Separation of the metal phase from chondrites

A few hundred mg of chondrite samples were crushed in an agate mortar until most of metal grains were isolated, as observed under a binocular microscope. The metallic particles were separated by means of a hand magnet. When coarse metallic particles contained many small silicate inclusions, usually in the case of H chondrites, a medium-size fraction, $-50+100$ mesh, was separated. The metallic particles were treated with 46–50% HF in an ultrasonic bath for *ca.* 30 min, and visible non-magnetic minerals were removed by using a hand magnet. The magnetic fraction was washed well in a dilute HCl and water, dried under an infrared lamp and weighed. A few tens mg of metal were obtained.

2.2. Chemical procedure (*Fig. 1*)

Reagents used in this procedure were purified by distillation, by ion exchange (for H_2O_2) or by solvent extraction (for isopropyl ether) in our laboratory. The

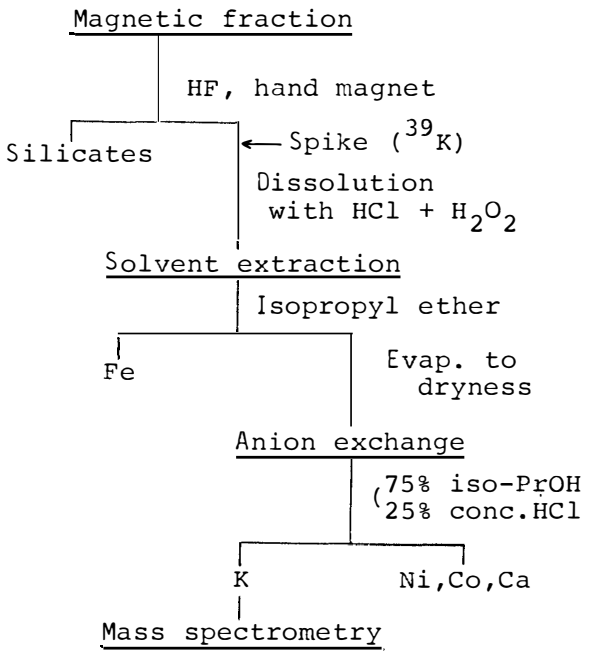

Fig. 1. Chemical procedure for separation of K fraction in metal phase of meteorites. A simplified separation scheme for K and others.

chemical operations were carried out in a clean hood passing through filtrated air. The weighed metal sample was dissolved with 1 m*l* of concentrated HCl in a 5 m*l* teflon vessel, after adding enriched ^{39}K as a spike. Iron in the solution was sufficiently oxidized by an addition of one drop of 30% hydrogen peroxide, and was extracted into isopropyl ether from the 8 N HCl solution. The aqueous phase was evaporated to dryness. An anion exchange resin column (Diaion SA #100, 100–200 mesh, volume: 0.5–1 m*l*) was used for the separation of potassium. The column was conditioned with a mixture of 25% concentrated HCl and 75% isopropyl alcohol. The sample was dissolved in a small volume of the mixture and charged to the column. Nickel, Co, Ca and others were adsorbed in the resin. Potassium fraction was recovered in the effluent of the first two column volumes of the mixture. But the potassium fraction seems to contain small amounts of impurities such as phosphate and sodium.

The level of contamination during the process was determined by using ^{39}K spike for every step and for reagents. Essentially no detectable contamination has been introduced in the step of sample mounting on the filament or evaporation. Anion exchange and extraction steps are relatively free from the contamination except for reagents especially HCl. The contamination level has been about 5 ppbK in HCl. Minimal use of the acid helps to reduce the contamination. One full operation of the chemistry resulted in an addition of about 5 ngK. On the whole,

the greater part of the contamination seemed to be attributed to the meteorite sample.

2.3. Massspectrometry

The size of samples to be analyzed is in the order of 10^{-9} gK, and the amount of cosmogenic ^{40}K in the separated sample is about 10^8 atoms of ^{40}K. This could be determined massspectrometrically sensitively by employing surface ionization techniques. The sample solution was dried on a single tungsten filament which had been cleaned by preheating. In the case of reagent potassium, a large excess of H_3PO_4 was added to the solution. For the sample of meteorites no further treatment seems to be necessary. Probably the presence of some impurities may help to emit K^+ ion smoothly.

