NEUTRON CAPTURE EFFECTS IN YAMATO-74191 AND RARE GAS COMPOSITION IN YAMATO-75258

Nobuo TAKAOKA

Department of Earth Sciences, Faculty of Science, Yamagata University, Yamagata 990

and

Keisuke NAGAO

Graduate School, Science University of Okayama, Ridai-cho 1–1, Okayama 700

***Abstract*:** Results are summarized of mass spectrometrical measurements of rare gases in the Yamato-74191 (L3) chondrite.

The rare gas compositions in the Yamato-75258 amphoterite (type 6) were measured. Spallogenic gases dominate in He and Ne. Cosmic-ray irradiation and gas retention ages were tentatively calculated to be 12.4 Ma and 30 Ga, respectively. The concentration of trapped ^{132}Xe agrees with that inferred from the petrologic type.

1. Introduction

The unequilibrated hypersthene chondrite Yamato-74191 contains large amounts of neutron-produced ^{80}Kr and ^{82}Kr isotopes, attributable to epithermal neutron-capture on Br. Small excesses at ^{126}Xe and ^{128}Xe have been found, and the ^{128}Xe-excess was also attributed to the neutron-capture on I. However, the origin of the ^{126}Xe-excess was not obvious in our previous report (NAGAO and TAKAOKA, 1979).

This chondrite is unique in containing the highest concentrations of the neutron-produced Kr isotopes so far determined in chondrites (*e.g.* EUGSTER *et al.*, 1969). So we measured mass spectrometrically the rare gas compositions released at various temperatures to determine the neutron-capture effects on rare gas isotopes. Neutrons in a meteorite are produced by cosmic-ray irradiation and decelerated by collisions with atoms of meteoritic constituent. Thus the total intensity and the energy distribution of neutrons in the meteorite give a constraint on models for the cosmic-ray irradiation history and information about the preatmospheric size of the meteorite.

The Yamato-75258 meteorite is an amphoterite (LL6) chondrite. It is brittle breccia with many crumbs (YANAI, 1979). These features suggest that the chondrite may be gas-rich. So we studied the rare gas composition in this meteorite.

2. Results and Discussion

A grain-size fraction finer than 147 μm (100 mesh) of Yamato-74191 was wrapped with thin Al foil (27.5 mg) and degassed at about 80°C for a night. The sample was heated in a thoroughly degassed Mo crucible at successively higher temperatures of 700, 900, 1100, 1300, 1500 and 1750°C for 25 min. For the Yamato-75258 chondrite, a powder sample finer than 200 mesh (72.0 mg) was melted at 1750°C. Details of experimental techniques have been described elsewhere (TAKAOKA, 1976; NAGAO and TAKAOKA, 1979; TAKAOKA and NAGAO, 1980).

2.1. Neutron-capture effects in the Yamato-74191 meteorite

Full data of the temperature experiment and results for the neutron-capture effects of Yamato-74191 have been given by TAKAOKA and NAGAO (1980). This paper discusses a few points not given in detail earlier and presents a brief survey of the results for the neutron-capture effects on the Kr and Xe isotopes for convenience of ready reference.

Fig. 1 shows release patterns of rare gas components. Spallogenic ^{3}He was

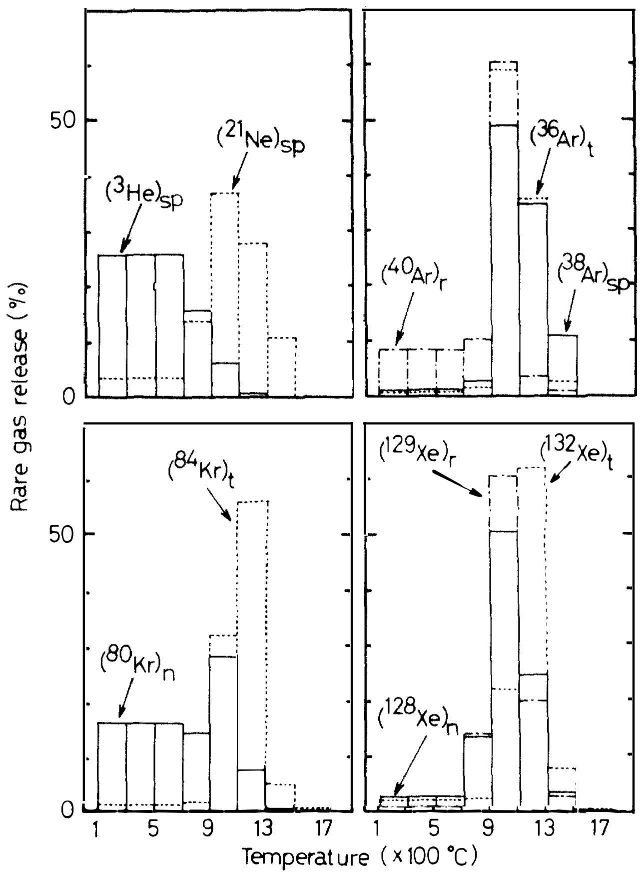

Fig. 1. Release patterns of rare gas components in Yamato-74191.

