

PRIMORDIAL RARE GASES IN BELGICA-7904 (C2) CARBONACEOUS CHONDRITE

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Abstract: Belgica-7904(C2) carbonaceous chondrite was analyzed for rare gas isotopic compositions with stepwise heating extraction method. Primordial rare gases are dominant, whereas the concentrations of radiogenic ^4He and ^{40}Ar are lower than those to be expected from radioactive decay. Cosmic-ray exposure age of 0.7 my was obtained with cosmogenic ^{21}Ne . Ne-E component was found in a 900–1100°C range and its concentration was $1.2 \times 10^{-9} \text{ cm}^3\text{STP/g}$. Excessive heavy Xe isotopes in a 900°C fraction have CCF-Xe spectrum. ^{20}Ne , ^{84}Kr and ^{132}Xe in the highest temperature fraction of 1800°C are enriched. The enrichments may be due to a stellar nucleosynthesis or due to a neutron irradiation of high temperature condensates.

1. Introduction

Though carbonaceous chondrites are in a minor group among the meteorites, they are regarded to have primitive elemental and isotopic compositions and to be important for the investigation of an early stage of the solar system. In rare gas studies on the carbonaceous chondrites with the techniques of stepwise heating, mineral separation and chemical leaching, it has been proved that they are composed of matters of various origin which are different in rare gas concentrations, relative elemental abundances and isotopic ratios. To get a consistent explanation for complicated rare gas data, more data on rare gas compositions for primitive meteorites are requested.

Many carbonaceous chondrites were collected by a great effort of the teams of Japanese Antarctic Research Expedition. Among the antarctic carbonaceous chondrites, we analyzed the Belgica-7904 C2 chondrite for all rare gas isotopic ratios with a conventional stepwise heating technique. These results will give us fundamental data useful for further investigation on this meteorite.

2. Sample Description and Experimental Procedure

The Belgica-7904 carbonaceous chondrite was recovered by YANAI *et al.* in December 19, 1979 at Belgica Mountains, Antarctica. It consists of abundant dark phyllosilicate matrix, chondrules and inclusions (NATIONAL INSTITUTE OF POLAR RESEARCH, 1982). The meteorite is classified as CM2 (type 2 carbonaceous chondrite of Mighei group) and is moderately altered (KOJIMA *et al.*, 1984).

The sample used in this work was of dark matrix and was less weathered. Several

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small pieces weighing 0.1696 g were separated from a block and were used for rare gas mass spectrometry. The pieces were wrapped with Al-foil 10 μm thick and loaded in a sample holder of a rare gas extraction and purification system. The system was baked out and the sample was heated in vacuum at about 150°C overnight to reduce an atmospheric contamination. Rare gases were extracted by stepwisely heating at temperatures of 700, 900, 1100, 1300, 1500 and 1800 °C. The evolved gases were purified by exposing them on Ti-Zr getter heated at about 800 °C and separated into three fractions of He and Ne, Ar, and Kr and Xe by adsorption and desorption with a charcoal trap. A mass spectrometer used in this work was of a single focusing 90° sector type with 30 cm radius of ion curvature and was made of stainless steel. It was equipped with an electron bombardment ion source and a single collector with a secondary electron multiplier. Resolving power of the mass spectrometer was adjusted to about 1500. Sensitivity and mass discrimination of the mass spectrometer were determined by analyzing known amounts of atmospheric rare gases with the same procedure as that applied to the sample analysis.

3. Results and Discussion

3.1. Depletion of radiogenic ^4He and ^{40}Ar , and of light rare gases

Rare gas isotopic compositions and concentrations of six temperature fractions are listed in Tables 1, 2 and 3. The total ^4He and ^{40}Ar concentrations are 0.748×10^{-5} and $0.121 \times 10^{-5} \text{ cm}^3\text{STP/g}$, respectively. If the mean values of U and K concentrations for C2 chondrites (MORGAN, 1971; GOLES, 1971) are assumed, ^4He and ^{40}Ar concentrations

Table 1. Concentrations and isotopic compositions of He, Ne and Ar.

