Mass Spectrometrical Study of Rare Gas Compositions and Neutron Capture Effects in Yamato-74191 (L 3) Chondrite

Nobuo Takaoka * and Keisuke Nagao **

Department of Physics, Faculty of Science, Osaka University, Toyonaka, Osaka 560, Japan

Z. Naturforsch. **35a**, 29–36 (1980); received December 5, 1979

Dedicated to Prof. H. Hintenberger on the occasion of his 70th birthday

The unequilibrated hypersthene chondrite Yamato-74191 was studied mass spectrometrically for rare gases released at various temperatures. Cosmogenic gases dominate in He and Ne. The meteorite contains large amounts of trapped Ar, Kr and Xe, and radiogenic ^{40}Ar and ^{129}Xe. Cosmic-ray irradiation and K-Ar ages were determined.

In addition to spallogenic components of Kr and Xe, isotopic excesses of ^{80}Kr, ^{82}Kr, ^{128}Xe and ^{126}Xe relative to AVCC-Kr and -Xe were found. The ratio of ^{80}Kr-excess to ^{82}Kr-excess is 2.66 after correction for spallogenic Kr. A correlation between ^{128}Xe/^{132}Xe and ^{129}Xe/^{132}Xe was found The ^{129}Xe/^{132}Xe ratio for trapped Xe in Yamato-74191 was determined as 1.12 ± 0.29 with the correlation plot.

The excesses found in Yamato-74191 are best explained by epithermal neutron capture on Br and I, and by the ^{127}I(n, 2nβ) ^{126}Xe reaction. Using neutron-produced ^{80}Kr, the neutron slowing-down density was estimated to be 0.14 ± 0.03 cm^{-3} sec^{-1}.

A minimum mass and a preatmospheric radius was estimated to be 470 kg and 32 cm, respectively.

1. Introduction

The unequilibrated hypersthene chondrite Yamato-74191 contains large amounts of trapped Ar, Kr and Xe, and the isotopic ratio of Kr in this chondrite shows large excesses of ^{80}Kr and ^{82}Kr relative to AVCC-Kr [1]. The ratio of excessive ^{80}Kr to ^{82}Kr was found to be 2.7, and these excesses were attributed to products of epithermal neutron capture on Br [2]. The isotopic composition of Xe was identical with the AVCC-Xe composition, except for small excesses of ^{128}Xe and ^{126}Xe. The ^{128}Xe excesses were also attributed to neutron capture on I, but the origin of the ^{126}Xe excess was not clear in our previous work [2].

One of the neutron sources in meteorites is cosmic-ray irradiation, which produces high-energy secondary neutrons. Excesses of Kr and Xe isotopes which originate from neutron capture on Br, Se and I in meteorites have been reported previously [3–5]. Since the total intensity and the energy distribution of neutrons in the meteorite depend on the duration of cosmic-ray irradiation and the moderation depth of neutrons in the meteorite, the determination of neutron-capture effects on rare gas isotopes can give a constraint on models for cosmic-ray irradiation history and information about the preatmospheric size of the meteorite.

In this paper we report on results of rare gas measurements and conspicuous neutron-capture effects on Kr and Xe isotopes in the Yamato-74191 chondrite.

2. Experimental Techniques and Results

The meteorite was crushed in a stainless-steel mortar and a grain size fraction finer than 147 μm (100 mesh) was used for the measurements. The sample, weighing 0.427 g, was wrapped with thin aluminium foil (27.5 mg in weight) and degassed at about 80 °C for a night in the side arm of a sample holder of the extraction line. After a blank run, the sample was dropped into a thoroughly degassed molybdenum crucible and heated at successively higher temperatures of 700, 900, 1100, 1300, 1500 and 1750 °C. Each temperature was kept constant for 25 minutes, and the evolved gases were analysed with a high-sensitivity mass spectrometer. Details of the instrument and rare gas analysis techniques have been given elsewhere [2, 6].

The concentrations and isotopic compositions of the rare gases are given in Table 1. Blank correc-

* Reprint requests to Prof. N. Takaoka to the present address: Department of Earth Sciences, Faculty of Science, Yamagata University, Koshirakawa-cho 1-4-12 Yamagata 990, Japan.

** Present address: Graduate School, Okayama University of Science, Ridai-cho 1-1, Okayama 700, Japan.

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Table 1. Concentration and isotopic composition of rare gases in Yamato-74191(0—147 μm).

