RARE GAS STUDIES OF ANTARCTIC METEORITES

Keisuke NAGAO* and Nobuo TAKAOKA**

Department of Physics, Faculty of Science, Osaka University, Machikaneyama, Toyonaka-shi, Osaka 560

Abstract: Concentrations and isotopic ratios of five stable rare gases were determined in six antarctic meteorites. Yamato-74191 chondrite (L3) contains large amounts of trapped heavy rare gases. Allan Hills No. 8 and Mt. Baldr No. 1 chondrites revealed short ages for cosmic-ray exposure, and are comprised in a cluster around 4 m.y. in a histogram of exposure ages for H-chondrites. This suggests a common parent body to these two chondrites. Yamato-74013 and -74097 diogenites are in close resemblance in amounts and isotopic ratios of cosmogenic He, Ne and Ar, and thus in their cosmic-ray exposure ages. Furthermore, their exposure ages resemble Yamato-692 diogenite but differ from other diogenites known to date. This strongly suggests a parent body common to these Yamato diogenites but not to the other ones. The U/Th–He ages are found to be shorter than the K–Ar ages. This is attributed to diffusive partial loss of He. Such loss was also observed for cosmogenic ^{3}He. The K–Ar ages for Allan Hills No. 8 and No. 1, Mt. Baldr No. 1 and Yamato-74191 in order of antiquity are 4.5±0.2, 4.4±0.2, 4.1±0.2 and 3.4±0.2 b.y., respectively.

^{80}Kr and ^{82}Kr anomalies resulting from epithermal neutron capture by Br have been found in Yamato-74191. Allan Hills No. 1 contains cosmogenic Kr the isotopic composition of which is: ^{78}Kr/^{80}Kr/^{82}Kr/^{83}Kr/^{84}Kr/^{86}Kr$=0.16\pm 0.02/0.59\pm0.06/0.85\pm0.12/=1/0.47\pm0.28/=0$. All chondrites investigated revealed enrichment in ^{129}Xe. Among others excess ^{129}Xe in Yamato-74191 amounts to 3.02×10^{-9} cm^{3}STP/g. An isotopic composition of trapped Xe in Allan Hills No. 8 which is depleted in Xe was identical with that of Xe-rich chondrites such as AVCC-Xe. Relation between trapped heavy rare gases and petrologic type is discussed.

1. Introduction

Meteorites contain rare gases of various origins, acquired by the meteorites through physical processes such as the decay of natural radioactivity, the production by particle bombardment, and trapping of ambient gases. Data on the

Present address:
* Department of Fundamental Science, Okayama University of Science, 1, Ridai-cho 1-chome, Okayama 700.
** Department of Earth Sciences, Faculty of Science, Yamagata University, Yamagata 990.

rare gases contained in the meteorites can provide us useful information on the ages of the meteorites, the histories of cosmic-ray exposures, and the early history of the solar system, which would help distinguish events during various stages in the history of the universe.

In addition, there are problems to be solved which are proper to the antarctic meteorites. As well known, the Yamato meteorites are an assemblage of meteorites first found in a limited area of bare ice (now called the "Meteorite Ice Field") near the Yamato Mountains in East Antarctica. In 1969 and 1973, 21 stony meteorites of various classes were found there (YOSHIDA *et al.*, 1971; SHIRAISHI *et al.*, 1976). In 1974, 663 meteorites were collected in the same bare ice field by the meteorite searching team of the 15th Japanese Antarctic Research Expedition (YANAI, 1976; YANAI, 1978a).

Preliminary studies on chemical composition and petrologic structure of these meteorites have revealed that the collection contains a variety of kinds of meteorites, and that some of them are different in appearence from usual meteorites. They have, for instance, neither fusion crust nor chondrules and resemble terrestrial rocks in appearence. Detection of cosmogenic nuclides in such stones is, therefore, essential to clarify extraterrestrial origin of these stones.

The cause and mechanism for concentration of the Yamato meteorites have never been understood completely, as discussed by some authors (NAGATA, 1978; SHIMA *et al.*, 1978; YANAI, 1978a; CASSIDY *et al.*, 1977). Rare gas data are useful to distinguish individual falls from meteorite showers.

In this paper we report the results of mass-spectrometrical analyses of rare gases in the Yamato meteorites and in the meteorites found in Victoria Land, Antarctica by a Japan-U.S. joint party for meteorite search during the period of December 15, 1976 to January 20, 1977 (YANAI, 1978b; CASSIDY *et al.*, 1977).

2. Sample and Experimental Technique

2.1. *Sample*

Meteorites studied in the present work are listed in Table 1. Detailed description of finding and occurrence of the Yamato meteorites and the Allan Hills and Mt. Baldr meteorites has been given by YANAI (1978a, 1978b) and by CASSIDY *et al.* (1977).

Both Yamato-74013 and -74097 meteorites are classified as diogenites (TAKEDA *et al.*, 1978; YABUKI *et al.*, 1978). It has been reported that they consist of recrystallized orthopyroxene of uniform mineral composition and that a granoblastic texture of equigranular orthopyroxene in these diogenites resembles that observed in Yamato-692 diogenite (TAKEDA *et al.*, 1978; YABUKI *et al.*, 1978). The texture of these diogenites is rather unusual compared with the other diogenites known to date. Another feature of Yamato-74013 and -74097 diogenites is that

Table 1. List of samples.

Sample	Weight (kg) recovered	Classification
Yamato-74013	2.0595[a]	Diogenite[b][c]
Yamato-74097	2.1939[a]	Diogenite[b][c]
Yamato-74191	1.0916[a]	Hypersthene chondrite of type 3[c][d]
Allan Hills No. 1	20.151[f]	Hypersthene chondrite[e]
Allan Hills No. 8	1.150[f]	Bronzite chondrite[e]
Mt. Baldr No. 1	4.108[f]	Bronzite chondrite of type of 5–4[d]

References: a) YANAI (1978a), b) TAKEDA *et al.* (1978), c) YABUKI *et al.* (1978), d) YANAI *et al.* (1978), e) Classification based on iron content given by NISHIIZUMI *et al.* (1978b), f) YANAI (1978b).

they have no fusion crust on the smooth surface and seem to be two of fragments from a single fall (YANAI, 1978a; TAKEDA *et al.*, 1978).