released mostly at 700°C, whereas most of spallogenic ^{21}Ne appeared in the temperature fractions higher than 1100°C. In He no trapped component was found and the radiogenic and spallogenic gases dominate. Small amounts of trapped Ne appeared in the 700, 1100 and 1300°C fractions. Trapped Ne (4×10^{-9} cc/g) at 700°C is not due to atmospheric contamination because the $^{38}Ar/^{36}Ar$ ratio indicates no atmospheric Ar and the $^{20}Ne/^{36}Ar$ ratio is higher than that in the atmosphere. Release of trapped Ne (1×10^{-9} cc/g) at 1100 and 1300°C seems to correlate with release of spallogenic ^{21}Ne at these temperatures, and may be due to a target mineral for spallogenic Ne, such as pyroxene.

The trapped component was found in heavy rare gases. Release patterns of trapped, spallogenic and radiogenic components of Ar have their peaks at 1100°C. The release of radiogenic ^{40}Ar was enhanced at lower temperatures compared with the spallogenic and trapped components. The release patterns are similar between spallogenic ^{21}Ne and ^{38}Ar, and trapped ^{36}Ar. This reflects similar retentivity of target minerals for spallogenic Ne and Ar. A contrast of low Ne and higher Ar in the trapped component means, therefore, an inefficient trapping mechanism for Ne in the ambient solar nebula, such as physical adsorption at low temperature and/or poor Ne-retentivity in the host phase of trapped gas, or that the host phase of trapped gas is different from the target minerals for spallogenic Ne and Ar.

The release pattern of neutron-produced ^{80}Kr shows two peaks at 700 and 1100°C, while trapped ^{84}Kr gives a single peak at 1300°C. In contrast, the release patterns of neutron-produced ^{128}Xe and radiogenic ^{129}Xe, both resembling each other, have a single peak at 1100°C, and are different from the pattern of trapped ^{132}Xe which was degassed mostly at 1300°C. We find here a significant difference in the

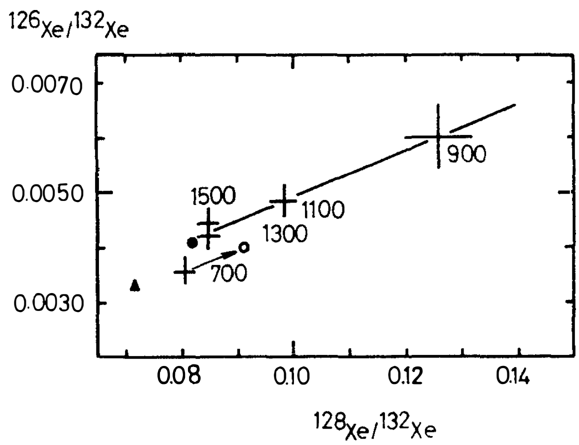

Fig. 2. Correlation plot between $^{126}Xe/^{132}Xe$ and $^{128}Xe/^{132}Xe$. Open circle: the 700°C fraction after correction for atmospheric contamination; solid circle: AVCC-Xe; solid triangle: atmospheric Xe.

rare gas retentivity between Br- and I-bearing minerals.

To the ^{126}Xe-excess unexplained in the previous report (NAGAO and TAKAOKA, 1979), the production of ^{126}Xe through the ^{127}I $(n, 2n\beta)$ ^{126}Xe reaction could give an answer. Fig. 2 shows a correlation plot between ^{126}Xe/^{132}Xe and ^{128}Xe/^{132}Xe. Judging from the isotopic composition, the fissiogenic contribution is negligible at ^{132}Xe. Therefore, the correlation in Fig. 2 is not owing to the contribution of fissiogenic Xe, but to the ^{126}Xe contribution correlating with the ^{128}Xe-excess. This supports the above suggestion that the ^{126}Xe-excess originated from I by the $(n, 2n)$ reaction. The production of ^{126}Xe from ^{127}I via the $(n, 2n)$ reaction has been reported for Bjurböle chondrules irradiated by a hard neutron flux in a reactor (SRINIVASAN *et al.*, 1972). A threshold for this reaction is 9.2 MeV. Because of the moderation of cosmic-ray secondary neutrons, the neutron energy is supposed to be distributed from thermal to several times ten MeV. Thus energetic neutrons could induce the $(n, 2n)$ reaction on ^{127}I.