The cosmogenic ^{40}K is measured in an enrichment above natural ^{40}K peak, and the absolute amount is determined by dilution analysis employing a spike, enriched ^{39}K.

The AEI MS5 massspectrometer was used with or without a 17 stage electron multiplier, EM (Hamamatsu TV Co.), for ion beams of higher than 10^{-12} amp. The beam current was amplified, using high resisters and vibrating reed electrometers, the Takeda TR 84S ($10^{+9}\,\Omega$) with EM and the CARY 401 ($10^{+10}\,\Omega$) without EM. For a higher beam, the peak height ratios of 41/39 were measured without EM and the system was switched to the EM line and 40/41 ratios were measured in a separate group.

The output of the electrometer was read with a digital voltmeter and the working conditions were controlled with a microprocessor. The system was operated under a controlled switching of the magnetic fields from one peak to another. The readings for the base were performed at a slightly higher mass position than every peak, $+0.2$ mass, by reducing about 20 V from the accelerating potential, 6 kV. The peak matching is controlled by adjusting the magnetic fields, 2100 Gauss, within a maximum range of about ± 1 Gauss in 16 steps.

The tailing of ^{39}K has been determined from time to time, by a scanning covering both sides of the ^{40}K peak. With the reagent, the effect reaches $2\pm 0.3\%$ of the K-40 peak height. In the case of potassium, a normalization, or a correction for mass discrimination and fractionation, could not be applied after the addition of the enriched isotope as a spike. The fractionation gives a serious effect. In later stages of a run, 41/39 ratios increase more than 5%. To reduce this uncertainty, we have to read out the beam continuously from the earliest stage of the run to the very end.

After all, the errors caused by a lower ion beam intensity of ^{40}K and caused by fractionations are predominant in the uncertainty in sample runs. Although the reproducibility of the data seems to be better, it is not easy to perform the measurement with lower than $\pm 1\%$ (σ) for the ^{40}K abundance. For this reason the sample

preparation must be performed better than 1 ppmK in general, which may result in $\pm 5\%$ or higher enrichment to the K-40.

In this work we have realized a very high ionization and collection efficiency for K^+. For example, with $5 \cdot 10^{-9}$ g size sample the beam intensities of 10^{-12}–10^{-11} amp were obtained continuously for more than 30 hrs, and 1600 sets of the ratios could be recorded. This corresponds to that total about 3% of K atoms in the sample were brought to the collector.

3. Results

The results of determinations are compiled in Tables 1 and 2. The reproducibility of the measurement for the enrichment in ^{40}K peak is depending largely on the contamination level and on the contents of cosmogenic ^{40}K, other than the errors due to dilution analysis for the potassium content. When the enrichment exceeds $+10\%$, the determination is performed relatively easily. With the extreme cases of $+50\%$ or higher, the experimental errors might depend on other uncertainties, such as weighing and aliquoting errors, and an overall error seems to be as low as $\pm 3\%$.

^{40}K, ^{36}Ar and ^{38}Ar are the products of >100 MeV particles in Fe+Ni. Recoil effects at the surface of metal grains will result in a loss of the product from the surface. In the case of ^{40}K, our sample treatment seems to be useful to minimize this loss, as we etch and dissolve the surface layers of the grains, using HF and HCl.

Table 1. Peak height ratios of reagent K and ^{39}K spike.