Temp. (°C)	$^4\text{He}^*$	$^3\text{He}/^4\text{He}$ ($\times 10^{-4}$)	$^{20}\text{Ne}^*$	$^{20}\text{Ne}/^{22}\text{Ne}$	$^{21}\text{Ne}/^{22}\text{Ne}$	$^{36}\text{Ar}^*$	$^{38}\text{Ar}/^{36}\text{Ar}$	$^{40}\text{Ar}/^{36}\text{Ar}$
700	20.9	277 ± 6	0.216	2.98 ± 0.06	0.703 ± 0.010	1.84	0.1877 ± 0.0037	16.00 ± 0.42
900	285	5.84 ± 0.14	2.21	6.78 ± 0.08	0.234 ± 0.003	2.90	0.1937 ± 0.0038	0.59 ± 0.27
1100	353	2.17 ± 0.06	4.73	5.97 ± 0.05	0.135 ± 0.001	25.7	0.1884 ± 0.0036	2.82 ± 0.04
1300	82.0	3.46 ± 0.12	1.52	7.33 ± 0.07	0.157 ± 0.003	22.2	0.1884 ± 0.0036	0.45 ± 0.04
1500	6.96	11.5 ± 0.5	0.432	7.36 ± 0.08	0.386 ± 0.010	6.96	0.1916 ± 0.0037	0.52 ± 0.11
1800	0.17	64 ± 60	0.023	2.91 ± 0.08	0.845 ± 0.033	0.11	0.1901 ± 0.0043	35.1 ± 8.4
Total	748		9.13			59.7		

*Concentrations in unit of $10^{-8} \text{ cm}^3\text{STP/g}$.

Table 2. Kr isotopic composition.

Temp. (°C)	$^{84}\text{Kr}^*$	^{78}Kr	^{80}Kr	^{82}Kr	^{83}Kr	^{84}Kr	^{86}Kr
700	2.87	0.593 ± 0.023	4.04 ± 0.06	20.29 ± 0.12	20.21 ± 0.30	=100	30.66 ± 0.27
900	2.81	0.602 ± 0.027	3.95 ± 0.17	19.96 ± 0.40	20.30 ± 0.47	=100	30.91 ± 0.36
1100	25.1	0.617 ± 0.013	3.96 ± 0.04	20.30 ± 0.20	20.26 ± 0.16	=100	30.67 ± 0.26
1300	16.3	0.602 ± 0.019	3.94 ± 0.04	20.33 ± 0.16	20.18 ± 0.15	=100	30.77 ± 0.19
1500	5.77	0.615 ± 0.021	3.95 ± 0.14	20.44 ± 0.18	20.41 ± 0.12	=100	31.03 ± 0.28
1800	0.12	0.588 ± 0.068	3.87 ± 0.11	19.46 ± 0.48	19.67 ± 0.40	=100	29.70 ± 0.77

*Concentration in unit of $10^{-10} \text{ cm}^3\text{STP/g}$.

Table 3. Xe isotopic composition.

Temp. (°C)	¹³² Xe*	¹²⁴ Xe	¹²⁶ Xe	¹²⁸ Xe	¹²⁹ Xe	¹³⁰ Xe	¹³¹ Xe	¹³² Xe	¹³⁴ Xe	¹³⁶ Xe
700	6.66	0.362 ±0.008	0.349 ±0.009	7.270 ±0.060	97.41 ±0.65	15.35 ±0.11	78.88 ±0.68	=100	38.72 ±0.24	33.14 ±0.37
900	4.17	0.447 ±0.022	0.401 ±0.012	8.089 ±0.111	100.64 ±0.61	16.04 ±0.13	81.93 ±0.57	=100	40.59 ±0.11	35.10 ±0.28
1100	25.2	0.434 ±0.010	0.392 ±0.010	8.163 ±0.071	101.34 ±0.43	16.28 ±0.06	81.95 ±0.14	=100	38.75 ±0.21	32.88 ±0.20
1300	15.1	0.435 ±0.012	0.393 ±0.010	7.985 ±0.070	101.44 ±0.62	16.05 ±0.16	81.52 ±0.37	=100	38.37 ±0.10	32.47 ±0.13
1500	6.54	0.428 ±0.016	0.388 ±0.015	7.997 ±0.120	100.95 ±0.61	16.16 ±0.13	81.19 ±0.44	=100	38.32 ±0.16	32.66 ±0.22
1800	0.154	0.459 ±0.015	0.379 ±0.059	7.688 ±0.168	100.23 ±1.35	15.73 ±0.39	80.77 ±1.23	=100	38.97 ±0.86	31.80 ±0.73