From the isotopic data of temperature experiment on Kr and Xe in Yamato-

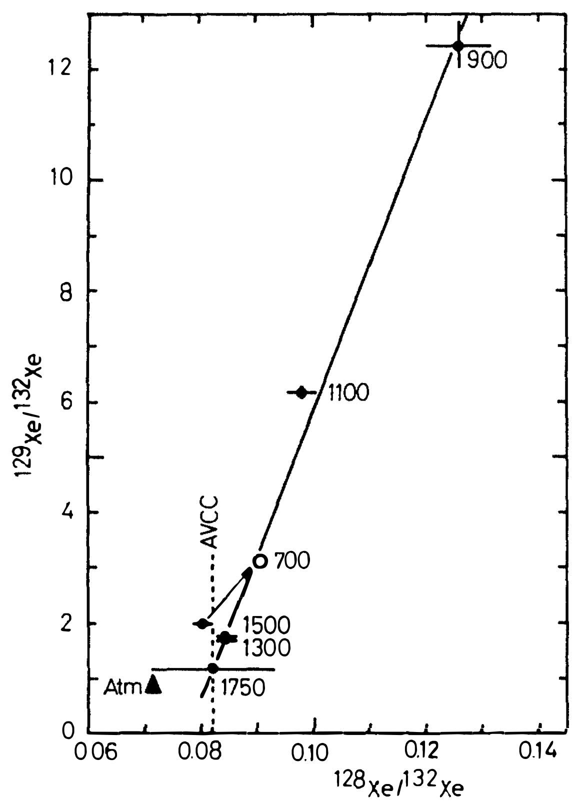

Fig. 3. Correlation diagram between $^{128}Xe/^{132}Xe$ and $^{129}Xe/^{132}Xe$. The 700°C fraction after correction for atmospheric contamination is given by an open circle. By extrapolating the correlation line to the $(^{128}Xe/^{132}Xe)_{AVCC}$=0.082 point, the trapped $^{129}Xe/^{132}Xe$ ratio in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

74191, the following resulted (TAKAOKA and NAGAO, 1980). As judged from the agreement in the isotopic ratio, except for ^{80}Kr and ^{82}Kr, between the present sample and AVCC-Kr (EUGSTER *et al.*, 1967), spallogenic and fissiogenic contributions were so small that we could consider a two-component mixture of trapped Kr and neutron-produced one. The production ratio of neutron-produced ^{80}Kr and ^{82}Kr was estimated to be 2.66, which agrees well with our previous result (NAGAO and TAKAOKA, 1979). This ratio agrees with the theoretical value for products of the epithermal neutron-capture on Br.

As given in Fig. 3, a correlation plot shows clearly that the ^{128}Xe-excess in Yamato-74191 was produced by the neutron-capture on I because the ^{129}Xe-excess originated from beta-decay of extinct ^{129}I ($T_{1/2}=17$ Ma). From the intercept extrapolated at the ^{128}Xe/^{132}Xe ratio of AVCC-Xe (EUGSTER *et al.*, 1967), the trapped ^{129}Xe/^{132}Xe ratio in this meteorite is estimated to be 1.12 ± 0.29.

The neutron moderation in meteorites depends on the shielding depth at the surface and its chemical composition. According to EBERHARDT *et al.* (1963), the reduction of neutron energy from E_0 to E corresponds to a Fermi age of the neutron

$$\tau = \ln (E_0/E)/3\xi \textstyle\sum_{tot} \cdot \sum_{tr}.$$

The slowing-down density q in a chondrite is calculated by

$$q = [(^{80}\mathrm{Kr})_n/^{79}\mathrm{Br}] \cdot [\xi \textstyle\sum_{tot}/R \cdot T].$$

In the present case, $E_0=3.7$ MeV, a mean neutron energy produced in meteorites by cosmic-ray irradiation, and $E=165$ eV, a mean of 30 to 300 eV. ξ is the average logarithmic energy decrement per collision, $\sum_{tot}$ the macroscopic total cross section, and $\sum_{tr}$ the macroscopic transport cross section. For the chondritic composition, $\xi\sum_{tot}=0.0354\ \mathrm{cm}^{-1}$ and $\sum_{tr}=0.339\ \mathrm{cm}^{-1}$. $R=110$ barns (MARTI *et al.*, 1966), the resonance integral for epithermal neutron capture on ^{79}Br, and $T=8.3$ Ma, the cosmic-ray irradiation age. The Br content amounts to 11.2 ppm (O. NITO, unpublished). With these numerical values, we have $\tau=280\ \mathrm{cm}^2$ for the Fermi age of neutron and $q=0.14\ \mathrm{cm}^{-3}\ \mathrm{s}^{-1}$ for the slowing-down density for $(^{80}\mathrm{Kr})_n=1.77\times 10^{-10}\ \mathrm{cm}^3$ STP/g. They give a minimum radius 32 cm on the assumption of a spherical meteoroid. So this meteorite was not as small in space as supposed from the recovered mass of 1.092 kg.