(Ion beam: ^{39}K)	41/39 $\times 10^{-5}$	40/39 $\times 10^{-7}$	40/41 $\times 10^{-6}$	Normalized to 41/39=7216×10^{-5} 40/39 $\times 10^{-7}$
Reagent K				
Method 1 ($\sim 3\times 10^{-12}$ amp)	7100	1274±8	—	1284±8
Method 2 ($\sim 10^{-11}$ amp)	7100	—	1746±10	1250±7
Spike; Enriched ^{39}K	114±2	176±10	—	—
Abundance ratios* (reagent)	7216±4	—	—	1251±4

Method 1; with EM. 41/39 ratio=7000 → 7200 (fractionation).
Method 2; with EM for 40/41 and without EM for 41/39. 41/39=7100 → 7300.
Corrections for ^{39}K tailing were made; Errors attached to peak height ratios are indicating $\pm 1\sigma$ among runs.
* GARNER *et al.* (1975).

4. Discussion

4.1. ^{40}K as a high energy products

According to high energy thick target experiments, the maximum productions

Table 2. Some typical empirical data for determinations of cosmogenic ^{40}K.

Name (Class)			Metal Sample mg	Spike ng^{39}K	Peak height ratios 41/39 $\times10^{-5}$	40/39 $\times10^{-7}$	K ppm	Enrich. ^{40}K+%	Cosmogenic $^{40}K\times10^{10}$ atom/g metal
Yamato	-7301	(H4)	28.0	1.89	6402	1342	0.71	13	15.7±1.9
	-7304	(L6)	10.97	2.4	5921	1171	1.3	14	32±5
			16.3	8.36	6213	1263	3.5	6	42±12
	-74080	(L6)	19.4	5.54	5597	1518	0.98	49	92±5
	-74116	(L)	10.9	5.17	4205	932	0.63	12	14.7±2.9
	-74192	(H6)	18.1	3.92	4626	1640	0.39	87	66±2
	-74418	(H6)	23.9	7.93	4798	1028	0.68	10	13.4±2.7
	-74454	(L6)	26.9	5.63	4510	1131	0.33	21	14.5±3.7
	-74663	(L6)	10.1	4.74	3833	984	0.54	27	28±2.5
	-75097	(L4-5)	24.1	9.4	2792	1044	0.24	80	38.9±1.7
	-75258	(LL6)	6.8	2.57	4882	1137	0.81	20	31±5
Allan Hills	-761	(L6)	23.4	13.0	4620	1230	0.94	40	68±3
	-768	(H6)	17.7	5.99	3516	778	0.33	4	2.7±1.1
	-769	(L6)	29.4	11.7	5331	1155	1.24	12	30±5
	-762	(iron)	24.6	8.3	3835	5782	0.40	700	564±17
	-77272	(L6)	11.9	3.68	4524	955	0.49	11	9.2±0.9
Peace River		(L6)	110.5	9.58	6116	1858	0.48	40	61±2[59±7]
Bruderheim		(L6)	18.4	10.8	5558	1233	2.1	15	54±10
			17.3	26.1	4042	936	2.0	15	55±5[53±4]
Kirin		(H5)	10.0*	4.45	4111	851	0.57	6	6.5±1.6
			17.6**	4.84	4784	1013	0.52	10	10±1
Abee		(E4)	36.8	9.28	6649	1266	3.0	3	16±4
Udei Station		(iron)	45.1	11.6	4050	3154	0.34	300	214±6
Crab Orchard		(Mesos.)	21.4	4.84	4698	1800	0.45	100	88±3
Bencubbin		(Uniq.)	54.4	11.53	5761	1512	0.83	45	73±3
Emery		(Mesos.)	14.4	1.98	6476	2090	1.6	90	240±3
Budulan		(Mesos.)	33.7	5.03	4720	1630	0.29	80	44±2

[ ] IMAMURA *et al.* (1975); * LJ 1 (U.S.A.), ** No. 1A (Tokyo), HONDA *et al.* (1980).