Temperature (°C)	700	900	1100	1300	1500	1750	Total	Yamato-74191[a] (47—417 μm)
^{3}He	11.0	2.22	0.857	0.0736	0.0053	0.0001	14.16	10.6
^{4}He	498.0	127.0	51.3	9.92	0.328	0.0241	686.6	545.0
^{3}He/^{4}He	0.0220 ± 0.0004	0.0175 ± 0.0003	0.0167 ± 0.0003	0.00729 ± 0.00015	0.0161 ± 0.0012	0.0004	0.0206 ± 0.0004	0.0191 ± 0.0002
^{21}Ne	0.510	0.640	1.73	1.31	0.500	0.00046	4.69	3.02
^{22}Ne/^{21}Ne	1.277 ± 0.006	1.174 ± 0.006	1.168 ± 0.011	1.142 ± 0.014	1.043 ± 0.005	— — —	1.160 ± 0.010	1.185 ± 0.010
^{21}Ne/^{20}Ne	0.545 ± 0.005	1.016 ± 0.007	0.983 ± 0.007	0.951 ± 0.008	1.027 ± 0.006	— — —	0.904 ± 0.007	0.870 ± 0.009
^{36}Ar	0.595	0.447	19.6	11.8	0.816	0.017	33.28	34.1
^{38}Ar/^{36}Ar	0.212 ± 0.003	0.212 ± 0.003	0.198 ± 0.003	0.200 ± 0.003	0.245 ± 0.002	0.190 ± 0.007	0.200 ± 0.003	0.196 ± 0.002
^{40}Ar/^{36}Ar	1752.0 ± 14.0	922.0 ± 3.0	140.2 ± 1.7	11.2 ± 0.2	33.4 ± 0.4	266.0 ± 13.0	131.2 ± 1.4	102.0 ± 1.0
^{84}Kr	2.41	0.430	7.15	12.4	1.12	0.127	23.64	22.6
^{78}Kr/^{84}Kr	0.0063 ± 0.0006	0.0070 ± 0.0015	0.00602 ± 0.00026	0.00615 ± 0.00012	0.0058 ± 0.0004	— — —	0.00612 ± 0.00025	0.00650 ± 0.00022
^{80}Kr/^{84}Kr	0.485 ± 0.011	0.757 ± 0.014	0.124 ± 0.002	0.0525 ± 0.0008	0.0495 ± 0.0026	0.0398 ± 0.0028	0.131 ± 0.003	0.103 ± 0.002
^{82}Kr/^{84}Kr	0.371 ± 0.010	0.477 ± 0.009	0.236 ± 0.004	0.208 ± 0.003	0.207 ± 0.004	0.201 ± 0.007	0.238 ± 0.004	0.227 ± 0.001
^{83}Kr/^{84}Kr	0.202 ± 0.004	0.208 ± 0.005	0.207 ± 0.003	0.205 ± 0.002	0.203 ± 0.004	0.201 ± 0.007	0.205 ± 0.003	0.205 ± 0.003
^{86}Kr/^{84}Kr	0.307 ± 0.005	0.313 ± 0.004	0.313 ± 0.003	0.311 ± 0.003	0.307 ± 0.005	0.304 ± 0.008	0.311 ± 0.003	0.311 ± 0.005
^{132}Xe	2.46	0.397	3.89	11.0	1.36	0.040	19.14	22.2
^{124}Xe/^{132}Xe	0.00398 ± 0.00023	0.0054 ± 0.0005	0.00492 ± 0.00029	0.00468 ± 0.00029	0.0050 ± 0.0004	— — —	0.00468 ± 0.00029	0.00463 ± 0.00007
^{126}Xe/^{132}Xe	0.00359 ± 0.00025	0.0060 ± 0.0006	0.00485 ± 0.00032	0.00423 ± 0.00024	0.0044 ± 0.0003	— — —	0.00432 ± 0.00027	0.00432 ± 0.00005
^{128}Xe/^{132}Xe	0.0805 ± 0.0018	0.126 ± 0.006	0.0981 ± 0.0024	0.0848 ± 0.0020	0.0848 ± 0.0019	0.082 ± 0.011	0.0878 ± 0.0021	0.0853 ± 0.0005
^{129}Xe/^{132}Xe	1.970 ± 0.024	12.46 ± 0.42	6.19 ± 0.09	1.692 ± 0.019	1.74 ± 0.03	1.17 ± 0.04	2.868 ± 0.043	2.34 ± 0.01
^{130}Xe/^{132}Xe	0.156 ± 0.002	0.164 ± 0.005	0.163 ± 0.003	0.163 ± 0.002	0.164 ± 0.004	0.156 ± 0.011	0.1615 ± 0.0024	0.162 ± 0.001
^{131}Xe/^{132}Xe	0.799 ± 0.011	0.807 ± 0.019	0.817 ± 0.010	0.815 ± 0.009	0.822 ± 0.011	0.840 ± 0.018	0.8140 ± 0.0098	0.819 ± 0.005
^{134}Xe/^{132}Xe	0.381 ± 0.006	0.377 ± 0.005	0.375 ± 0.005	0.379 ± 0.004	0.378 ± 0.005	0.382 ± 0.011	0.3785 ± 0.0046	0.382 ± 0.002
^{136}Xe/^{132}Xe	0.319 ± 0.009	0.321 ± 0.006	0.316 ± 0.006	0.318 ± 0.005	0.312 ± 0.008	0.323 ± 0.012	0.3175 ± 0.0060	0.320 ± 0.003