2.2. *Rare gas composition in the Yamato-75258 (LL6) chondrite and its cosmic-ray irradiation and gas retention ages*

The concentrations of five rare gas isotopes and the isotopic ratios of He, Ne and Ar are given in Table 1. In He and Ne, the trapped gases are completely lost and the spallogenic and radiogenic components are predominant. Argon is a mixture of three components, as judged from the isotopic composition. ^{132}Xe is regarded as mostly trapped on the isotopic composition of Xe which is not shown in Table 1.

Table 1. Rare gas composition in Yamato-75258. Rare gas concentration is given in unit of 10^{-8} cm^3 STP/g.

Isotope	Yamato-75258
^{3}He	30.1
^{4}He	1010
$^{3}He/^{4}He$	0.0298 ± 0.0005
^{21}Ne	5.76
$^{20}Ne/^{21}Ne$	0.947 ± 0.006
$^{22}Ne/^{21}Ne$	1.134 ± 0.006
^{36}Ar	0.668
$^{38}Ar/^{36}Ar$	0.699 ± 0.021
$^{40}Ar/^{36}Ar$	3935 ± 36
^{84}Kr	0.011
^{132}Xe	0.012

Table 2. Spallogenic ^{3}He, ^{21}Ne and ^{38}Ar, and cosmic-ray irradiation age of Yamato-75258.

Sample	Yamato-75258	Unit
$(^{3}He)_{sp}$	30.1	10^{-8}cm^{3}STP/g
$(^{21}Ne)_{sp}$	5.76	
$(^{38}Ar)_{sp}$	0.388	
$(^{3}He/^{21}Ne)_{sp}$	5.23	
$(^{21}Ne/^{38}Ar)_{sp}$	14.9	
P_3	2.48	10^{-8}cm^{3}STP/gMa
P_{21}	0.466 (0.478)[a]	
P_{38}	0.0586 (0.0649)[a]	
P_3/P_{21}	5.32 (5.19)[a]	
P_{21}/P_{38}	7.95 (7.37)[a]	
T_3	12.1	Ma
T_{21}	12.4 (12.1)[a]	
T_{38}	6.62 (5.98)[a]	

a: Production rates and cosmic-ray irradiation age calculated by BOGARD and CRESSY (1973) are given in parentheses.

The concentration of trapped ^{132}Xe agrees with that inferred from the petrologic type (MARTI, 1967).

Table 2 shows the concentrations of spallogenic ^{3}He, ^{21}Ne and ^{38}Ar, and the cosmic-ray irradiation age. No correction for the trapped component was applied to ^{3}He and ^{21}Ne, but the trapped component was corrected at ^{38}Ar. The production rates of spallogenic isotopes were calculated after HERZOG and ANDERS (1971), STAUFFER (1962), and BOGARD and CRESSY (1973) with the chemical composition of Yamato-75258 (YANAI, 1979). They are also listed in Table 2. 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 spallogenic gases determined. The agreement is good between P_3/P_{21} and $^{3}He/^{21}Ne$, which suggests little loss of ^{3}He. But there is a large discrepancy between P_{21}/P_{38} and $^{21}Ne/^{38}Ar$. This is due to the low spallogenic ^{38}Ar concentration in this sample. Since the Ar retention in meteoritic matter is better than the He retention, a diffusive loss of spallogenic Ar is out of consideration, and the low spallogenic ^{38}Ar should be attributed to the reduced production of Ar. The chemical inhomogeneity is one reason for the low ^{38}Ar production. In the cosmic-ray spallation reaction, the Ar production is enhanced from K and Ca. The K content of 700 ppm is reasonable to produce the radiogenic ^{40}Ar determined. With the typical target chemistry for amphoterite, Ca and Fe produce most of spallogenic Ar to the same extent. The Yamato-75258 chondrite is breccia with large gray rounded and irregular clasts in a lighter yellowish-gray matrix (YANAI, 1979). The target with low contents of Ca and Fe such as an aubritic composition could give a significantly high $^{21}Ne/^{38}Ar$ ratio, because of low contents of these elements. As shown in Fig. 4, high $(^{21}Ne/^{38}Ar)_{sp}$ ratios have been observed in amphoterite chon-