of cosmogenic nuclides are expected at the center of spheres having radii (R) of 140 and 60 g/cm^2 for Ne and ^{40}K respectively. These figures are important factors indicating the cosmic ray flux as a function of meteorite size and sample location in the meteorites. In samples of the internal parts of a large body of $R\geqq150$ g/cm^2, the shielding effects are important for "high energy products" such as ^{40}K, ^{38}Ar, ^{39}Ar, ^{37}Ar and ^{36}Cl in metals. The high energy products are mainly produced by a high energy cosmic ray flux of 10^8–10^9 eV and the production decreases with depth

relatively faster after the maximum in an equilibrium region with low energy secondaries. They are the products (A) of $\Delta A=56-A=16-20$, where 56 is a target mass. On the other hand, the production of Ne isotopes is not affected much at a depth of <250 g/cm^2, but the depth effect is prominent in near-surface samples of a large body, or in objects having smaller radii than that of the maximum production described above, ($R<140$ g/cm^2). The effect causes a steep variation with depth in productions of isotopes of Ne, ^{22}Na, ^{26}Al, ^{53}Mn, ^{54}Mn and other "low energy products" of $\Delta A=2-3$. The low energy products are produced by low energy cosmic ray secondaries, mainly fast neutrons of 10^7-10^8 eV. And the build-up is seen in the increase of the productions with depth near the surface.

The preatmospheric size of chondrites is usually estimated to be smaller than 1 ton, $R=130$ g/cm^2, and the former shielding effect in the equilibrium region is not serious, but the latter depth effect, in the transient, build-up, region of secondaries, must be considerably high. If the ablation loss from the surface was not extensive in recovered samples, and if the irradiation history was simple, the effect is demonstrated in the concentration gradient.

The situation is substantially different in the cases of irons and mesosiderites (stony-iron), as shown in this report. The preatmospheric size of irons and stony-iron meteorites is usually larger than that of chondrites by far. The shielding is a major factor giving a serious effect to high energy products in them. To obtain an exposure age of irons or stony-irons, using ^{40}K data, we have to know about the production rate from the saturation activity of ^{36}Cl or ^{39}Ar. In the case of antarctic meteorites, however, this is almost impossible because of the decay of their radioactivities due to the long terrestrial ages.

4.2. Production rate of ^{40}K

In this work, we determined the production rate of ^{40}K in Fe+Ni directly relative to other rates of ^{36}Ar and ^{36}Cl. We received five metal phase samples separated by Dr. BEGEMANN from 3 mesosiderites, 1 iron and 1 unique metallic meteorites. The natures of the samples and the contents of ^{36}Cl and ^{36}Ar have already been studied by BEGEMANN *et al.* (1976). They are convenient samples in this work because of their intermediate ages, which are still short for ^{40}K. The determinations of ^{40}K in these samples were performed without any difficulty and the data tabulated in Table 3 show relatively small errors.

The contents of ^{40}K and reported data of ^{36}Ar could be compared directly, and the production ratio of ^{36}Ar/^{40}K was found to be 0.78 ± 0.04 (σ) from four specimens. And the ratio ^{40}K/^{36}Cl is obtained by using the ^{36}Ar/^{36}Cl ratio which is well established. Here we apply the same ratio, as obtained in somewhat large objects, to the chondrites without any correction.

A nominal production rate of ^{40}K calculated in Table 3 is actually higher by *ca.* 30% than our old figure. The latter was referred to 20 dpm ^{36}Cl/kg Fe+Ni,

Table 3. Comparison of ^{36}Ar and ^{40}K contents in metals in some irons and mesosiderites and calculation for production rate of ^{40}K.

Name	Class	^{36}Cl# dpm/kg metal	^{36}Ar# $\times 10^{-8}$ cc/g metal	^{40}K $\times 10^{10}$ atom/g metal	$^{36}Ar/^{40}K$ atom/atom
Bencubbin	Uniq.	24.3±1.5	2.08±0.20	73 ± 3	0.766
Budulan	Mesos.	13.7±3.0	1.15±0.07	44 ± 2	(0.702) **
Crab Orchard	Mesos.	17.3±1.5	2.62±0.15	88 ±3	0.800
Emery	Mesos.	24.0±1.0	7.55±0.40	250*±10	0.812
Udei Station	Iron	19.6±0.8	6.0 ±0.30	222*± 6	0.725
				mean:	0.776±39 (σ)

P(^{36}Cl) = 25 atoms/min kg metal (reference value for chondrites).
P(^{36}Ar) = 25/0.83 = 30 〃 (〃).
P(^{40}K) = 30/0.776 = 39 〃 ≡(2.0±0.1)×10^{10} atoms/my g metal.