Yamato-74191 meteorite is classified as unequilibrated hypersthene chondrite (L3) (YANAI *et al.*, 1978; YABUKI *et al.*, 1978). Petrological data for Allan Hills No. 1 and No. 8 meteorites were not available. Mt. Baldr No. 1 meteorite is classified as the chemical and petrologic type H4 or 5 by YANAI *et al.* (1978).

Meteorite samples we used are rather fresh specimens of meteorite chips except Mt. Baldr No. 1. After fusion crust was removed, the meteorites were crushed in a stainless steel mortar and then sifted to separate fine powder by passing through a 200-mesh screen. Grain samples of the size between 32- and 200-mesh were employed for measurements of the rare gas composition and the potassium content. For Mt. Baldr No. 1, the sample we had was a chip of meteorite including fusion crust and a zone revealing wethering. The sample used for the rare gas analysis was prepared from a portion which did not exhibit signs of wethering. Sample sizes used were from 0.09 to 0.31 g for a single analysis of rare gases, and about 0.15 g for the potassium content. Duplicated analyses were carried out for the isotopic and elemental compositions of rare gases except for Yamato-74191 and Mt. Baldr No. 1. The potassium content was determined from a single analysis except for the Allan Hills No. 8 meteorite. For Allan Hills No. 8, an agreement between the duplicated analyses of the potassium content was satisfactory (*i.e.*, 650 and 670 ppm). The potassium analyses were carried out with an atomic absorption technique by Mr. H. MINARI in the laboratory of chemical analysis, Central Workshop of Osaka University.

2.2. Mass spectrometry

Details of the instrument and experimental technique used for the rare gas analysis have been published elsewhere (TAKAOKA, 1976; TAKAOKA and NAGAO, 1978). Experimental procedures changed from those described previously are as follows: extraction temperature was 1700°C in the first run and was increased to 1800°C in the duplicated run. In both runs, extraction yields were tested by reheating the samples. The rare gases purified were separated into four fractions of He–Ne, Ar, Kr and Xe. The He–Ne fraction was separated with activated charcoal at liquid nitrogen temperature. Ar, Kr and Xe fractions were separated from each other with an activated charcoal trap kept at −50, −30 and 100°C, respectively. An atmospheric standard which was prepared for calibration of the instrumental sensitivity was too low in abundance of ^{3}He to determine the isotopic ratio of He with sufficient precision. To correct a mass discrimination effect on the ^{3}He/^{4}He ratio, an artificial ^{3}He–^{4}He mixture was prepared from metered amounts of pure ^{3}He and ^{4}He. The ^{3}He/^{4}He ratio of the mixture was 1.34×10^{-4}. The concentrations and isotopic ratios of rare gases in the Berkeley standard, Bruderheim (REYNOLDS, J. H., 1973, written communication) were measured. The results are given in Tables 2, 3 and 4. They are in good agreement with mean values from different laboratories all over the world.

3. Results and Discussion

The concentrations and isotopic ratios of He, Ne and Ar in six antarctic meteorites are shown in Table 2. The concentrations and isotopic ratios of Kr and Xe are given in Table 3 and 4, respectively. Data on Kr and Xe in Yamato-74013 and -74097 diogenites are not given. The isotopic ratios of Kr and Xe in these diogenites were atmospheric.

He, Ne and Ar in Yamato-74013 and -74097 diogenites are mostly cosmogenic. These diogenites are similar in both concentrations and isotopic ratios of He, Ne and Ar. This suggests a possibility that these may be two pieces from a single fall, as noted by YANAI (1978a). Yamato-74191 chondrite contains a small amount of trapped Ne and large amounts of trapped Ar, Kr and Xe. Enrichment in the trapped heavy rare gases is common to unequilibrated chondrites. Allan Hills No. 1 and No. 8, and Mt. Baldr No. 1 chondrites contain larger amounts of radiogenic ^{4}He compared with three Yamato meteorites investigated. He, Ne and Ar in Allan Hills No. 1 chondrite are the mixture of cosmogenic and radiogenic gases. Allan Hills No. 8 (a) and Mt. Baldr No. 1 have trapped Ne. Because of differences in the isotopic ratio of Ne and in the concentrations of Kr and Xe between the duplicated analyses, data of the separate runs for Allan Hills No. 8 are given in Tables 2, 3 and 4. These differences are beyond experimental errors and are attributed to inhomogeneous sample composition.

3.1. Cosmic-ray exposure age

A cosmic-ray exposure age T_i is given by

$$T_i = N_i / P_i \ ,$$

Table 2. Concentrations and isotopic ratios of He, Ne and Ar.

Sample	^{4}He*	$^{3}He/^{4}He$	^{22}Ne*	$^{20}Ne/^{22}Ne$	$^{21}Ne/^{22}Ne$	^{36}Ar*	$^{38}Ar/^{36}Ar$	$^{40}Ar/^{36}Ar$
Yamato-74013	531	0.130±0.002	18.4	0.846±0.003	0.916±0.003	0.900	1.47±0.04	35.7±5.3
Yamato-74097	459	0.151±0.004	19.5	0.849±0.005	0.919±0.005	0.834	1.42±0.01	31.9±1.8
Yamato-74191	545	0.0194±0.0002	3.58	0.969±0.006	0.843±0.007	34.1	0.196±0.002	102±1
Allan Hills No. 1	1080	0.0528±0.0003	14.3	0.842±0.005	0.913±0.004	1.44	1.16±0.03	3400±40
Allan Hills No. 8 (a)	1100	0.00217±0.00003	0.657	1.226±0.025	0.874±0.007	0.524	0.287±0.002	>6000
Allan Hills No. 8 (b)	1180	0.00208±0.00002	0.668	0.865±0.007	0.927±0.005	0.654	0.311±0.003	7710±70
Mt. Baldr No. 1	1600	0.00484±0.00010	2.40	1.610±0.015	0.885±0.011	0.931	0.462±0.003	5200±90
Bruderheim**	514	0.102±0.002	9.77	0.840±0.001	0.912±0.004	1.53	1.07±0.006	898±20

* Unit in 10^{-8} cm^{3} STP/g. Uncertainties in concentrations are 6, 12 and 10% for ^{4}He, ^{22}Ne and ^{36}Ar, respectively.

** Standard sample prepared at Berkeley.

Table 3. Concentration and isotopic ratios of Kr.

