RARE GAS STUDIES OF YAMATO-7301 (j), -7304 (m) AND -7305 (k)

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Abstract: Yamato-7301 (H4 chondrite) and Yamato-7304 and -7305 (both L5 chondrites) were investigated for concentrations and isotopic compositions of rare gases. From the data of isotopic analyses of rare gases, cosmic-ray exposure ages on cosmogenic ^{3}He, ^{21}Ne and ^{38}Ar and gas-retention ages on radiogenic ^{4}He and ^{40}Ar were calculated as follows:

Meteorite	Cosmic-ray exposure age (m.y.)			Gas-retention age (b.y.)	
	T_3	T_{21}	T_{38}	T_4	T_{40}
Yamato-7301	8.19±1.78	13.2±3.8	11±3	3.7±0.9	4.1±0.4
7304	13.3±2.6	17.6±2.4	14±3	0.5±0.2	1.7±0.4
7305	16.9±1.7	21.7±2.4	23±3	2.0±0.6	4.1±0.2

Partial loss of helium was suggested for three samples. There were definitely observed ^{129}Xe-excesses and systematic enrichment in the light isotopes of xenon in all three meteorites. Isotopic anomalies were observed in the heavy isotopes of xenon in Yamato-7304 and the excess pattern resembles the fission yield pattern of ^{244}Pu.

1. Introduction

The Japanese Antarctic Research Expedition team found four different classes of nine meteorites at the southeast end of the Yamato Mountains in East Antarctica, in 1969 (YOSHIDA *et al.*, 1971). They were named "Yamato meteorites". Four of them have been investigated for chemical compositions and rare gas contents (SHIMA *et al.*, 1973). Following the finding of these Yamato meteorites, twelve meteorites were collected in the same area in 1973 (SHIRAISHI *et al.*, 1976). The Yamato meteorites are the first finding of an assemblage of meteorites in a limited area except the case of a meteorite shower.

It is known that in meteorites there exist, in general, five components of rare gases of different origins: 1) a cosmogenic component, 2) a radiogenic component, 3) a fissiogenic component, 4) a trapped component of primordial, ambient gases in the solar nebula absorbed by meteorite matters, 5) an atmospheric contamination. The quantitative determination of these components enables us to calculate the cosmic-ray exposure age and the gas-retention age of the meteorite, to estimate the abundance of extinct nuclides such as ^{129}I and ^{244}Pu and thereby to calculate the formation interval between the end of nucleosynthesis and the beginning of gas retention in the meteorite matter, and to put a constraint on the cosmochronology and the age of elements (WASSERBURG *et al.*, 1969; SCHRAMM and WASSERBURG, 1970). The primordial, trapped component gives information regarding the circumstances in the primitive solar nebula where the meteoritic bodies grew.

In this paper we report the result of isotopic analyses of rare gases in three Yamato meteorites collected in 1973.

2. Experimental Method

Meteorites used in this work are Yamato-7301(j), -7304(m) and -7305(k). Yamato-7301 has been classified as H4 chondrite in the VAN SCHMUS and WOOD classification (1967) and both Yamato-7304 and -7305 as L5 chondrites. Yamato-7301 was delivered to us in the forms of 0.6 gram of fragments and 0.4 gram of fine powder, and one gram each of Yamato-7304 and -7305 was delivered in fine powder. 0.1 to 0.4 gram of the samples were used for a rare gas measurement without any pretreatment such as mineral separation.