* Minor corrections were made for the decay of ^{40}K (see Table 2).
** Excluded from a calculation of mean $^{36}Ar/^{40}K$, because of the relatively low ^{36}Cl and ^{53}Mn contents.
\# BEGEMANN *et al.* (1976).

Table 4. ^{40}K exposure ages and others determined in several meteorites.

Meteorite (class)		^{53}Mn* dpm/kg Fe+Ni/3	Exposure Age: ^{40}K Age++ (my)	Exposure Age: Others (my)
Peace River	(L6)	411	37± 2(1.6)	38#
Bruderheim	(L6)	420	24± 5(2.2) / 25± 2	24#
Kirin**	(H5) LJ 1	84	3.2±0.7	
	(H5) No. 1A	187	5.0±0.5	
Abee	(E4)	338	9± 2(1.7)	6.7#
Bencubbin	(Uniq.)	525	38± 2(1.9)	36±5+
Budulan	(Mesos.)	398	40± 9(1.1)	36±8+
Crab Orchard	(〃)	520	64± 6(1.1)	64±7+
Emery	(〃)	422	130±10(1.9)	134±12+
Udei Station	(Iron)	575	139± 6(1.6)	130±10+

* See Table 5.
** See Table 1. ^{40}K ages are indicating the apparent values.
\+ BEGEMANN *et al.* (1976) ^{36}Cl-^{36}Ar age.
++ Production rate, unit: $\times 10^{10}$ atoms/my g, normalized from ^{36}Cl or ^{39}Ar content, BEGEMANN *et al.* (1969, 1976).
\# Data were collected from the compilation prepared by SCHULTZ and KRUSE (1978).

which is a higher value among small iron and stony-iron meteorites. However, if we take 25 dpm/kg Fe+Ni as the average in chondrites, a higher production rate may be more convenient for many chondrites (BEGEMANN and VILCSEK, 1969), though the experimental errors for ^{36}Cl determinations are usually higher, ±10%. Whenever we use the rate, the reference level of the irradiation, P(^{36}Cl)=25 atom/min kg or an equivalent, has to be referred to.

There are several chondrites which have been studied extensively; the ^{39}Ar and/or ^{36}Cl in the metal, ^{21}Ne in bulk and ^{40}K in metal have been examined already.

Table 5. ^{40}K exposure ages and others of Antarctic meteorites.

Meteorite (Class)			^{53}Mn* dpm/kg Fe+Ni/3	Exposure Age	
				^{40}K Age** (my)	Others (my)
Yamato	-7301	(H4)	101	7.9±0.9	13±4#
	-7304	(L6)	412	16±2 21±6	18±2#
	-74080	(L6)	345	46±2	
	-74116	(L)	63	7.6±0.7	
	-74192	(H6)	578	33±2	
	-74418	(H6)	275	7.0±1.3	
	-74454	(L5)	387	7.2±1.6	
	-74663	(L6)	255	14±1	
	-75097	(L4-5)	424	19±1	
	-75258	(LL6)	350	16±2	
Allan Hills	-761	(L6)	443	34±2	29±4#, 33±4####
	-768	(H6)	22	1.3±0.7	1.7±0.2####, 1.4±0.2#
	-769	(L6)	477	15±3 15±2	13±2####
	-762	(iron)	556	410±12(1.5)***	540±125##
	-77272	(L6)	215	4.6±0.5	4±1###

* ^{53}Mn: NISHIIZUMI *et al.* (1978, 1979, 1980); IMAMURA *et al.* (1978, 1979); HONDA *et al.* (1980).

** ^{40}K age: 2.0×10^{10} atoms/my g was assumed.

*** Assumed $\times10^{10}$ atoms/my g. The assumption was made approximately based on the Mn-53 content.

\# TAKAOKA and NAGAO (1978); NAGAO and TAKAOKA (1979), Ne age.
^{21}Ne contents: 5.72±1.78(7301), 8.19±1.01(7304), 13.1±15% (761), 0.605±15% (768), $\times10^{-8}$ cc/g.