Rare gas concentrations are given in units of 10^{-8} cm^{3} STP/g for He, Ne and Ar, and in units of 10^{-10} cm^{3} STP/g for Kr and Xe. Uncertainties in concentration are 6, 12, 10, 10 and 8 per cent for He, Ne, Ar, Kr and Xe, respectively.

[a] Nagao and Takaoka [2].

tions were made for the concentrations and the isotopic ratios. The blank correction was typically less than 2%, except for the 1750 °C fraction. The errors given in Table 1 are based on the statistical errors of ratio measurements, the uncertainties in mass discrimination and blank correction. In the 1750 °C fraction only ^{21}Ne exceeded the blank. Therefore, the concentration of ^{21}Ne in this temperature fraction is given in Table 1, but the isotopic ratios of Ne are not given.

Cosmogenic gases dominate in He and Ne. Compared with our previous results [2], given in Table 1 for comparison, the present sample released more He and Ne of cosmic-ray origin. The concentrations of trapped Ar, Kr and Xe are practically the same in the two samples, whereas radiogenic ^{40}Ar and ^{129}Xe, and the isotopic excesses of ^{80}Kr, ^{82}Kr and ^{128}Xe are enhanced in the present sample.

Figure 1 shows release patterns of Kr and Xe components. The radiogenic ^{129}Xe was calculated as the ^{129}Xe-excess beyond the trapped $^{129}Xe/^{132}Xe$ ratio obtained from the correlation plot of $^{128}Xe/^{132}Xe$ against $^{129}Xe/^{132}Xe$ (Figure 3). As discussed by Nagao and Takaoka [2] and will be discussed later, the ^{80}Kr and ^{82}Kr-excesses and ^{128}Xe-excess found in Yamato-74191 are attributable to epithermal neutron capture on Br and I, respectively. The release pattern of neutron-produced ^{80}Kr shows two peaks at 700 °C and 1100 °C, while trapped ^{84}Kr gives a single peak at 1300 °C. The release patterns of neutron-produced ^{128}Xe and radiogenic ^{129}Xe are quite similar to each other but different from the pattern of trapped ^{132}Xe release which has a peak at 1300 °C, similar to trapped ^{84}Kr. The significant degassing of neutron-produced ^{80}Kr at 700 °C may be due to poor retentivity of rare gases in Br-bearing minerals.

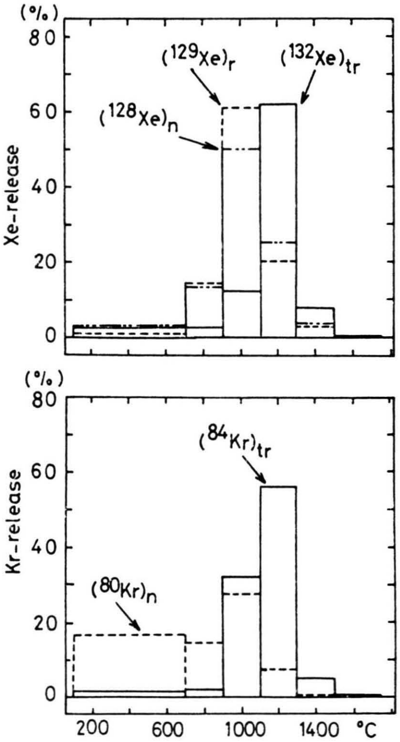

Fig. 1. Release patterns for neutron-produced ^{80}Kr and trapped ^{84}Kr, and for neutron-produced ^{128}Xe, radiogenic ^{129}Xe and trapped ^{132}Xe.