Sample	^{84}Kr*	^{78}Kr	^{80}Kr	^{82}Kr	^{83}Kr	^{84}Kr	^{87}Kr
Yamato-74191	22.6	0.650±0.022	10.3±0.2	22.7±0.1	20.5±0.3	=100	31.1±0.5
Allan Hills No. 1	0.758	1.79±0.09	8.16±0.28	25.7±0.5	27.0±0.6	=100	29.9±0.5
Allan Hills No. 8 (a)	0.38	—	4.45±0.13	20.6±0.3	20.3±0.5	=100	30.9±0.4
Allan Hills No. 8 (b)	0.890	0.652±0.036	4.45±0.15	20.3±0.5	19.9±0.4	=100	31.1±0.4
Mt. Baldr No. 1	0.478	0.684±0.024	4.82±0.50	20.6±0.4	20.6±0.3	=100	30.6±0.7
Bruderheim	0.71	1.51±0.04	6.64±0.13	23.0±0.3	24.6±0.4	=100	30.1±0.7
AVCC-Kr**	—	0.610	3.919	20.15	20.26	=100	30.98

* Concentration in 10^{-10} cm^{3} STP/g. Error in concentration is 10%.

** EUGSTER *et al.* (1967).

Table 4. Concentration and isotopic ratios of Xe.

Sample	^{132}Xe*	^{124}Xe	^{126}Xe	^{128}Xe	^{129}Xe	^{130}Xe	^{131}Xe	^{132}Xe	^{134}Xe	^{136}Xe
Yamato-74191	22.2	0.463 ± 0.007	0.432 ± 0.005	8.53 ± 0.05	234 ± 1	16.2 ± 0.1	81.9 ± 0.5	=100	38.2 ± 0.2	32.0 ± 0.3
Allan Hills No. 1	0.668	0.721 ± 0.056	0.807 ± 0.061	8.59 ± 0.22	182 ± 3	16.0 ± 0.3	82.9 ± 0.6	=100	38.4 ± 0.5	31.9 ± 0.4
Allan Hills No. 8 (a)	1.39	0.473 ± 0.044	0.418 ± 0.022	7.91 ± 0.14	134 ± 2	16.1 ± 0.2	81.0 ± 0.8	=100	38.4 ± 0.7	32.4 ± 0.7
Allan Hills No. 8 (b)	0.786	0.460 ± 0.035	0.426 ± 0.033	8.05 ± 0.17	135 ± 2	16.0 ± 0.3	81.6 ± 0.8	=100	38.6 ± 0.4	32.5 ± 0.3
Mt. Baldr No. 1	0.557	0.524 ± 0.064	0.481 ± 0.045	8.33 ± 0.13	135 ± 2	16.4 ± 0.5	81.9 ± 1.4	=100	38.1 ± 1.1	33.4 ± 0.9
Bruderheim	1.01	0.57 ± 0.02	0.63 ± 0.01	8.25 ± 0.04	129 ± 2	16.3 ± 0.2	82.0 ± 0.3	=100	38.8 ± 0.1	32.6 ± 0.1
AVCC-Xe**	—	0.459	0.410	8.20	—	16.08	81.7	=100	38.2	32.1

* Concentration in 10^{-10} cm^3 STP/g. Uncertainty in concentration is 8%.
** EUGSTER *et al.* (1967).

Table 5. Cosmogenic ^3He, ^{21}Ne and ^{38}Ar and cosmic-ray exposure ages.

Sample	^3He (10^{-8} cm^{3}STP/g)	^{21}Ne	^{38}Ar	$^3He/^{21}Ne$	$^{21}Ne/^{38}Ar$	P_3 (10^{-8} cm^{3}STP/g/m.y.)	P_{21}	P_{38}	P_3/P_{21}	P_{21}/P_{38}	Cosmic-ray exposure age (m.y.) T_3	T_{21}	T_{38}
Yamato-74013	69.2	16.8	1.32	4.12	12.7	2.65	0.517	0.0344	5.03	15.0	26	32	38
Yamato-74097	69.5	17.9	1.17	3.88	15.3	2.65	0.517	0.0344	5.03	15.0	26	35	34
Yamato-74191	10.6	3.01	0.350	3.52	8.60	2.48	0.466	0.0560	5.32	8.32	4.3	6.5	6.3
Allan Hills No. 1	57.0	13.1	1.60	4.35	8.19	2.48	0.466	0.0560	5.32	8.32	23	28	29
Allan Hills No. 8 (average)	2.44	0.605	0.082	4.03	7.38	2.48	0.433	0.0614	5.73	7.05	0.98	1.4	1.3
Mt. Baldr No. 1	7.75	2.12	0.292	3.66	7.26	2.48	0.433	0.0614	5.73	7.05	3.1	4.9	4.8

Uncertainties in exposure ages are estimated to be 12, 15 and 17% for T_3, T_{21} and T_{38}, respectively. The uncertainties were calculated from those of rare gas determination and production rates.

where N_i and P_i are concentration and production rate of cosmogenic nuclide i, respectively. Table 5 shows the concentrations of cosmogenic ^{3}He, ^{21}Ne and ^{38}Ar, and the cosmic-ray exposure ages for six antarctic meteorites. All of ^{3}He determined was regarded to be cosmogenic. Cosmogenic ^{21}Ne and ^{38}Ar were calculated by correcting for the trapped component. The following isotopic ratios were assumed: $(^{21}Ne/^{22}Ne)_t=0.0290$ and $(^{38}Ar/^{36}Ar)_t=0.187$, and $(^{21}Ne/^{22}Ne)_c=0.92$ and $(^{38}Ar/^{36}Ar)_c=1.55$ for trapped and cosmogenic Ne and Ar, respectively.

The production rates of ^{3}He (P_3) and ^{21}Ne (P_{21}) were calculated according to HERZOG and ANDERS (1971). The ^{21}Ne production rate used is in good agreement with that given by BAGARD and CRESSY (1973). The production rate of cosmogenic ^{38}Ar was calculated using an empirical production ratio of cosmogenic ^{21}Ne to ^{38}Ar given by STAUFFER (1962) and the absolute production rate of ^{21}Ne given by HERZOG and ANDERS (1971). Since there were no available data on chemical compositions of bulk meteorites used in this work except for Yamato-74013 diogenite (YABUKI, 1978), the average chemical compositions (MASON, 1971) were employed for the calculation of P_{21}/P_{38} ratio. The production rates of cosmogenic ^{3}He, ^{21}Ne and ^{38}Ar are listed in Table 5. The ratios of production rates, P_3/P_{21} and P_{21}/P_{38}, are compared with the ^{3}He/^{21}Ne and ^{21}Ne/^{38}Ar ratios for cosmogenic gases determined. As shown in Table 5, the ^{3}He/^{21}Ne ratio is always lower than the P_3/P_{21} ratio, which suggests diffusive partial loss of He. An agreement between ^{21}Ne/^{38}Ar and P_{21}/P_{38} is excellent. For this reason the exposure ages calculated on cosmogenic ^{21}Ne and ^{38}Ar are more reliable than those on cosmogenic ^{3}He.