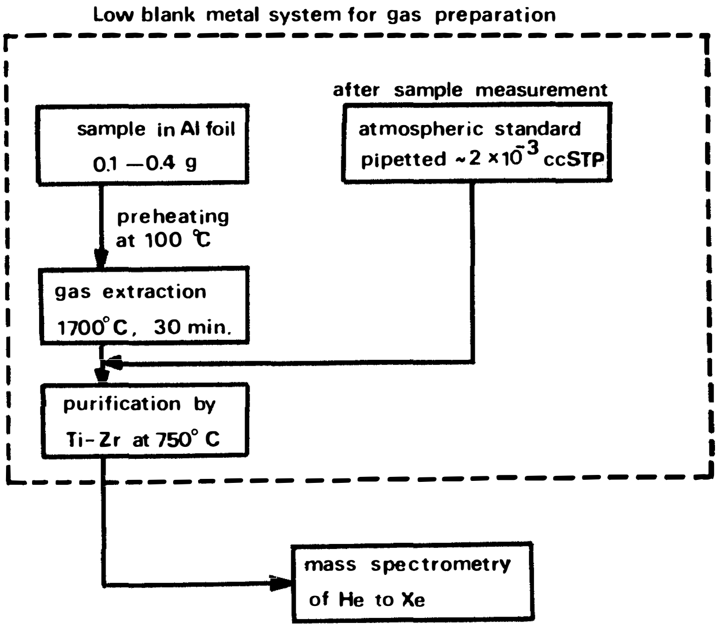

Fig. 1. Schematic diagram of experimental procedure. A gas preparation system is made of stainless steel to prevent the permeation of atmospheric helium. After extraction and purification of sample gases, rare gases were admitted into a mass spectrometer tuned to the mass resolution of 600 and analyzed with static operation. After a sample analysis, known amount of atmospheric standard gas was analyzed with the same procedure as applied to the sample gases to determine sensitivity and mass discrimination of the system.

Details of the instrument and the experimental technique used for the rare gas analysis have been published elsewhere (TAKAOKA, 1976). A schematic diagram of the analytical procedure is given in Fig. 1. The rare gas preparation system is made of stainless steel to prevent the permeation of atmospheric helium, except a sample holder and a vacuum gage which are made of glass, and is pumped out with mercury diffusion pumps. After the samples wrapped with 10 μm thick aluminium foil were mounted in the sample holder, the whole system was thoroughly baked out at 250°C for a night. The samples mounted in the sample holder were separately heated at about 100°C for a night to remove the adsorbed air. After sufficient degassing of a crucible, a blank was run. The blank for rare gases was negligible relative to sample gases, so no correction for blank was applied. Then the sample was dropped into the crucible and heated at 1,700°C for 30 minutes. Released gases were exposed to titanium-zirconium getters heated at 750°C to purify the rare gases by adsorbing the reactive gases. The purified gases were separated for the mass spectrometries into three fractions by the help of adsorption on activated charcoal at liquid nitrogen and dry-ice temperatures: helium-neon, argon and krypton-xenon fractions. Vapor pressure of krypton is relatively high at dry-ice temperature. An appreciable amount (approximately 15 per cent) of krypton appeared in the argon fraction. The amount of krypton which was lost in the argon fraction was corrected. However, since data of krypton showed relatively wide variations, they should be re-examined and are not presented here. Doubly-charged ions of ^{40}Ar and CO_2 interfering with the measurements of ^{20}Ne and ^{22}Ne respectively were negligibly low and hydrocarbon peaks were separated from rare gas peaks with the high resolution power of 600.

3. Results and Discussion

The rare gas concentrations and their isotopic ratios determined for three Yamato chondrites are shown in Tables 1 and 4. They are weighted means of several analyses by sample weight. Errors cited are standard deviations ($\pm$one σ). The isotopic ratios of atmospheric rare gases are also given for comparison. The data of Yamato-7301 showed a larger scattering than those of the other two samples. Uncertainties in the sensitivity of machine were not more than 10 per cent. One of causes to consider is an effect of weathering in a long terrestrial age. As discussed later, this stone may have an extremely long terrestrial age (NISHIIZUMI *et al.*, 1978). The weathering in the long terrestrial age might enhance inhomogeneous gas losses from mineral to mineral. As the rare gases in Yamato-7301 were analyzed using relatively coarse fragments, inhomogeneity might follow in the gas concentration.

Helium in three meteorites could be understood in terms of mixture of radiogenic ^{4}He and cosmogenic helium. The concentration of radiogenic ^{4}He was estimated by $^{4}He_{rad} = {}^{4}He - 5 \times {}^{3}He$, on the assumptions that all of ^{3}He are cosmogenic and that the $(^{3}He/^{4}He)_{cos}$ ratio is equal to 0.2. The contents of

Table 1. Concentrations and isotopic ratios of helium, neon and argon in Yamato-7301, -7304 and -7305 chondrites. Concentrations in $10^{-8}cm^3STP/g$.