3. Discussion

3.1. Cosmic-ray irradiation and K-Ar ages

A cosmic-ray irradiation age T_i was calculated by

$$T_i = N_i / P_i ,$$

where N_i and P_i are the concentration and produc-

Table 2. Cosmic-ray irradiation and K-Ar ages for Yamato-74191.

Sample	Yamato-74191 (This work)	Yamato-74191 (Nagao and Takaoka [2])
$^{3}He_c$ [a]	14.2	10.6
$^{21}Ne_c$ [a]	4.69	3.01
$^{38}Ar_c$ [a]	0.503	0.350
$^{40}Ar_r$ [a]	4096.	3490.
$(^{3}He/^{21}Ne)_c$	3.03	3.52
$(^{21}Ne/^{38}Ar)_c$	9.32	8.60
P_3 [b]	2.48	2.48
P_{21} [b]	0.466 (0.505) [d]	0.466 (0.505) [d]
P_{38} [b]	0.0503 (0.0696) [d]	0.0503 (0.0696) [d]
P_3/P_{21}	5.32 (4.91) [d]	5.32 (4.91) [d]
P_{21}/P_{38}	9.26 (7.26) [d]	9.26 (7.26) [d]
T_3 [c]	5.73	4.27
T_{21} [c]	10.1 (9.28) [d]	6.46 (5.96) [d]
T_{38} [c]	10.0 (7.23) [d]	6.96 (5.03) [d]
T_{40} [c]	3530. [e]	3400. [e]

[a] Concentrations in units of 10^{-8} cm^3 STP/g.
[b] Production rates in units of 10^{-8} cm^3 STP/(g.my).
[c] Cosmic-ray irradiation and K-Ar ages in units of million years (my).
[d] Production rates and cosmic-ray irradiation ages calculated with Bogard and Cressy [9] are shown in parentheses.
[e] For the calculation of the K-Ar age, the decay constants $\lambda_{\beta^-} = 4.96 \times 10^{-10}$/y and $\lambda_{\beta^+} + \lambda_e = 0.581 \times 10^{-10}$/y were used.

tion rate of cosmogenic nuclide i, respectively. Table 2 shows the concentrations of cosmogenic ^{3}He, ^{21}Ne and ^{38}Ar, and the cosmic-ray irradiation ages. 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: $(^{22}Ne/^{21}Ne)_{tr} = 34.5$ and $(^{38}Ar/^{36}Ar)_{tr} = 0.187$, and $(^{22}Ne/^{21}Ne)_{c} = 1.14$ and $(^{38}Ar/^{36}Ar)_{c} = 1.55$ for trapped and cosmogenic Ne and Ar, respectively.

The production rates given by Herzog and Anders [7] were used for cosmogenic ^{3}He and ^{21}Ne. The production rate of cosmogenic ^{38}Ar was calculated using the empirical production ratio of cosmogenic ^{21}Ne and ^{38}Ar given by Stauffer [8] and the ^{21}Ne production rate employed above. The production rates of cosmogenic ^{21}Ne and ^{38}Ar given by Bogard and Cressy [9] were also calculated with the bulk chemical composition of Yamato-74191 meteorite, listed in Table 3, and the results are given in Table 2. The recalculated results of our previous measurements are given in Table 2. The agreement is good between the ^{21}Ne ages calculated with both production rates mentioned. However, there is a difference between the present result and the previous one. This is due to the difference in the concentration of cosmogenic ^{21}Ne. The reason why the fine grained sample used in this work contains greater amounts of cosmogenic nuclides than the coarse grained sample used in the previous work is not obvious. The difference in the rare gas concentration between them is beyond experimental error. One possibility is chemical inhomogeneity resulting from the grain size separation of the crushed sample. A wide variation of chemical composition has been reported for this meteorite [10]. Enrichments of alkali and alkaline-earth elements could produce higher concentrations of cosmogenic Ne and Ar. Enrichments of minor elements such as K and Br in the fine grains are suggested by higher concentrations of radiogenic ^{40}Ar and the neutron-produced Kr isotopes in the present sample.