*Concentration in unit of 10^{-10} cm³STP/g.

to be expected from radioactive decay of U, Th and ⁴⁰K in 4.5×10^9 years are about 2×10^{-5} and 3×10^{-5} cm³STP/g, respectively. Both of the concentrations observed are much lower than the estimated values. However, since the U, Th and K concentrations have not been measured, the depletions of radiogenic rare gases are not conclusive at present.

The ³He concentration is about a half of that of cosmogenic ³He expected from the exposure age of 0.7 my calculated from cosmogenic ²¹Ne. This indicates a partial loss of cosmogenic He. Some of the trapped He should be lost when the cosmogenic He was partially lost. ²⁰Ne is also slightly depleted compared with a typical value of ²⁰Ne concentration for C2 chondrites. In contrast to the depletions in radiogenic ⁴He and ⁴⁰Ar, and in light rare gases ³He and ²⁰Ne, trapped ³⁶Ar, ⁸⁴Kr and ¹³²Xe concentrations are in the range of concentrations for C2 chondrites reported (*e.g.* MAZOR *et al.*, 1970).

The loss of radiogenic rare gases may be caused by weathering of U-, Th- and K-bearing minerals of the meteorite. Cosmogenic and trapped rare gases in these minerals should be also lost. The other possible cause which can explain the depletion of both radiogenic and light rare gases is a moderate heating of the meteorite in the past.

3.2. Helium

The release patterns of ³He and ⁴He are shown in Fig. 1. Most of ³He was released at the lowest temperature of 700°C. About a half of total ⁴He was released in the 1100°C fraction and most of the remaining was released at 900 and 1300°C. Observed ³He/⁴He ratios for the 900, 1100 and 1300°C fractions are in the range of 10^{-4} , which is similar to that of trapped He. The high ³He/⁴He ratios in the 700 and 1800°C fractions are due to the spallation component with a high ³He/⁴He ratio.

3.3. Neon

A three-isotope plot of Ne for all temperature fractions is shown in Fig. 2. Ne for the 700°C fraction is mostly cosmogenic. The isotopic ratios for three temperature fractions of 700, 900 and 1300°C lie along a mixing line between spallation Ne (Ne-S) and planetary Ne (Ne-A). A point for the 1500°C fraction is along a mixing line con-

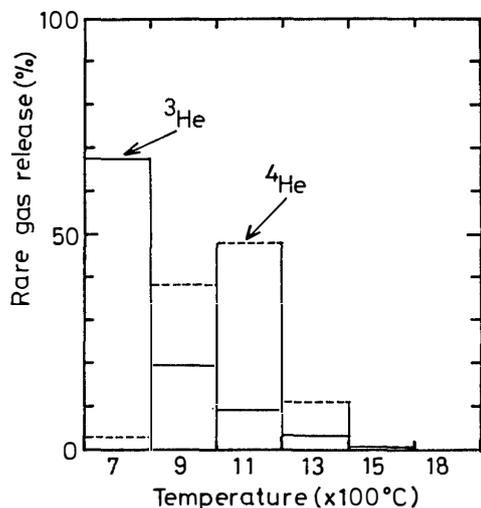


Fig. 1. The release patterns of ^3He and ^4He . The total concentrations of ^3He and ^4He are 0.859 and 748 in unit of $10^{-8} \text{ cm}^3\text{STP/g}$, respectively. $^3\text{He}/^4\text{He}$ ratios for the 900, 1100 and 1300°C fractions are in the range of 10^{-4} .