Sample	Yamato-7301	Yamato-7304	Yamato-7305	Atmosphere
^3He	20.3 ± 4.0	33.0 ± 5.6	41.8 ± 1.8	—
^4He	1360 ± 270	344 ± 58	987 ± 42	—
$^3He/^4He$	0.0150 ± 0.0008	0.0960 ± 0.0009	0.0424 ± 0.0005	1.3×10^{-6}*
^{22}Ne	6.45 ± 1.78	8.98 ± 1.11	11.2 ± 1.0	—
$^{20}Ne/^{22}Ne$	0.828 ± 0.029	0.975 ± 0.009	0.885 ± 0.005	9.800**
$^{21}Ne/^{22}Ne$	0.889 ± 0.008	0.914 ± 0.008	0.908 ± 0.012	0.0290**
^{36}Ar	1.83 ± 0.36	4.87 ± 1.00	4.04 ± 0.14	—
^{40}Ar	5980 ± 1190	1170 ± 240	6910 ± 240	—
$^{38}Ar/^{36}Ar$	0.511 ± 0.060	0.333 ± 0.003	0.470 ± 0.007	0.187***
$^{40}Ar/^{36}Ar$	3270 ± 630	240 ± 2	1710 ± 20	295.5***

* COON (1949), ** EBERHARDT *et al.* (1965), *** NIER (1950a).

cosmogenic ^4He in Yamato-7301, -7304 and -7305 were 8, 48 and 21 per cent of the total ^4He, respectively. Neon in Yamato-7301 and -7305 is composed mostly of the cosmogenic component. Hence, no correction was applied. Neon in Yamato-7304 was a mixture of the cosmogenic and the minor trapped components. ^{36}Ar and ^{38}Ar in three meteorites could be understood in terms of the mixture of the cosmogenic and the trapped components. The trapped components accounted for 80 per cent of ^{36}Ar measured in Yamato-7301 and -7305 and for 90 per cent of ^{36}Ar in Yamato-7304. The trapped argon in Yamato-7304 will be discussed later in a relation with the ^{132}Xe concentration. Most of ^{40}Ar was radiogenic. The isotopic ratios of xenon will be discussed in a separate section.

3.1. Cosmic-ray exposure age

The cosmic-ray exposure age is calculated using the concentration of cosmogenic stable nuclides produced by the cosmic-ray irradiation and the production rate of the cosmogenic nuclides. The production rate P_i (i=mass number) of cosmogenic rare gases, which depends on the elemental composition of meteorite and on the energy spectrum and intensity of cosmic-rays, has been empirically formulated as a function of the elemental composition by several authors.

Table 2 presents the concentrations of cosmogenic ^3He, ^{21}Ne and ^{38}Ar, and the cosmic-ray exposure ages calculated. All of ^3He were assumed to be cosmogenic for three samples. The concentrations of cosmogenic ^{21}Ne and ^{38}Ar were calculated from the following equations,

$$(^{21}Ne)_c=(21/22)_c\frac{(21/22)_m-(21/22)_t}{(21/22)_c-(21/22)_t}(^{22}Ne)_m\,,$$

and

Table 2. Cosmogenic rare gases and cosmic-ray exposure ages.

Sample	Yamato-7301	Yamato-7304	Yamato-7305
$(^{3}He)_c$*	20.3±4.0	33.0±5.6	41.8±1.8
$(^{21}Ne)_c$*	5.72±1.58	8.19±1.01	10.1±0.9
$(^{38}Ar)_c$*	0.676±0.186	0.808±0.172	1.30±0.08
P_3**	2.48±0.23	2.48±0.23	2.48±0.23
P_{21}**	0.433±0.029	0.466±0.031	0.466±0.031
P_{38}**	0.061±0.007	0.056±0.005	0.056±0.005
$(^{3}He/^{21}Ne)_c$	3.54±1.20	4.03±0.85	4.14±0.41
P_3/P_{21}	5.73±0.66	5.32±0.61	5.32±0.61
$(^{21}Ne/^{38}Ar)_c$	8.46±3.30	10.1±2.5	7.77±0.84
P_{21}/P_{38}	7.1±0.6	8.3±0.6	8.3±0.6
T_3 (m.y.)	8.19±1.78	13.3±2.6	16.9±1.7
T_{21} (m.y.)	13.2±3.8	17.6±2.4	21.7±2.4
T_{38} (m.y.)	11±3	14±3	23±3