The K-Ar age was calculated from the radiogenic ^{40}Ar given in Table 2 and the potassium concentration given in Table 3. No correction was made for cosmogenic and trapped components to determine the amount of radiogenic ^{40}Ar. The K-Ar age obtained for Yamato-74191 is typical for hypersthene chondrites and shows that this meteorite did not suffer recent metamorphism.

Table 3. Chemical composition of Yamato-74191.

Element	Concentration (wt %)	Remarks
Na	0.72	a
Mg	15.01	a
Al	1.53	a
Si	28.08	a
S	1.83	a
Cl	223. ppm	b
K	0.091	c
	0.11	a
Ca	1.28	a
Ti	0.14	a
Mn	0.27	a
	0.29	d
Cr	0.51	a
Fe	20.25	a
	19.3	d
Ni	0.85	a
	1.05	d
Br	11.2 ppm	b

a Ikeda and Takeda [10],
b Nito, S., unpublished data,
c Nagao and Takaoka [2],
d Nishiizumi et al. [14].

3.2. *Neutron-capture effects on rare gas isotopes*

As given in Table 1, the isotopic ratios $^{80}Kr/^{84}Kr$, $^{82}Kr/^{84}Kr$ and $^{128}Xe/^{132}Xe$, are significantly higher than those of AVCC-Kr and Xe. These ex-

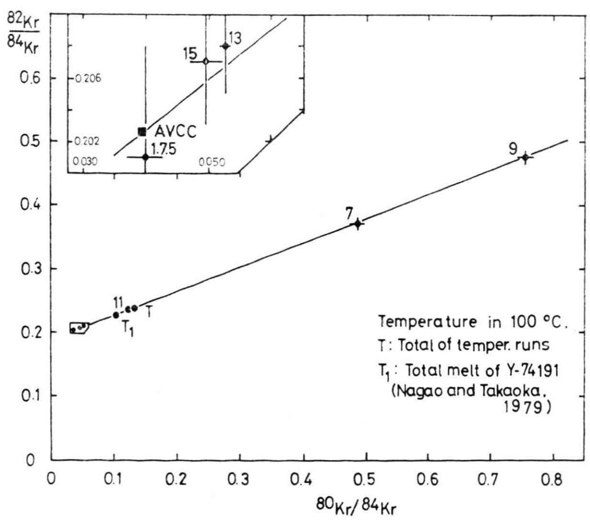

Fig. 2. Correlation diagram between $^{80}Kr/^{84}Kr$ and $^{82}Kr/^{84}Kr$. Data define a correlation line passing AVCC-Kr.

cesses could be attributed to the epithermal neutron capture on Br and I in the meteorite [2]. Figure 2 shows a correlation diagram between $^{80}Kr/^{84}Kr$ and $^{82}Kr/^{84}Kr$. All data fall on a straight line. As following from the agreement in the isotopic ratios, except for ^{80}Kr and ^{82}Kr, between the present sample and AVCC-Kr, the spallogenic and fissiogenic contributions are so small that the sample may considered to be a two-component mixture consisting of trapped Kr and neutron-produced one. Enrichments of neutron-produced ^{80}Kr and ^{82}Kr are striking in the 900 °C fraction. From a slope of the correlation line, the ratio of ^{80}Kr-excess to ^{82}Kr-excess is given as 2.62. Correction for spallogenic Kr gives a slightly higher ratio of 2.66. This value agrees well with our previous result [2].

The Yamato-74191 chondrite contains a large amount of radiogenic ^{129}Xe which originated from beta-decay of extinct ^{129}I ($T_{1/2} = 17.2$ my). Figure 3 shows a correlation plot between $^{128}Xe/^{132}Xe$ and $^{129}Xe/^{132}Xe$ for this meteorite. Except a point at 700 °C, the data define a correlation line. The slope is 263 and the intercept at the $^{128}Xe/^{132}Xe$ ratio of AVCC-Xe is 1.12 ± 0.29. This value is regarded as the trapped $^{129}Xe/^{132}Xe$ ratio in this meteorite. Xenon released at 700 °C contains a contamination of atmospheric Xe. From the $^{130}Xe/^{132}Xe$ ratio, the ^{132}Xe found in the 700 °C fraction is estimated to be a mixture of 54% atmospheric Xe and 46% AVCC-Xe. After correction for the atmospheric contamination, the data fit the correlation line, as shown by an open circle in Figure 3. Figure 3 shows clearly that the ^{128}Xe-excess found in the Yamato-74191 chondrite was produced by neutron capture on I.