In Fig. 1, the cosmic-ray exposure ages determined from cosmogenic ^{21}Ne are compared with those of other meteorites (MEGRUE, 1968; SHIMA *et al.*, 1973; WASSON, 1974). The exposure ages for Yamato-7301, -7304 and -7305 meteorites given by TAKAOKA and NAGAO (1978) are also plotted. Bronzite chondrites, Allan Hills No. 8 and Mt. Baldr No. 1, belong to a sharp cluster of 4 m.y. These meteorites seem to have been stored in a single parent body which was disrupted about 4 m.y. ago. However, the contents of cosmogenic ^{53}Mn (NISHIIZUMI *et al.*, 1978b) and ^{26}Al (FRUCHTER and EVANS, 1977) in Allan Hills No. 8 chondrite have been found significantly lower than those observed in stone meteorites of long exposure age. A similar low content of ^{53}Mn has been reported in Yamato-7301 chondrite, and possible mechanisms responsible for the low activity of ^{53}Mn were discussed by NISHIIZUMI *et al.* (1978a).

According to them, the most plausible mechanism is a two-stage irradiation in which the meteorite has been subjected to a long period (*ca.* 10^2 m.y.) of heavy shielding in a deep place of large meteoroid before it was bombarded by cosmic-rays for a short time (*ca.* 1 m.y.) in a small-size meteoroid. The low ^{53}Mn and ^{26}Al contents in the Allan Hills No. 8 meteorite can be also attributed to the two-stage irradiation (NISHIIZUMI *et al.*, 1978b; IMAMURA *et al.*, 1978). In this context,

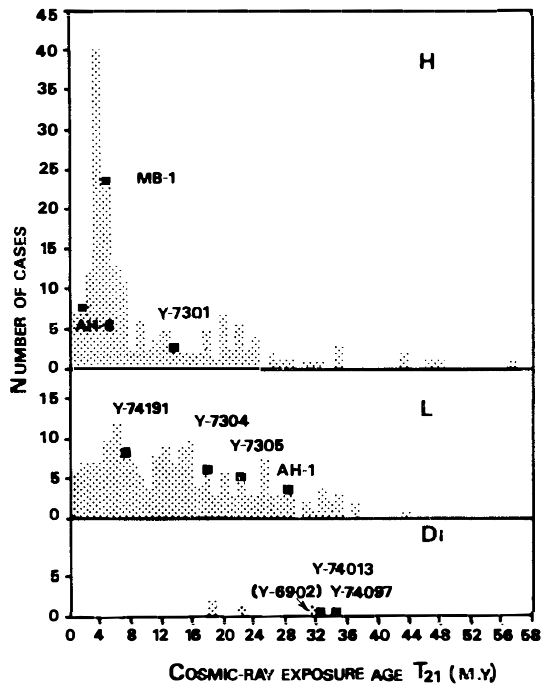

Fig. 1. Comparison of cosmic-ray exposure ages for the antarctic meteorites with those for other meteorites. Data for H- and L-chondrites were reproduced from WASSON *(1974) and data for diogenites from* MEGRUE *(1968) and* SHIMA *et al. (1973). Allan Hills No. 8 and Mt. Baldr No. 1 are comprised in a sharp cluster around 4 m.y. Yamato-74013 and -74097 diogenites as well as Yamato-692 diogenite make a cluster at about 34 m.y., whereas the ages for other diogenites known to date gather around 20 m.y.*

the cosmic-ray irradiation history of Allan Hills No. 8 is probably the second stage irradiation for a few tenth million years following a long period of irradiation with heavy shielding. It is noted that the irradiation time of 0 to 1.4 m.y. has been proposed for the second stage of irradiation of Yamato-7301 chondrite (NISHIIZUMI *et al.*, 1978a).

3.2. *U/Th–He and K–Ar ages*

U/Th–He and K–Ar ages are given in Table 6, together with the concentrations of radiogenic ^{4}He and ^{40}Ar, and the content of potassium. Radiogenic ^{4}He was calculated by the following equation,

$$^4He_r = {}^4He_m - 5\cdot{}^3He_c - 330\cdot{}^{20}Ne_t,$$

where subscripts r, m, c and t represent radiogenic, measured, cosmogenic and trapped components, respectively. The following isotopic ratios are assumed: $(^3He/^4He)_c=0.2$ and $(^4He/^{20}Ne)_t=330$. The $^4He/^{20}Ne$ ratio for the trapped component is a mean value of Pesyanoe (ZÄHRINGER, 1962) and Orgueil (MAZOR *et al.*, 1970). ^{40}Ar determined is mostly radiogenic, and no correction was made for the

Table 6. Radiogenic ^{4}He and ^{40}Ar and U/Th–He and K–Ar ages.

Sample	$^{4}He^{*}$	$^{40}Ar^{*}$	K (ppm)	Gas retention ages (b.y.)	
				U/Th–He	K–Ar
Yamato-74191	320±54	3490±310	910	0.9±0.3	3.4±0.2
Allan Hills No. 1	795±80	5270±280	730	2.0±0.6	4.4±0.2
Allan Hills No. 8	1170±120	5040±430	660	3.3±0.3	4.5±0.2
Mt. Baldr No. 1	880±180	4840±260	790	2.7±0.5	4.1±0.2

* Concentrations in 10^{-8} cm^{3} STP/g.
Decay constants used in calculation of ages are based on those recommended by the IUGS Subcommission on Geochronology (STEIGER and JÄGER, 1977).

cosmogenic and trapped components.