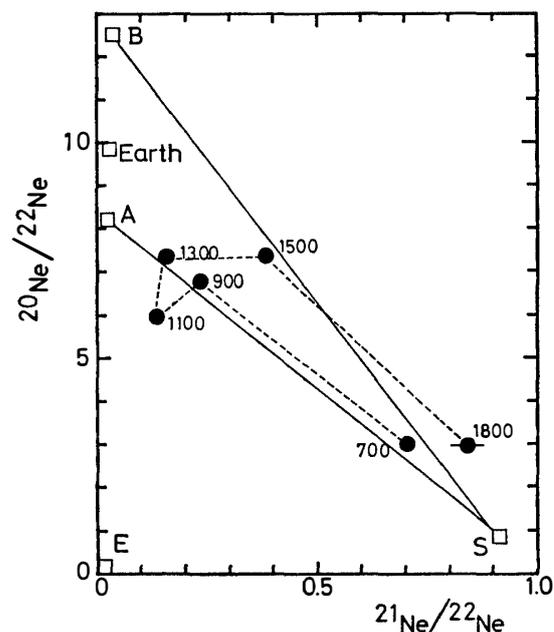


Fig. 2. Three isotope plot of Ne. Ne-E is released in the 1100°C fraction. $^{20}\text{Ne}/^{22}\text{Ne}$ ratio in the 1800°C fraction is higher than that of Ne-S.

necting spallation Ne and solar Ne (Ne-B). Those four points are in the triangular region formed by three components S, A and B.

The isotopic ratios for the 1100°C fraction are plotted below the mixing line between the Ne-S and Ne-A components. The ratios are presumably lowered by mixing of Ne-E, which is composed practically of pure ^{22}Ne (e.g. MEIER *et al.*, 1980). The release temperature for this fraction is consistent with the stepwise extraction data reported for other carbonaceous chondrites (BLACK and PEPIN, 1969). Usually Ne-E is released at 1000°C from the carbonaceous chondrites according to BLACK and PEPIN. In Belgica-7904 about a half of total ^{20}Ne is released in a temperature range between 900 and 1100°C. The amount of Ne-E in this fraction of Belgica-7904 is estimated to be $1.2 \times 10^{-9} \text{ cm}^3 \text{ STP/g}$ on the assumption of mixture of three components (A, E and S). This concentration of Ne-E is practically the same as that reported for carbonaceous chondrites by BLACK and PEPIN (1969), for which Ne-E amounts to $1.3 \times 10^{-9} \text{ cm}^3 \text{ STP/g}$ with ambiguity of a factor 2 at 1000°C. These strongly suggest the presence of Ne-E in Belgica-7904.

In Fig. 2, the $^{20}\text{Ne}/^{22}\text{Ne}$ ratio in the 1800°C fraction is exceptionally higher than the ratio of spallation Ne. Though the amount of Ne in this fraction used for the mass spectrometry was small ($4 \times 10^{-11} \text{ cm}^3 \text{ STP}$), the hindering ions ($^{40}\text{Ar}^{++}$, $\text{H}_2^{18}\text{O}^+$ and HF^+) were completely separated from ^{20}Ne . The peak height of CO_2^{++} ion calculated with empirically determined $\text{CO}_2^{++}/\text{CO}_2^+$ ratio and CO_2^+ peak height was 16% of the peak height of $M/e=22$ ion. Since the fluctuations of the $\text{CO}_2^{++}/\text{CO}_2^+$ ratio and of

the CO_2^+ peak height were small during the Ne analyses of the meteorite measurements, the high $^{20}\text{Ne}/^{22}\text{Ne}$ ratio is difficult to be attributed to an erroneous correction for CO_2^{++} at $M/e=22$. The ^{20}Ne excess may be produced by α capture of ^{16}O in stellar nucleosynthesis or by $^{19}\text{F}(n, \beta) ^{20}\text{Ne}$ reaction in the meteorite. We plan to re-examine the Ne isotopic ratio for this meteorite.