A neutron source pertinent to the present excesses of rare gas isotopes are cosmic-ray secondaries. Cosmic-ray produced neutrons have their mean energy around 4 MeV and are moderated by elastic collisions with atoms of chondritic constituents. Based on the ^{80}Kr-excess to ^{82}Kr-excess ratio, epithermal neutron reactions are considered to have dominated in this meteorite. However, the actual neutron energy was not confined in a narrow range but distributed from thermal to several times ten MeV because of the moderation of cosmic-ray secondary neutrons. Energetic neutrons could induce an (n, 2n) reaction on ^{127}I, which would produce ^{126}Xe. A threshold for this reaction is 9.2 MeV. Figure 4 shows a correlation between $^{124}Xe/^{132}Xe$ and $^{126}Xe/^{132}Xe$. The isotopes ^{124}Xe and ^{126}Xe can be produced by spallation of heavier elements such as Ba and rare-earth elements. From the slope of the correlation line, the excess ratio of ^{124}Xe to ^{126}Xe is 0.41. This ratio is lower than the ratio of spallogenic ^{124}Xe to ^{126}Xe (approximately 0.6) usually

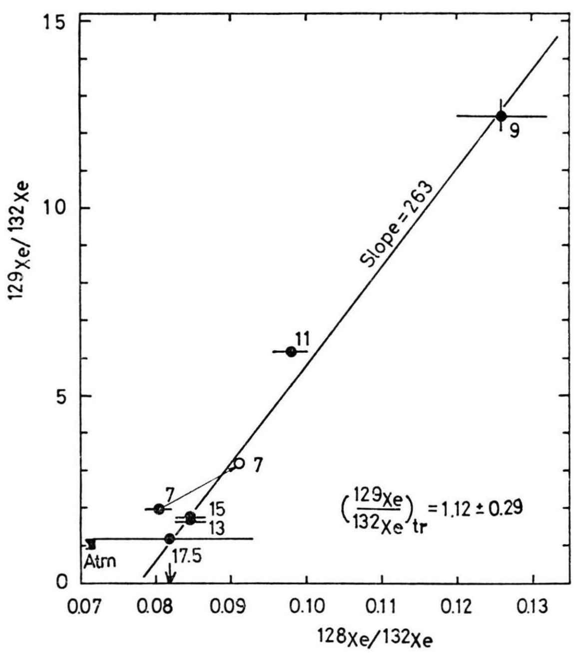

Fig. 3. Correlation diagram between $^{128}Xe/^{132}Xe$ and $^{129}Xe/^{132}Xe$ in Yamato-74191. After correction for atmospheric contamination, the 700 °C fraction is given by an open circle. Using the correlation line, the trapped $^{129}Xe/^{132}Xe$ ratio in Yamato-74191 was found to be 1.12 ± 0.29 by extrapolating to the $(^{128}Xe/^{132}Xe)_{AVCC} = 0.082$ point, which is shown by an arrow.

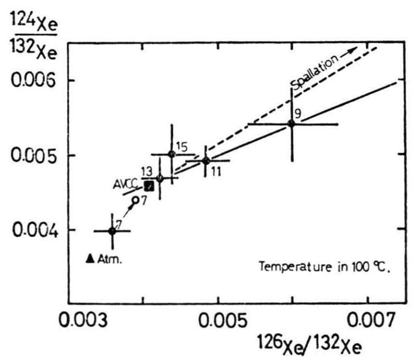

Fig. 4. Correlation plot of $^{124}Xe/^{132}Xe$ against $^{126}Xe/^{132}Xe$. The 700 °C fraction, corrected for atmospheric contamination, is given by an open circle.

found in stone meteorites. The effect of fissiogenic contributions to ^{132}Xe is to displace the observed point along the line passing through it and the origin. Judging from the isotopic composition determined, the fissiogenic contribution is negligible for ^{132}Xe. The production of ^{126}Xe by the $^{127}I(n, 2n\beta)$ ^{126}Xe reaction could give an answer to the ^{126}Xe excess in this meteorite. The occurence of ^{126}Xe excess produced by this reaction shows a contribution from high-energy cascade neutrons in the meteorite. No excess of ^{134}Xe, as shown in Table 1, suggests that the neutron energy in this meteorite was relatively high because thermal neutron capture on U would give a high fission yield at this isotope.