Since we had no avairable data on the content of uranium in the meteorites studied in this work, mean contents of uranium for L-chondrite (U=15±5 ppb) and for H-chondrite (U=12±1 ppb) in literature (MORGAN, 1971) were used with an assumption of Th/U=3.6. As shown in Table 6, the U/Th–He ages are systematically younger than the K–Ar ages. This can be attributed to partial loss of He. Such diffusive loss of He has been observed in cosmogenic ^{3}He, as discussed in Section 3. 1. Compared the present results for the antarctic meteorites with those for other meteorites in literature (*e.g.*, WASSON, 1974), both U/Th–He and K–Ar ages for the antarctic chondrites fall in the typical distribution of ages for the relevant chemical group of chondrites.

3.3. Isotopic anomaly in Kr

The isotopic composition of Kr in Yamato-74191 chondrite shows large excesses in ^{80}Kr and ^{82}Kr compared with average carbonaceous chondrite (AVCC) Kr (EUGSTER *et al.*, 1967). The amount of excessive ^{80}Kr is $(1.44\pm0.05)\times10^{-10}$ cm^{3}STP/g. A small excess in ^{78}Kr is presumably of cosmogenic origin. Contribution from cosmogenic ^{80}Kr is estimated to be 4×10^{-12} cm^{3}STP/g on the basis of the small excess in ^{78}Kr and an isotopic yield of cosmogenic (Allan Hills No. 1) Kr as will be discussed later. The origin of excesses in ^{80}Kr and ^{82}Kr can be attributed to neutron capture by Br. However, the thermal neutron capture by Br could not interpret the observed $^{80}Kr/^{82}Kr$ ratio for excess Kr. The $^{80}Kr/^{82}Kr$ ratio resulting from the thermal neutron capture by Br is 3.6, while the observed one is 2.7. MARTI *et al.* (1966) have shown that the epithermal neutron capture by Br produces ^{80}Kr and ^{82}Kr, the isotopic ratio of which is around 2.5. According to MARTI *et al.* (1966), the amounts of neutron capture products were estimated for the mean contents of halogen atoms in L-chondrites (GOLES and ANDERS, 1962; GUNTEN *et al.*, 1965), normalizing the concentration of excess ^{80}Kr to 1.40×10^{-10} cm^{3}STP/g. Table 7 shows estimates of ^{80}Kr, ^{82}Kr, ^{36}Ar and ^{128}Xe produced by the

Table 7. Comparison between excess isotopic abundances observed in Yamato-74191 chondrite (L3) and calculated (n, γβ) products.

Target	Product	Observed excess (10^{-12} cm^3 STP/g)	Neutron energy		
			Thermal –10 (eV)	30–300 (eV)	10–100 (keV)
^{79}Br	^{80}Kr	140±8	=140	=140	=140
^{81}Br	^{82}Kr	52±11	39.2	52.2	84.1
^{35}Cl	^{36}Ar	<3.4×10^5	5.1×10^5	710	4000
^{127}I	^{128}Xe	7.8±2.1	7.3	18	23

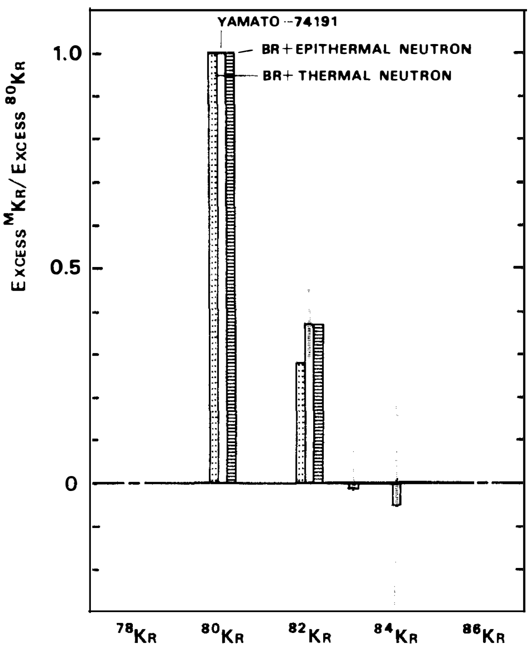

Fig. 2. ^{80}Kr and ^{82}Kr anomalies in Yamato-74191 compared with isotopic abundance of Kr originating from neutron capture by Br. The ^{80}Kr and ^{82}Kr anomalies have been corrected for the trapped (AVCC) and cosmogenic (Allan Hills No. 1) Kr. Errors cited are one σ and include uncertainties both in the isotopic ratio measurement and in the correction for cosmogenic Kr. Relative abundances of ^{80}Kr and ^{82}Kr isotopes resulting from neutron captures by Br in energy range of thermal—10 eV (referred to thermal neutron in the figure) and 30–300 eV (referred to epithermal neutron in the figure) are given. An agreement is excellent between the observed spectrum and the calculated one in the epithermal range of neutron energy.

neutron capture in three ranges of neutron energy. Comparison with the observed isotopic ratio of ^{80}Kr to ^{82}Kr excess gives the appropriate neutron energy of 30 to 300 eV. Fig. 2 represents a comparison of the isotopic ratio for the excess Kr observed with the calculated one for thermal neutrons and for epithermal neutrons of energy between 30 and 300 eV. An agreement is satisfactory between the observed and the calculated spectra for Kr produced by the epithermal neutron capture.

The isotopic ratios of Kr in Allan Hills No. 1 chondrite are enriched in the light isotopes compared with AVCC Kr. The concentration of excess ^{83}Kr is 5.7×10^{-12} cm^{3}STP/g, and the isotopic composition of excessive Kr is: ^{78}Kr: ^{80}Kr: ^{82}Kr : ^{83}Kr : ^{84}Kr : ^{86}Kr$=0.16\pm0.02$: 0.59 ± 0.06: 0.85 ± 0.12: $=1.00$: 0.47 ± 0.28: $=0$. In calculation of excess Kr, a spallogenic yield in ^{86}Kr was assumed to be zero. As shown in Fig. 3, the isotopic composition of excessive Kr agrees with a