\## ^{53}Mn-^{54}Cr (22±5 ppb) age.

\### From ^{53}Mn, ^{26}Al and ^{36}Cl data.

\#### WEBER and SCHULTZ (1980), Ne age.
^{21}Ne contents: 16.7±0.5(761), 0.81±0.02(768), 6.8±0.2(769), $\times10^{-8}$ cc/g.

Here we selected the Bruderheim and Peace River chondrites from the group and repeated our ^{40}K measurements. The data were the same as before. After the normalization with their ^{36}Cl data, respective ^{40}K production rates were obtained, and exposure ages based on their ^{21}Ne data were compared (Table 4). Just for comparison, the ^{40}K–^{36}Cl ages of the mesosiderites and others were recalculated and tabulated with individual meteorite.

The results of determinations with antarctic samples are tubulated in Table 5. Data of ^{40}K may directly be applied to obtain exposure ages of the chondrites. In the table we assumed the constant ^{40}K production rate. In some chondrites, Ne contents are available.

4.3. Comparison with light rare gases

In fact, rare gases, especially Ne isotopes, are quite convenient elements to be

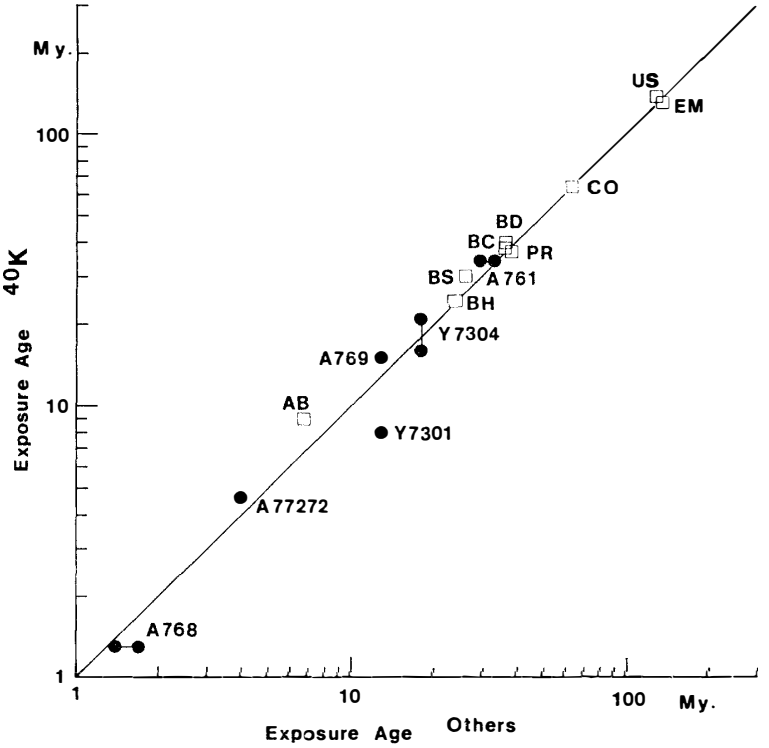

Fig. 2. Comparison of exposure ages based on ^{40}K contents and other data.
US: Udei Station. EM: Emery. CO: Crab Orchard. BD: Budulan. BC: Bencubbin. PR: Peace River. BS: Breitsheid (IMAMURA et al., 1975). BH: Bruderheim. AB: Abee. A: Allan Hills. Y: Yamato.
In this figure we also plotted our previous data of the BREITSHEID. The contents of Ne isotopes are not quite certain. Here we adopted the Ne age based on the highest figure determined with light phase of the meteorite by SIGNER and SUESS (1963).

measured and they provide exposure ages of chondrites after corrections for the cosmic ray flux, by using $^{22}Ne/^{21}Ne$ and $^{3}He/^{21}Ne$. However, we have to be careful about compositions of the meteorite samples and about a diffusion loss of rare gases by heating, shock or weathering, of the meteorites, during the whole history. Although the former is a troublesome problem, the effect is not seriously large, if the sample represents the average composition of chondrites properly. The latter, on the other hand, is a serious problem in some cases. Among antarctic meteorites, the loss of Ar isotopes from metal is common resulting from rusting. Recently even with the observed fall, a considerable amount of the loss of rare gases, including Ne, has been discussed in the Kirin chondrites, H5 (BEGEMANN *et al.*, 1980). Potassium-40 and other nuclides in metal phases seem to be essentially free from these problems.