* Observed concentrations in 10^{-8} cm^3 STP/g.
** Production rates in 10^{-8} cm^3 STP/g/m.y. (see text).

$$(^{38}Ar)_c = \cancel{(38/36)_c}\frac{(38/36)_m-(38/36)_t}{(38/36)_c-(38/36)_t}(^{36}Ar)_m ,$$

where suffixes c, m and t represent cosmogenic, measured and trapped, respectively. The following isotopic ratios were assumed: $(21/22)_c=0.90$ and $(38/36)_c=1.55$, and $(21/22)_t=0.030$ and $(38/36)_t=0.187$. The production rates of cosmogenic ^{3}He and ^{21}Ne are: $P_3=(2.48\pm0.23)\times10^{-8}$ cm^3 STP/g/m.y. for both H and L chondrites; $P_{21}=(4.33\pm0.29)\times10^{-9}$ and $(4.66\pm0.31)\times10^{-9}$ cm^3 STP/g/m.y. for H and L chondrites, respectively (HERZOG and ANDERS, 1971). The production rate of ^{38}Ar was calculated using the 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 meteorites used in this work, the average chemical compositions for H and L chondrites (MASON, 1971) were employed for calculation of P_{21}/P_{38}. In Table 2, the ratios 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. Agreement between P_{21}/P_{38} and $(^{21}Ne/^{38}Ar)_c$ is good for three samples investigated, whereas the $(^{3}He/^{21}Ne)_c$ ratios are systematically lower than the P_3/P_{21} ratios, *i.e.* 37, 25 and 23 per cent lower for Yamato-7301, -7304 and -7305, respectively. This suggests diffusive partial loss of helium through meteorite minerals. For this reason the exposure ages calculated on cosmogenic ^{3}He are less reliable. The mean cosmic-ray exposure ages on Ne and Ar are 12±2.5, 16±1.9 and 23±1.7 m.y. for Yamato-7301, -7304 and -7305, respectively. These ages are in the typical range of exposure ages for H and L chondrites (ZÄHRINGER, 1968).

However, the content of cosmogenic ^{53}Mn in Yamato-7301 is approximately one-fourth of that expected for the exposure age estimated from the cosmogenic rare gases (NISHIIZUMI *et al.*, 1977). This is suggesting an extremely old terrestrial age for this stone, a lower cosmic-ray irradiation in a large body of $\sim$100 tons with heavy shielding, multistage irradiation history or a combined mechanism (NISHIIZUMI *et al.*, 1977). The ^{21}Ne production rate is relatively sensitive to shielding, and the ^{22}Ne/^{21}Ne and ^{3}He/^{21}Ne ratios are also functions of the shielding. EBERHARDT *et al.* (1966) and WRIGHT *et al.* (1973) have argued a relation between the ^{21}Ne production rate and the ^{22}Ne/^{21}Ne and ^{3}He/^{21}Ne ratios. According to them, increased shielding enhances the ^{21}Ne production and reduces the ^{22}Ne/^{21}Ne ratio. For instance, a very slightly shielded sample of the Saint Séverin meteorite gave a low ^{21}Ne production rate (0.274×10^{-8} cm^3 STP/g/m.y.) and a high ^{22}Ne/^{21}Ne ratio (1.215) (MARTI *et al.*, 1969). The ^{22}Ne/^{21}Ne ratio is insensitive to gas loss compared with the ^{3}He/^{21}Ne ratio. The ^{22}Ne/^{21}Ne ratio for Yamato-7301 is 1.12 $\pm$0.01, a typical value for chondrites. Using this ratio and the result according to Fig. 5 in EBERHARDT *et al.* (1966), we find $P_{21}=0.40\times 10^{-8}$ cm^3 STP/g/m.y. as the ^{21}Ne production rate, which is in agreement with the value used in the present work. However, there have been so far no measurement of the cosmogenic rare gases in a large chondrite of the order of about 10^2 tons. The relation between the ^{21}Ne production rate and the ^{22}Ne/^{21}Ne ratio might not be simply extrapolated to a deep interior of a large-size stone (HONDA, 1977).