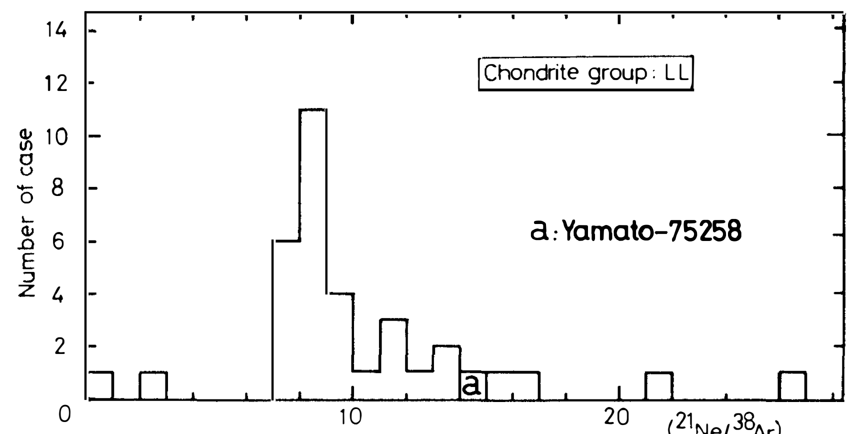

Fig. 4. Spallogenic $^{21}Ne/^{38}Ar$ ratio distribution in amphoterite chondrites. The ratios were calculated on data given by SCHULTZ and KRUSE (1978). A cluster around 8 agrees with the theoretical production ratio calculated with the target chemistry typical of amphoterites. Yamato-75258 is found in a tail to the higher side of $(^{21}Ne/^{38}Ar)_{sp}$ ratio.

drites. The present sample seems to deviate from the chemical composition reported by YANAI (1979). Thus the cosmic-ray age given in Table 2 is regarded as tentative. The aubritic composition would result in a higher production rate for ^{21}Ne and therefore the cosmic-ray age calculated with ^{21}Ne would be reduced to about 10 Ma. However, in any case, the cosmic-ray age of this chondrite falls in a cluster around 10 Ma for amphoterite chondrites, as shown by WASSON (1974).

The U/Th-He and K-Ar ages were calculated to be 2.7 and 3.0 Ga, respectively. For calculation of the gas retention ages, U = 12.6 ppb and Th/U = 3.81 (MORGAN, 1971), and K = 700 ppm (YANAI, 1979) were assumed.

Acknowledgments

The authors are greatly indebted to Prof. T. NAGATA and Dr. K. YANAI for the meteorite samples. They thank Prof. M. HONDA and Dr. O. NITO for invaluable comments and discussion, and for the permission to use unpublished data on the Br content, respectively. This work was supported in part by the Grant in Aid for Scientific Research of the Ministry of Education, Science and Culture, grant No. 339024.

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.

EBERHARDT, P., GEISS, J. and LUTZ, H. (1963): Neutrons in meteorites. Earth Science and Meteoritics, comp. by J. GEISS and E. D. GOLDBERG. Amsterdam, North-Holland, 143.

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.

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

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

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

MORGAN, J. W. (1971): Thorium and uranium. Handbook of Elemental Abundances in Meterorites, ed. by B. MASON. New York, Gordon and Breach, 517 and 529.

NAGAO, K. and TAKAOKA, N. (1979): Rare gas studies of Antarctic meteorites. Mem. Natl Inst. Polar Res., Spec. Issue, **12**, 207–222.

SCHULTZ, L. and KRUSE, H. (1978): Light noble gases in stony meteorites, a compilation. Nucl. Track Detect., **2**, 65–103.

SRINIVASAN, B., ALEXANDER, E. C., JR. and MANUEL, O. K. (1972): Radiation effects in ^{129}I–^{129}Xe dating of Bjurböle chondrules by neutron irradiation. Icarus, **16**, 571–576.

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

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

TAKAOKA, N. and NAGAO, K. (1980): Mass spectrometrical study of rare gas compositions and neutron capture effects in Yamato-74191 (L3) chondrite. Z. Naturforsch., **35a**, 29–36.

WASSON, J. T. (1974): Meteorites. Berlin, Springer-Verlag, 128.

YANAI, K. comp. (1979): Catalog of Yamato Meteorites. First edition. Tokyo, Natl Inst. Polar Res., 170 and 188.

(Received May 14, 1980; Revised manuscript received July 21, 1980)