Both data of ^{40}K and ^{21}Ne in antarctic meteorites, except Yamato-7301, seem to agree with each other (Fig. 2). The data of exposure age calculated from ^{21}Ne are quoted from original papers. Many workers are using the rate close to 0.47×10^{-8} cc/g my for ordinary size L chondrites, which has been given by HERZOG-ANDERS (1971). The number is affected with the composition of the targets and the cosmic ray flux in the sample. For example, for H chondrites, 0.43×10^{-8} cc/g my may be used. Normally $^{22}Ne/^{21}Ne=1.1$ and $^{3}He/^{21}Ne=5.3$ are observed. If the data were not different far from them, the rate could be corrected by a small factor. However, the details are still not quite simple. We examined the data to be compared with our ^{40}K contents. Because the authors are using the above rate or somewhat modified one, which is not much different with individual meteorite, we may not discuss further this problem here.

4.4. Size and depth effect

Among antarctic meteorites, small size chondrites might be predominant, even after a correction for a secondary fragmentation at the fall. The efficiency to recover smaller group, <1 kg, seems much higher than usual. The small individual spherical objects must be interesting samples to us. Because the production of ^{40}K, ^{36}Cl and ^{39}Ar might still be high, but that of ^{21}Ne and ^{53}Mn would be lower. The Yamato-74080, -74418, -74663 and -75258 may be examples showing this effect.

4.5. Multi-exposure history

The meteorites, Yamato-7301 and Allan Hills-768 have been studied by our group on the contents of ^{36}Cl, ^{26}Al, ^{10}Be, ^{53}Mn and rare gases. And their multiple irradiation histories have been discussed. In this work, Yamato-74116 and Kirin meteorites were found to be additional examples of similar complex histories.

In the Yamato-7301, the concentration of ^{40}K seems to be relatively lower than the ^{21}Ne contents by 40%. We interpret the lower ^{40}K content to be attributed to the 1st stage irradiation in a large size object. The absolute intensity level would be

10% or so relative to the normal, as we discussed in earlier papers. The production ratio of $^{40}K/^{21}Ne$ would be lower by a factor 2, compared with ordinary levels, under a heavy shield.

Potassium-40 is literally radioactive in a time scale of 10^9 y. Earlier irradiation effects to the stable nuclides have been discussed with some unusual meteorites. In this problem ^{40}K is a useful nuclide. We may compare the ^{40}K content with stable nuclides. If the latter had been enriched with ancient components, the difference between the two, ^{40}K and the stable nuclides, would give us a crucial information. Therefore, the first stage event could also be explained as a result of the earliest stage irradiation, because the effect does not contribute to the ^{40}K but ^{21}Ne. This seems to be unlikely, however, even if such a possibility could not be completely excluded. If it was the case, the amount of stable nuclides could be the sum of productions in total 3 stages: the earliest, the first stage in a meteoroid and the second stage in a smaller body before the fall.

4.6. Final remarks

Practically sampling of the metal from chondrites may be a problem in this study. In some cases, such as with the Abee chondrite, a clean sampling was not successful by current techniques. But ^{53}Mn, ^{36}Cl, ^{40}K and Ar isotopes could be determined in a common metal sample. These nuclides and Ne will be useful in discussions of the terrestrial age, exposure history, and the size-depth effect of the antarctic meteorites. It is still our ultimate target, however, to find any proof of so-called early irradiation, because such an irradiation seems to be probable, but none of positive data has been available yet.

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