To know an exposure age of this meteorite a rough estimation of cosmogenic ^{21}Ne concentration was made. The cosmogenic ^{21}Ne concentration was calculated with the total concentrations for each isotope on the assumption of mixture of Ne-A ($^{20}\text{Ne}/^{22}\text{Ne}=8.2$, $^{21}\text{Ne}/^{22}\text{Ne}=0.024$), Ne-S ($^{20}\text{Ne}/^{22}\text{Ne}=0.85$, $^{21}\text{Ne}/^{22}\text{Ne}=0.92$) and Ne-E. The concentration of cosmogenic ^{21}Ne was $0.27 \times 10^{-8} \text{ cm}^3 \text{ STP/g}$, and the exposure age was estimated to be about 0.7 my with the production rate of $0.368 \times 10^{-8} \text{ cm}^3 \text{ STP/g/my}$ by CRESSY and BOGARD (1976). In this calculation the shielding effect was not considered. Carbonaceous chondrites classified as CM make a group with short exposure age distribution (MAZOR *et al.*, 1970). The short exposure age for the Belgica-7904 CM2 chondrite is also in the group of CM chondrites. However, the actual age might be somewhat longer than the age of 0.7 my because of the Ne loss mentioned in the previous section.

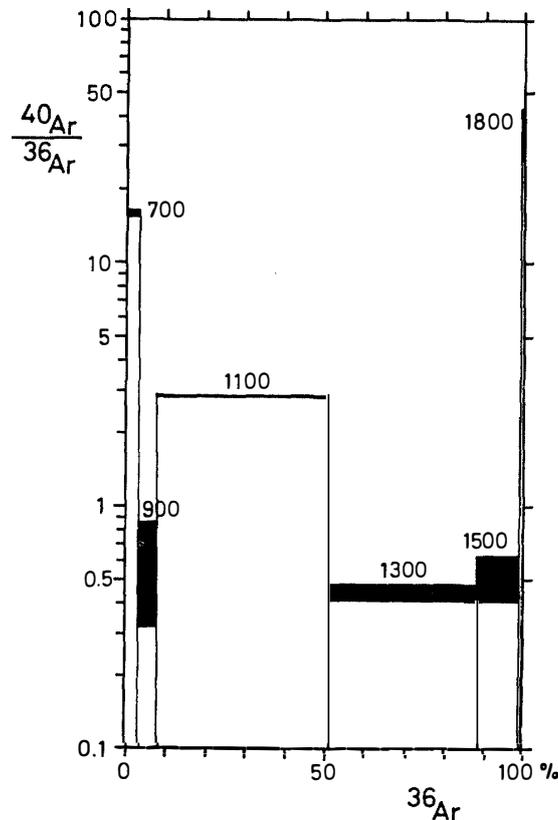


Fig. 3. The $^{40}\text{Ar}/^{36}\text{Ar}$ ratios are shown against the cumulative fractional release of ^{36}Ar . The shaded areas indicate the error of the $^{40}\text{Ar}/^{36}\text{Ar}$ ratios.

3.4. Argon

The $^{40}\text{Ar}/^{36}\text{Ar}$ ratios are shown in Fig. 3 against the cumulative fractional release of ^{36}Ar . The total trapped ^{36}Ar concentration is in the range of that for C2 chondrites.