BEGEMANN *et al.* (1976) have measured the rare gases and ^{36}Cl in the Bondoc mesosiderite and estimated the production rates of cosmogenic ^{21}Ne and ^{38}Ar. The Bondoc mesosiderite is known for its large preatmospheric size of about 200 cm (CRESSY and SHEDLOVSKY, 1965; BORN and BEGEMANN, 1975; NISHIIZUMI *et al.*, 1977). The production rates of cosmogenic ^{21}Ne from Mg and of cosmogenic ^{38}Ar from Ca in Bondoc, estimated by BEGEMANN *et al.* (1976), are one order of magnitude lower than small size meteorites. If this is true for other target elements and if Yamato-7301 is a fragment broken from near the center of a large parent body, say 200 cm radius, the production rates might be overestimated and should be reduced to about 1/7, and thus the cosmic-ray exposure age of Yamato-7301 would be extended to around 90 m.y. It is not inconsistent with a two-stage irradiation model by NISHIIZUMI *et al.* (1978), where the stone was irradiated more than 60 m.y. in a place deeper than a few meters from the surface of the parent body and in the next stage it was exposed to a normal level of cosmic-ray flux for less than 1.6 m.y. The low $(^{3}\mathrm{He}/^{21}\mathrm{Ne})_c$ ratio for Yamato-7301 may be due not only to the diffusive partial loss of ^{3}He, but also to the heavy shielding in a large parent body.

3.2. *U/Th-He and K-Ar ages*

The U/Th-He and K-Ar ages are given in Table 3. Since no direct measure-

Table 3. U/Th-He and K-Ar ages for Yamato-7301, -7304 and -7305.

Meteorite	Yamato-7301	Yamato-7304	Yamato-7305
$(^4He)_{rad}$*	1.3±0.3	0.18±0.06	0.78±0.04
U (ppb)**	11	15	15
U/Th-He age (b.y.)	3.7±0.9	0.5±0.2	2.0±0.6
$(^{40}Ar)_{rad}$*	5.98±1.19	1.17±0.24	6.91±0.24
K (ppm)***	1000	1100	1100
K-Ar age (b.y.)	4.1±0.4	1.7±0.4	4.1±0.2

U/Th-He ages were calculated on the basis of $\lambda_{232}=4.95\times10^{-11}$ y^{-1}, $\lambda_{235}=9.85\times10^{-10}$ y^{-1}, $\lambda_{238}=1.55\times10^{-10}$ y^{-1}, and K-Ar ages were on the basis of $\lambda=4.96\times10^{-10}$ y^{-1}, $\lambda_e=5.81\times10^{-11}$ y^{-1}. Errors cited in ages include uncertainties in rare gas, U and K concentrations. The uncertainties in U and K concentrations were assumed to be 30 and 10%, respectively.

* Rare gas concentration in 10^{-5} cm^3STP/g.
** MORGAN (1971), Th/U ratio of 3.6 was assumed.
*** SHIMA (1977).