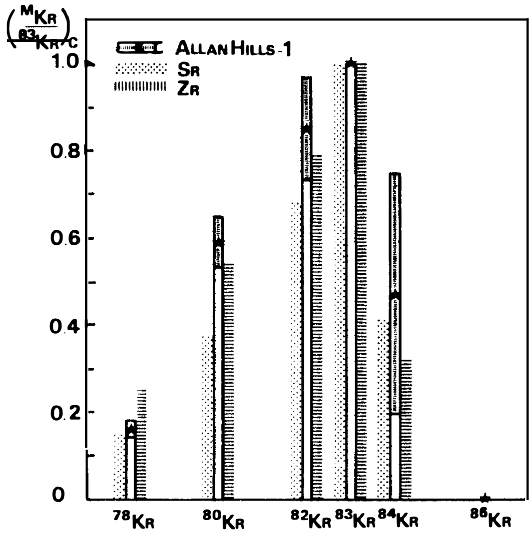

Fig. 3. Mass spectrum of cosmogenic Kr in Allan Hills No. 1 normalized to $^{83}Kr=1$. The amount of $^{83}Kr_c=5.7\times 10^{-12}$ cm^{3}STP/g. Theoretical isotopic compositions of Kr produced by spallation reactions on Sr and Zr are shown for comparison. In calculation of theoretical spectra of spallogenic Kr, an isobaric production rate inversely proportional to $(\Delta A)^n$ is assumed, where ΔA and n are a mass difference between target and product, and a parameter depending on hardness of irradiation, respectively. According to MARTI et al. (1966), the following numerical values were used: $n=2.0$; $g(^{78}Kr)=0.65$, $g(^{80,82\text{ and }83}Kr)=1$, $g(^{84}Kr)=0.25$ and $g(^{86}Kr)=0$ for isobaric fraction coefficients. ^{78}Kr, ^{83}Kr and ^{84}Kr can be understood in terms of mixture of theoretical spectra produced from Sr and Zr. ^{80}Kr and ^{82}Kr exceed theoretical values. This discrepancy is presumably due to contribution of neutron-capture products of Br.

theoretical spectrum of spallogenic Kr calculated from the production rate of an isobar which depends on the mass difference between target and product nuclides (MARTI *et al.*, 1966). Excess Kr except ^{80}Kr and ^{82}Kr is understood in terms of mixture of nuclides produced by cosmic-ray irradiation of Zr and Sr, whereas excess ^{80}Kr and ^{82}Kr seem to exceed the theoretical value. These discrepancies may be due to a small contribution of neutron capture products as discussed earlier.

3.4. Isotopic anomaly in Xe

Fig. 4 is isotopic amonalies of Xe which are represented by $\delta_{132}(\text{atm})$. The definition of $\delta_{132}(\text{atm})$ is given by

$$\delta_{132}(\text{atm})=\left[\frac{(^{m}\text{Xe}/^{132}\text{Xe})}{(^{m}\text{Xe}/^{132}\text{Xe})_{\text{atm}}}-1\right]\times 10^{3},$$

where subscript atm means atmosphere. AVCC-Xe (EUGSTER *et al.*, 1967) is also shown for comparison. Excess ^{129}Xe was observed in all chondrites studied. Among others the concentration of excess ^{129}Xe in Yamato-74191 unequilibrated L-chondrite is as high as 3.02×10^{-9} cm^{3}STP/g.

As indicated in Fig. 4, the isotopic composition of Xe in Yamato-74191 is identical with that of AVCC-Xe with the exception of small excesses in ^{126}Xe and ^{128}Xe. The isotopic composition of trapped Xe in Xe-rich ordinary chondrites has been found to agree with that known for carbonaceous chondrites (EUGSTER *et al.*, 1969). As discussed in Section 3.3, this chondrite contains appreciable amounts of

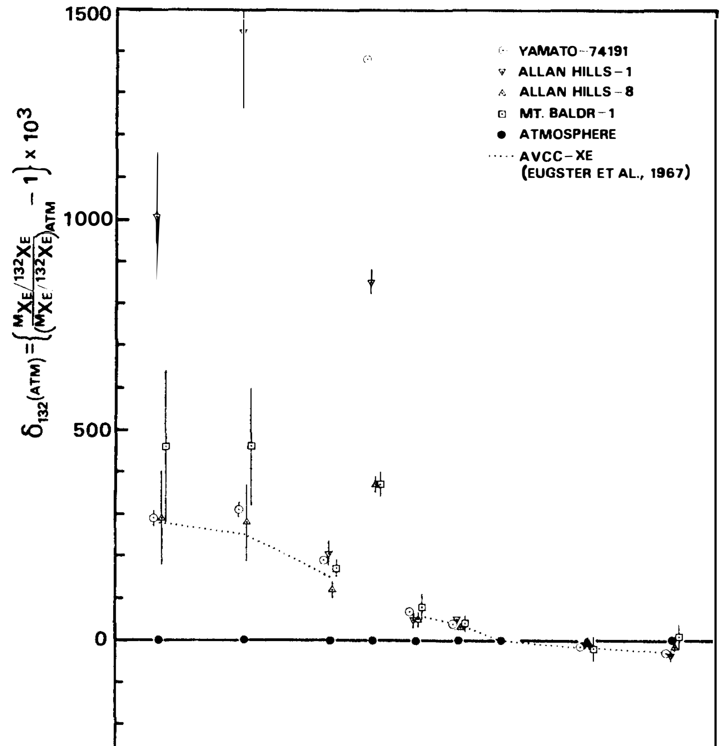

Fig. 4. Isotopic anomalies of Xe. Excess ^{129}Xe is seen in all chondrites. Amount of excess ^{129}Xe in Yamato-74191 is as high as 3.02×10^{-9} cm^{3}STP/g. Xe in Allan Hills No. 1 and Mt. Baldr No. 1 can be understood as a mixture of AVCC-Xe and cosmogenic Xe. Isotopic composition of Xe in Allan Hills No. 8 is identical with that of AVCC-Xe.

^{80}Kr and ^{82}Kr produced by neutron capture of Br. Excess ^{128}Xe is understood in terms of the neutron capture by ^{127}I. However, excess ^{126}Xe (5.0×10^{-13} cm^{3}STP/g) is left to clarify the origin.

The isotopic composition of Xe in Allan Hills No. 8 is identical with that of AVCC-Xe within experimental errors. This bronzite chondrite has a significantly short age of cosmic-ray irradiation, and thus contains little amount of cosmogenic Xe. Xenon determined in Allan Hills No. 8 consists mostly of trapped Xe. A petrologic type has not been reported for Allan Hills No. 8. As will be discussed later, we guess the petrologic type of this chondrite with a correlation between the petrologic type of chondrite and the amounts of trapped ^{36}Ar, ^{84}Kr and ^{132}Xe given by MARTI (1967). Data for Allan Hills No. 8 fall in the petrologic type 6 in the correlation plot (Fig. 5). If this is the case, the isotopic composition of trapped Xe in the highly metamorphozed chondrite is found to be identical with that in the undifferentiated primitive meteorites.