The $^{40}\text{Ar}/^{36}\text{Ar}$ ratio in the 700°C fraction is relatively higher than the other temperature fractions except for the 1800°C fraction. The high ratio is due to the release of radiogenic ^{40}Ar loosely retained in K bearing minerals. However, since the amount of radiogenic ^{40}Ar is smaller than the expected value from radioactive decay of ^{40}K in 4.5×10^9 y with typical concentration of from 300 to 500 ppm for C2 chondrites (GOLES, 1971), the $^{40}\text{Ar}/^{36}\text{Ar}$ ratio should be higher than the observed value.

80% of total ^{36}Ar was released at the higher temperatures of 1100 and 1300°C and the $^{40}\text{Ar}/^{36}\text{Ar}$ ratios are 2.82 ± 0.04 and 0.45 ± 0.04 for 1100 and 1300°C, respectively. The low $^{40}\text{Ar}/^{36}\text{Ar}$ ratios in these fractions suggest that the primordial Ar with low $^{40}\text{Ar}/^{36}\text{Ar}$ of 10^{-4} (CAMERON, 1973) is prevailing in these fractions.

The $^{38}\text{Ar}/^{36}\text{Ar}$ ratios are a little higher than the atmospheric ratio of 0.187 (NIER, 1950). In the 900°C fraction both the high $^{38}\text{Ar}/^{36}\text{Ar}$ ratio of 0.1937 ± 0.0038 and the

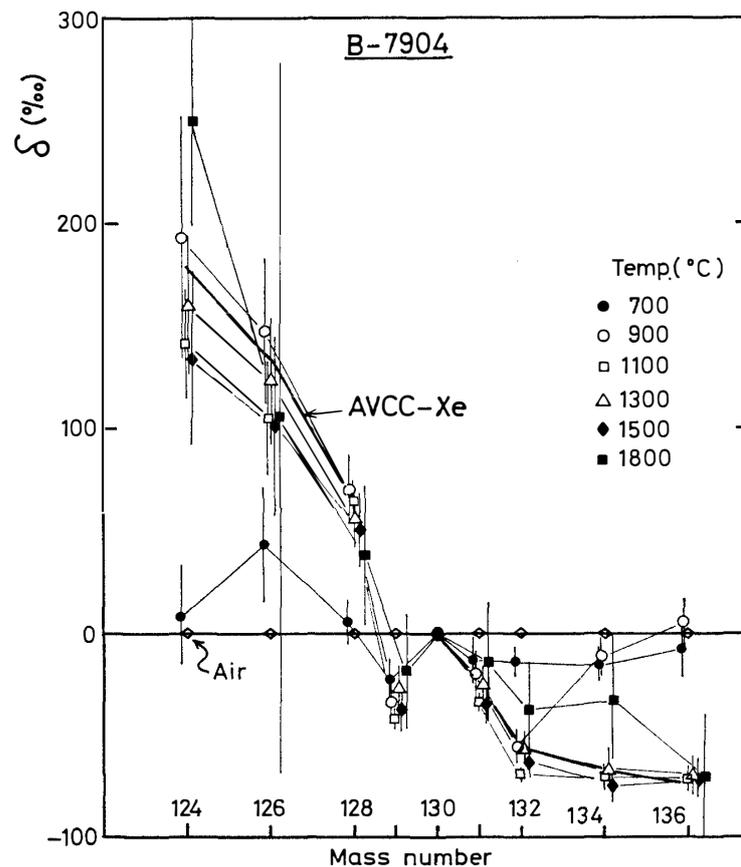


Fig. 4. Isotopic variations compared with atmospheric Xe are shown. δ is defined as:

$$\delta = [(^m\text{Xe}/^{130}\text{Xe})_s / (^m\text{Xe}/^{130}\text{Xe})_{\text{atm}} - 1] \times 10^3,$$

where subscripts "atm" and "s" mean atmospheric and sample, respectively. Isotopic composition of Xe in the 700°C fraction is similar to that of atmospheric Xe. Definite excesses of ^{134}Xe and ^{136}Xe are seen in the 900°C fraction.