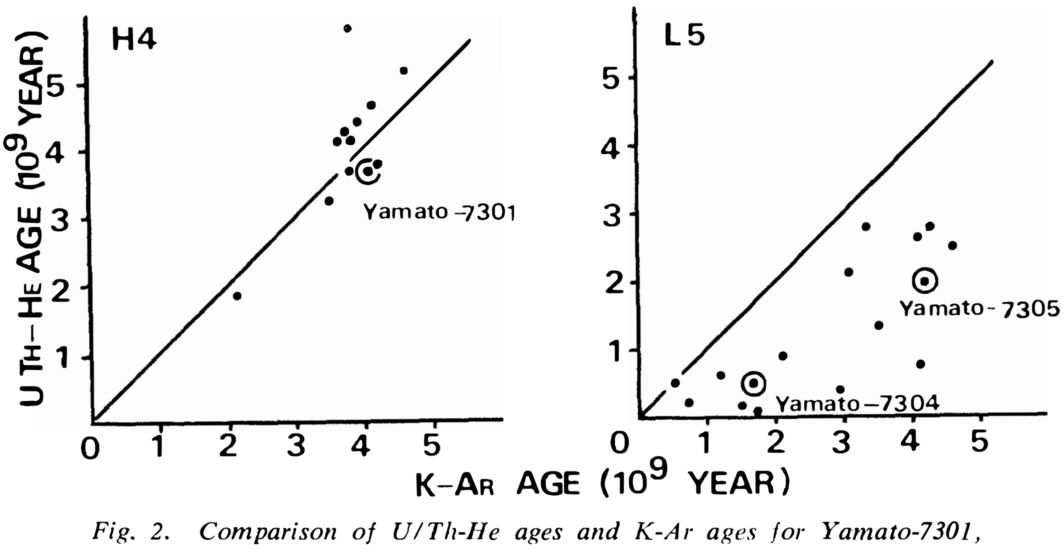

Fig. 2. Comparison of U/Th-He ages and K-Ar ages for Yamato-7301, -7304 and -7305 with those for other chondrites. Plots for other chondrites are according to FISHER (1972).

ment of the content of uranium in the Yamato meteorite was available, mean contents of uranium for H and L chondrites in literature (MORGAN, 1971) were used for calculation of the U/Th-He ages, *i.e.* 11 and 15 ppbU for H and L chondrites, respectively. The U/Th-He ages are systematically younger than the K-Ar ages for three samples. It is presumably due to partial loss of ^4He and is consistent with the low exposure ages obtained on cosmogenic ^3He.

The K-Ar ages obtained are 4.1, 1.7 and 4.1 b.y. for Yamato-7301, -7304 and -7305, respectively. As Yamato-7304 has a remarkably young age, this stone is considered to have suffered metamorphism in the later stage of its history and to have lost some parts of cumulate gases. In Yamato-7304 fine lamellar structures which are due to a shock effect have been reported by YAGI *et al.* (1977). However, the K-Ar ages obtained above fall in a cluster in the K-Ar histograms given by ZÄHRINGER (1968). Fig. 2 presents plots of correlation between the U/Th-He ages and the K-Ar ages together with the data for other chondrites in literature (FISHER, 1972). Yamato-7304 and -7305, both L5 chondrites, show large differences between the U/Th-He ages and the K-Ar ages, while Yamato-7301, H4 chondrite, gives a fair agreement between them.

It has been mentioned that the ratio of trapped ^{36}Ar to ^{132}Xe in ordinary chondrites is remarkably constant around 100 (MARTI, 1967; ZÄHRINGER, 1968; HEYMANN, 1971). This ratio for Yamato-7304 is appreciably high (*i.e.* 740 $\pm$200), whereas Yamato-7301 and -7305 resemble the literature value, 91 $\pm$36 for Yamato-7301 and 300$\pm$10 for Yamato-7305. The atmospheric contamination may be one of the causes of the high ^{36}Ar/^{132}Xe ratio for Yamato-7304. If this is the case, we could correct the trapped component for the atmospheric contamination assuming the U/Th-He age (0.5 b.y.) as a lower limit of the K-Ar age. The correction results in that 77 per cent of the trapped ^{36}Ar calculated earlier can be attributed to atmospheric argon and thus the ^{36}Ar/^{132}Xe ratio decreases to 300. It suggests that the K-Ar age of 1.7 b.y. for Yamato-7304 may be too old. To reach a definite conclusion regarding the extent of atmospheric contamination, more detailed studies with a refined technique are required.