Allan Hills No. 1 chondrite contains an appreciable amount of cosmogenic Xe ($^{126}Xe_c = 2.7 \times 10^{-13}$ cm^{3}STP/g). After correction of AVCC-Xe for the trapped component on the assumption that $^{136}Xe_c = 0$, the isotopic composition of cosmogenic Xe in Allan Hills No. 1 is: $^{124}Xe : {}^{126}Xe : {}^{128}Xe : {}^{131}Xe = 0.66 \pm 0.18 : = 1 : 1.1 \pm 0.6 : 4 \pm 3$. Xenon in Mt. Baldr No. 1 is also understood as a mixture of AVCC-Xe and a small amount of cosmogenic Xe.

3.5. Trapped ^{36}Ar, ^{84}Kr and ^{132}Xe and petrologic classification

ZÄHRINGER (1966) and MARTI (1967) have noted that the concentrations of trapped Ar, Kr and Xe are correlated with the petrologic type of chondrite. MARTI (1967) have shown definite linear correlations between trapped ^{36}Ar, ^{84}Kr and ^{132}Xe and given the concentration of trapped ^{132}Xe in different petrologic type as follows:

Type 6 : $^{132}Xe_t < 1.4$,
Type 5 : $1.4 \leqq {}^{132}Xe_t < 3.5$
Type 4 : $3.5 \leqq {}^{132}Xe_t < 10$
Type 3 : $10 \leqq {}^{132}Xe_t < 40$, in unit of 10^{-10} cm^{3}STP/g.

Trapped ^{36}Ar, ^{84}Kr and ^{132}Xe in the antarctic chondrites investigated in this work are plotted in Fig. 5. For comparison, data for other meteorites used in MARTI (1967) are also plotted. Yamato-74191 chondrite is plotted in the cluster formed by the chondrites of petrologic type 3. This classification deduced from trapped ^{132}Xe is consistent with that given from petrologic study by YANAI *et al.* (1978) and YABUKI *et al.* (1978). Allan Hills No. 1 and No. 8, and Mt. Baldr No. 1 chondrites are plotted in the left lower side of the correlation diagram in Fig. 5. This means that the petrologic type of these meteorites deduced from the concentration of trapped ^{132}Xe is 6. However, Mt. Baldr No. 1 has been assigned to the petrologic type 5 or 4 by YANAI *et al.* (1978). No petrologic data were

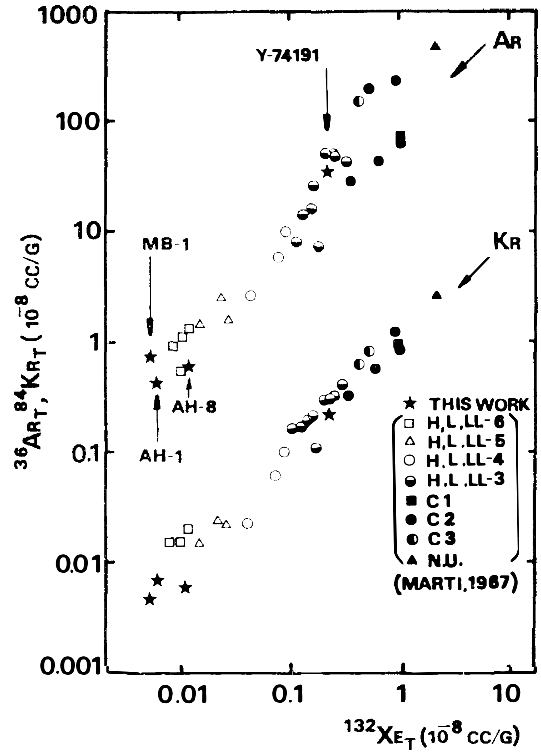

Fig. 5. Correlation diagram of trapped ^{36}Ar, ^{84}Kr and ^{132}Xe.

available for Allan Hills No. 1 and No. 8.

The antarctic meteorites are plausible candidates to study the relation between rare gas content and metamorphism. Consortial investigation of rare gas content, petrologic texture and other related subjects will be able to make great progress in studies on origin, diffusion, and fractionation of rare gas in terms of thermal history of meteorite.

Acknowledgments

The authors are indebted to Dr. K. YANAI, National Institute of Polar Research, for the meteorite samples used in this work. They are thankful to Mr. H. MINARI, Osaka University, for the atomic absorption analyses of potassium contents. This work is supported by the Ministry of Education (grant No. 234024).

References

BOGARD, D. D. and CRESSY, P. J., Jr. (1973): Spallation production of ^{3}He, ^{21}Ne and ^{38}Ar from target elements in the Bruderheim chondrite. Geochim. Cosmochim. Acta, **37**, 547–557.

CASSIDY, W. A., OLSEN, E. and YANAI, K. (1977): Antarctica: A deep-freeze storehouse for meteorites. Science, **198**, 727–731.

EUGSTER, O., EBERHARDT, P. and GEISS, J. (1967): Krypton and xenon isotopic composition in three carbonaceous chondrites. Earth Planet. Sci. Lett., **3**, 249–257.

EUGSTER, O., EBERHARDT, P. and GEISS, J. (1969): Isotopic analyses of krypton and xenon in fourteen stone meteorites. J. Geophys. Res., **74**, 3874–3896.

FRUCHTER, J. S. and EVANS, J. C. (1977): Personal communication quoted in NISHIIZUMI *et al.* (1978b).

GOLES, G. G. and ANDERS, E. (1962): Abundances of iodine, tellurium and uranium in meteorites. Geochim. Cosmochim. Acta, **26**, 723–737.

GUNTEN, H. R. VON, WYTTENBACH, A. and SCHERLE, W. (1965): Determination of chlorine in stony meteorites by neutron activation analysis. Geochim. Cosmochim. Acta, **29**, 475–480.

HERZOG, G. E. and ANDERS, E. (1971): Absolute scale for radiation ages of stony meteorites. Geochim. Cosmochim. Acta, **35**, 605–611.

IMAMURA, M., NISHIIZUMI, K., TAKAOKA, N., NAGAO, K. and HONDA, M. (1978): Cosmic-ray produced nuclides in antarctic meteorites. Abstract presented in Ann. Meeting Meteorol. Soc., 1978.