Table 4 presents the observed amounts of neutron-produced isotopes, theoretical production estimates and the neutron slowing-down density for the Yamato-74191 chondrite. Information for the neutron absorption cross sections of Cl, Br and I, and the neutron mean free path in chondrites were taken from Marti et al. [4]. The concentrations of Cl and Br are given in Table 3. Since no data on the iodine content in Yamato-74191 were available, we calculated from the $^{128}I/^{127}I$ ratio at the beginning of radiogenic ^{129}Xe retention by iodine-bearing minerals. The ratio is around 1×10^{-4} in many meteorites [11]. With it the radiogenic ^{129}Xe determined gives an iodine content of 170 ppb, which falls in a typical range of iodine contents for hypersthene chondrites [12]. The theoretical production of ^{128}Xe given in Table 4 was calculated for the epithermal neutron capture on I of this content.

Table 4. ^{80}Kr, ^{82}Kr, ^{128}Xe and ^{36}Ar excesses observed in Yamato-74191 and the theoretical production by epithermal neutron capture normalized to ^{80}Kr. Rare gas concentrations are given in units of 10^{-10} cm^3 STP/g.

Isotope	Excess observed		Theoretical production	
^{80}Kr	2.13[a]	1.40[b]	= 2.13	= 1.40
^{82}Kr	0.80[a]	0.52[b]	0.795	0.522
^{128}Xe	0.121[a]	0.078[b]	0.047	0.031
^{36}Ar	< 3300.[a]	< 3400.[b]	0.77	0.51
$^{80}Kr/^{82}Kr$	2.66[a]	2.69[b]	2.68	
Neutron slowing-down density q ($cm^{-3}\,sec^{-1}$)			0.17	0.11

[a] This work (0—147 μm).
[b] Yamato-74191 (47—417 μm); Nagao and Takaoka, [2].

3.3. Preatmospheric size and cosmic-ray irradiation history

Neutron moderation in meteorites depends on the shielding depth at the meteorite surface and its chemical composition. As discussed earlier, the ^{80}Kr- and ^{82}Kr-excesses and the ^{128}Xe-excess were produced by epithermal neutron capture on Br and I, respectively. According to Eberhardt et al. [13], neutrons with a mean energy of 3.7 MeV are moderated by elastic collision with atoms in chondrites. The reduction of the neutron energy from E_0 to E corresponds to a Fermi age of the neutron

$$\tau = \frac{\ln(E_0/E)}{3\,\xi\,\Sigma_{tot}\cdot\Sigma_{tr}}\,.$$

In our case $E_0 = 3.7$ MeV and $E = 165$ eV, the mean of 30 to 300 eV. ξ is the average logarithmic energy decrement per collision, Σ_{tot} the macroscopic total cross section, and Σ_{tr} the macroscopic transport cross section. For chondritic composition, $\xi\,\Sigma_{tot} = 0.0354\,cm^{-1}$ and $\Sigma_{tr} = 0.339\,cm^{-1}$ [13]. With these numerical values we find $\tau = 280\,cm^2$.

The slowing-down density q in a chondrite may be calculated by

$$q = [(^{80}Kr)_n/^{79}Br]\,[\xi\,\Sigma_{tot}/(R\,T_{21})],$$

where $R = 110$ barns, resonance integral for epithermal neutron capture, and T_{21} the cosmic-ray irradiation age. With Br = 11.2 ppm and $T_{21} = 8.3$ My, a mean value of two measurements, we have $q = 0.17$ and $0.11\,cm^{-3}\,sec^{-1}$ for $(^{80}Kr)_n = 2.13 \times 10^{-10}$ and 1.40×10^{-10} cm^3 STP/g, respectively. With these values and the Fermi age calculated earlier, we find the minimum mass and radius 470 kg and 32 cm on the assumption of a spherical meteorite for the Yamato-74191 chondrite.

Recently Nishiizumi et al. [14] have reported a high saturation activity of ^{53}Mn in Yamato-74191 relative to the average value of other meteorites. They assigned this apparently high value to an underestimation of the rare gas cosmic-ray irradiation age. The underestimation of the irradiation age could be caused by an erroneous estimation of various effects on the production rate such as the target chemical composition and shielding depth, and by a complex irradiation history of the meteorite. However, a recalculation using the newly determined ^{21}Ne age and the ^{53}Mn content in Yamato-74191 reduces the saturation activity of ^{53}Mn to 571 dpm/kg Fe. This value is equal to the theoretical content of ^{53}Mn estimated for a spherical meteorite of 30 cm preatmospheric radius, as determined by Nishiizumi et al. [14]. The preatmospheric radius of 30 cm is in a good agreement with our minimum radius for this meteorite. The Yamato-74191 meteorite was apparrently not so small in space as is supposed from the recovered mass.