3.3. *Isotopic anomalies in xenon*

The isotopic ratios of xenon listed in Table 4 show that there are definite excesses of ^{129}Xe, the decay products of extinct ^{129}I, and systematic enrichment in the light isotopes of xenon relative to atmospheric xenon, which are characteristic trends in isotopic composition of meteorite trapped xenon discovered by REYNOLDS (1960) and observed in many meteorites by many workers. Excesses in ^{124}Xe and ^{126}Xe relative to AVCC-Xe (Average Carbonaceous Chondrite-Xe) (EUGSTER *et al.*, 1967) are observed in three samples. The ratios of excessive ^{124}Xe to ^{126}Xe are around 0.5, the ratio characteristic of cosmogenic xenon. In addition, the isotopic anomalies in the heavy isotopes are found in Yamato-7304. The amount of excess xenon relative to AVCC-Xe was calculated. The amount of excessive ^{136}Xe is 1.5×10^{-12} cm^3 STP/g, which exceeds the fissiogenic ^{136}Xe produced from ^{238}U during 4.6 b.y. in the chondrite which contains 15 ppb uranium. Yamato-7304 has lost a great part of the radiogenic ^{40}Ar as mentioned already. It is a general trend for radiogenic ^{40}Ar to be released at relatively low temperature because potassium resides in low-temperature minerals. It is likely that the loss of fissiogenic xenon is less than radiogenic argon, since xenon has a smaller diffusion coefficient than argon. Fig. 3 presents the excess xenon pattern normalized to ^{136}Xe. It is suggesting that those excesses are due to fission products of uranium or a transuranium element. The fission yield patterns for ^{235}U (VANDENBOSCH and HUIZENGA, 1973), ^{238}U (WETHERILL,

Table 4. Isotopic ratios and concentrations of xenon in Yamato-7301, -7304 and -7305 chondrites. Isotopic ratios are normalized to $^{132}Xe=100$.

Sample	Yamato-7301	Yamato-7304	Yamato-7305	Atmosphere*
^{124}Xe	0.48±0.03	0.56±0.03	0.53±0.03	0.36
^{126}Xe	0.46±0.03	0.56±0.06	0.59±0.03	0.33
^{128}Xe	7.73±0.19	7.66±0.13	7.93±0.32	7.136
^{129}Xe	123±16	107±1	117±3	98.33
^{130}Xe	15.7±0.5	15.7±0.1	15.9±0.4	15.2
^{131}Xe	80.6±0.7	80.7±0.5	80.6±0.6	78.77
^{132}Xe	=100	=100	=100	=100
^{134}Xe	38.7±0.5	39.9±0.5	39.0±0.3	38.7
^{136}Xe	33.3±0.2	34.1±0.5	33.1±0.2	33.0
^{132}Xe ($10^{-10}cm^3STP/g$)	1.52±0.58	0.587±0.154	1.08±0.03	—

* NIER (1950b).

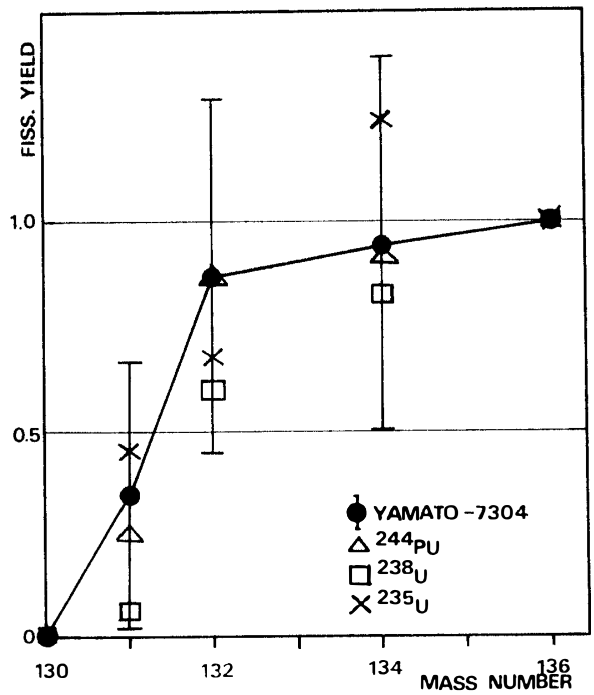

Fig. 3. Excess xenon pattern in Yamato-7304 meteorite normalized to $^{136}Xe=1$. AVCC-Xe was assumed as a trapped component. Excessive ^{136}Xe amounts to 1.5×10^{-12} cm^3STP/g. Relative fission yield patterns normalized to $^{136}Xe=1$ for ^{235}U (neutron-induced), ^{238}U and ^{244}Pu are plotted for comparison.

1953) and ^{244}Pu (ALEXANDER *et al.*, 1971) are plotted for comparison. Though the excess pattern is in good agreement with the fission yield pattern for ^{244}Pu, large error bars on the present result make the identification of the parent nuclide difficult.

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