MARTI, K., EBERHARDT, P. and GEISS, J. (1966): Spallation, fission and neutron capture anomalies in meteoritic krypton and xenon. Z. Naturforsch., **21a**, 398–413.

MARTI, K. (1967): Trapped xenon and the classification of chondrites. Earth Planet. Sci. Lett., **2**, 193–196.

MASON, B. (1971): Handbook of Elemental Abundances in Meteorities, ed. by B. MASON. New York, Gordon and Breach, 555 p.

MAZOR, E., HEYMANN, D. and ANDERS, E. (1970): Noble gases in carbonaceous chondrites. Geochim. Cosmochim. Acta, **34**, 781–824.

MEGRUE, G. H. (1968): Rare gas chronology of hypersthene achondrites and pallasites. J. Geophys. Res., **73**, 2027–2033.

MORGAN, J. W. (1971): Uranium. Handbook of Elemental Abundances in Meteorites, ed. by B. MASON. New York, Gordon and Breach, 529–548.

NAGATA, T. (1978): A possible mechanism of concentration of meteorites within the Meteorite Ice Field in Antarctica. Mem. Natl Inst. Polar Res., Spec. Issue, **8**, 70–92.

NISHIIZUMI, K., IMAMURA, M. and HONDA, M. (1978a): Cosmic-ray induced ^{53}Mn in Yamato-7301(j), -7305(k) and -7304(m) meteorites. Mem. Natl Inst. Polar Res., Spec. Issue, **8**, 209–219.

NISHIIZUMI, K., IMAMURA, M. and HONDA, M. (1978b): Nankyoku-san inseki-chû no uchû-sen seisei hôshanô (Cosmic-ray induced radioactivities in antarctic meteorites). Dai-3-kai Nankyoku Inseki Shinpojium kôen yôshi (Abstr. 3rd Symp. on Antarctic Meteorites), Tokyo, Natl Inst. Polar Res., 42–45.

SHIMA, MAKOTO, SHIMA, MASAKO and HINTENBERGER, H. (1973): Chemical composition and rare gas content of four new detected Antarctic meteorites. Earth Planet. Sci. Lett., **19**, 246–249.

SHIMA, MAKOTO, YABUKI, H. and YOSHIKAWA, K. (1978): Study on the extraterrestrial materials in Antarctica (VIII). On the distribution of the Yamato meteorites. Mem. Natl Inst. Polar Res., Spec. Issue, **8**, 101–109.

SHIRAISHI, K., NARUSE, R. and KUSUNOKI, K. (1976): Collection of Yamato meteorites, Antarctica, in December 1973. Nankyoku Shiryo (Antarct. Rec.) **55**, 49–60.

STAUFFER, H. (1962): On the production ratios of rare gas isotopes in stone meteorites. J. Geophys. Res., **67**, 2023–2028.

STEIGER, R. H. and JÄGER, E. (1977): Subcommission on geochronology: convention on the use of decay constants in geo- and cosmochronology. Earth Planet. Sci. Lett., **36**, 359–362.

TAKAOKA, N. (1976): A low-blank, metal system for rare gas analysis. Mass Spectr., **24**, 73–86.

TAKAOKA, N. and NAGAO, K. (1978): Rare gas studies of Yamato-7301(j), -7304(m) and -7305(k). Mem. Natl Inst. Polar Res., Spec. Issue, **8**, 209–219.

TAKEDA, H., MIYAMOTO, M., YANAI, K. and HARAMURA, H. (1978): A preliminary mineralogical examination of the Yamato-74 achondrites. Mem. Natl Inst. Polar Res., Spec. Issue, **8**, 170–184.

WASSON, J. T. (1974): Meteorites. Berlin, Springer-Verlag, 316 p.

YABUKI, H., YAGI, K. and ONUMA, K. (1978): Petrological studies of Yamato-74 meteorites (1). Mem. Natl Inst. Polar Res., Spec. Issue, **8**, 142–155.

YABUKI, H. (1978): Petrological studies on Yamato-74 meteorites. Sci. Papers Inst. Phys. Chem. Res., **72**, 14–40.

YANAI, K. (1976): 1974-nen no nankyoku-san Yamato inseki no tansa to saishû (Search and collection of Yamato meteorites, Antarctica, in November and December 1974). Nankyoku Shiryo (Antarct. Rec.), **56**, 70–81.

YANAI, K. (1978a): Yamato-74 meteorites collection, Antarctica, from November to December 1974. Mem. Natl Inst. Polar Res., Spec. Issue, **8**, 1–37.

YANAI, K. (1978b): First meteorites found in Victoria Land, Antarctica, December 1976 and January 1977—Report of the U.S.-Japan joint program titled "Antarctic Search for Meteorites" 1976–1977. Mem. Natl Inst. Polar Res., Spec. Issue, **8**, 51–69.

YANAI, K., MIYAMOTO, M. and TAKEDA, H. (1978): A classification for the Yamato-74 chondrites based on the chemical compositions of their olivine and pyroxene. Mem. Natl Inst. Polar Res., Spec. Issue, **8**, 110–120.

YOSHIDA, M., ANDO, H., OMOTO, K., NARUSE, R. and AGETA, Y. (1971): Discovery of meteorites near Yamato Mountains, East Antarctica. Nankyoku Shiryo (Antarct. Rec.), **39**, 62–65.

ZÄHRINGER, J. (1962): Isotopie-Effekt und Häufigkeiten der Edelgase in Steinmeteoriten und auf der Erde. Z. Naturforsch., **17a**, 460–471.

ZÄHRINGER, J. (1966): Primordial argon and the metamorphism of chondrites. Earth Planet. Sci. Lett., **1**, 379–382.

(*Received June 20, 1978*)

Note Added in Proof

Recently, OLSEN *et al.* gave the chemical and petrographic classification of Mt. Baldr No. 1, and Allan Hills No. 1 and No. 8 meteorites in "Meteoritics," **13**, 209 (1978). Mt. Baldr No. 1, and Allan Hills No. 1 and No. 8 are classified as H6, L6 and H6 chondrites, respectively. The petrologic type of these meteorites given by OLSEN *et al.* is in excellent agreement with that deduced from the concentration of trapped ^{132}Xe. It is noted that the isotopic composition of trapped Xe in the highly metamorphozed chondrite, Allan Hills No. 8 (H6), is identical with that of AVCC-Xe which is found in the undifferentiated primitive meteorites.