The $^{22}Ne/^{21}Ne$ ratio of cosmogenic Ne in Yamato-74191 is 1.14, a high value suggesting irradiation at shallow depth. However, Smith, and Huneke [15] have showed that cosmic-ray produced Ne in Na-rich oligoclase feldspar can be characterized by a $^{22}Ne/^{21}Ne$ ratio as high as 1.50. They give a high cosmogenic $^{22}Ne/^{21}Ne$ production ratio in Na of 2.9 and assign it to indirect production of ^{22}Ne from ^{23}Na via ^{22}Na. Low energy protons take part in this production. Cosmic-ray irradiation of Si and S also produces a higher $^{22}Ne/^{21}Ne$ ratio than 1.2 [16]. Since the ^{21}Ne production rate in Na is larger than those in Si and S, a variation of Na concentration could more sensitively affect the $^{22}Ne/^{21}Ne$ ratio in the meteorite. From the $^{22}Ne/^{21}Ne$ ratio, the Ne found at 700 °C and 900 °C is regarded to contain the cosmogenic component from Na, while Ne at 1500 °C consists mostly of the cosmogenic component from Mg and Fe because of the low $^{22}Ne/^{21}Ne$ ratio and the high release temperature. The bulk composition of Na is 0.72 per cent as given in Table 3. However, the Na concentration has been revealed to be variable from 10 per cent in glass to 0.02 per cent in chondrules from Yamato-74191 [10]. The enrichment of ^{22}Ne could be enhanced by cosmic-ray secondary neutrons through $^{23}Na(n, 2n)^{22}Na$ ($Q = -12.4$ MeV) and $^{25}Mg(n, \alpha)^{22}Ne$ ($Q = +0.48$ MeV). The latter reaction that is an exothermic process is unique in that neutrons with lower energy than one MeV could produce ^{22}Ne. Therefore a high $^{22}Ne/^{21}Ne$ ratio is not necessarily incompatible with a large preatmospheric size of Yamato-74191 chondrite.

4. Conclusions

(1) The unequilibrated hypersthene chondrite Yamato-74191 was studied mass spectrometrically for rare gases released at various temperatures. Cosmogenic gases dominate in He and Ne. The meteorite contains large amounts of radiogenic ^{40}Ar and ^{129}Xe, and trapped Ar, Kr and Xe.

(2) In addition to the spallogenic component of Kr and Xe, isotopic excesses produced by neutron capture on Br and I were found for ^{80}Kr, ^{82}Kr and ^{128}Xe. A large part of the neutron-produced ^{80}Kr and ^{82}Kr was released at 700 °C and 1100 °C, while trapped ^{84}Kr was dominately released at 1300 °C. In contrast to the poor retentivity for the neutron-produced Kr isotopes in Br-bearing minerals, the ^{128}Xe-excess and radiogenic ^{129}Xe were released mainly at 1100 °C, while trapped ^{132}Xe was released at 1300 °C. From a correlation plot between $^{128}Xe/^{132}Xe$ and $^{129}Xe/^{132}Xe$, the trapped $^{129}Xe/^{132}Xe$ ratio was determined to be 1.12 ± 0.29. A small excess of ^{126}Xe was attributable to the $^{127}I(n, 2n\beta)^{126}Xe$ reaction ($Q = -9.2$ MeV) by high-energy neutrons.

(3) With neutron-produced ^{80}Kr, the neutron slowing-down density was estimated to be 0.14 ± 0.03 using the method of Eberhardt et al. [13]. A minimum mass and preatmospheric radius were estimated as 470 kg and 32 cm, respectively. The preatmospheric radius estimated here is in good agreement with that found with the ^{53}Mn saturation activity. Apparently the Yamato-74191 chondrite was not so small in space as supposed by the recovered mass.

Acknowledgements

The authors are greatly indebted to Prof. T. Nagata and Prof. K. Yanai of the National Institute of Polar Research for the meteorite sample used in this work. They thank Prof. M. Honda and Dr. S. Nito for invaluable comments and discussion and for the permission to use unpublished data on the Cl and Br contents. This work was supported in part by the Fund for Scientific Research of the Ministery of Education, grant number 339024.

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