high $^{136}\text{Xe}/^{132}\text{Xe}$ ratio are observed. The correlation between the $^{38}\text{Ar}/^{36}\text{Ar}$ and $^{136}\text{Xe}/^{132}\text{Xe}$ ratios was found in Allende etched residues by LEWIS *et al.* (1977). Though the correlation found in Belgica-7904 might be the same one presented by LEWIS *et al.*, we cannot abandon a possibility of cosmogenic ^{38}Ar for the high $^{38}\text{Ar}/^{36}\text{Ar}$ ratio in the 900°C fraction. The estimated abundance of cosmogenic ^{38}Ar from the exposure age of 0.7 my can produce the high $^{38}\text{Ar}/^{36}\text{Ar}$ ratio if more than half of the cosmogenic ^{38}Ar is released in the 900°C fraction.

3.5. Krypton and xenon

Xe isotopic compositions relative to the atmospheric ratios are shown in Fig. 4 with δ -values. The isotopic composition of Xe released at the lowest temperature of 700°C is similar to the atmospheric one which indicates that the Xe in this fraction is mostly atmospheric contamination. The isotopic ratios of Xe which appeared at higher temperatures agree with those for AVCC-Xe except for the 900°C fraction. Heavy isotopes ^{134}Xe and ^{136}Xe in the 900°C fraction are definitely enriched relative to AVCC-Xe. In order to calculate the relative abundances of excess Xe isotopes, the trapped component was subtracted. The calculation was made for two cases: one is that the trapped Xe is AVCC, and the other is that the trapped Xe has a linear fractionation pattern extrapolated from light isotopes of AVCC-Xe determined by PEPIN and PHINNEY (1984). The relative abundance patterns calculated agree with each other within experimental errors, and are similar to the composition of CCF-Xe reported for Allende etched residue (LEWIS *et al.*, 1975) as represented in Fig. 5. Though the variation of Xe isotopic ratios from those of AVCC-Xe is small, the similar relative abundance seems to indicate the presence of CCF-Xe in this fraction. An estimated amount of ^{136}Xe from a spontaneous fission of ^{238}U in 4.5×10^9 y is much lower than the excess ^{136}Xe of $(1-3) \times 10^{-11}$ cm³

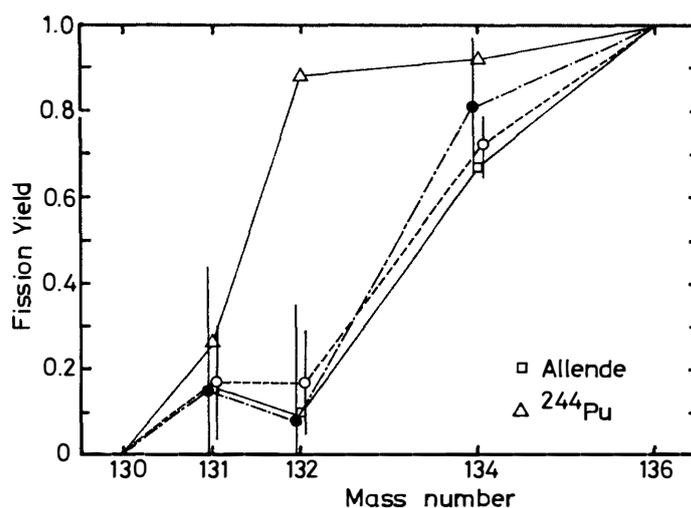


Fig. 5. Mass spectrum of CCF-Xe in the 900°C fraction normalized to ^{136}Xe . Solid and open circles correspond to spectra calculated on assumptions that the trapped Xe is AVCC and that the trapped Xe is a component of linear fractionation (PEPIN and PHINNEY, 1984), respectively. They agree with CCF-Xe reported for Allende etched residue (LEWIS *et al.*, 1975) and distinctly differ from the fission spectrum of ^{244}Pu .

STP/g in the 900°C fraction. The relative abundance pattern shown in Fig. 5 is different from that of fissionogenic Xe from ^{238}U . The pattern is also different from that of fission yield of ^{244}Pu .

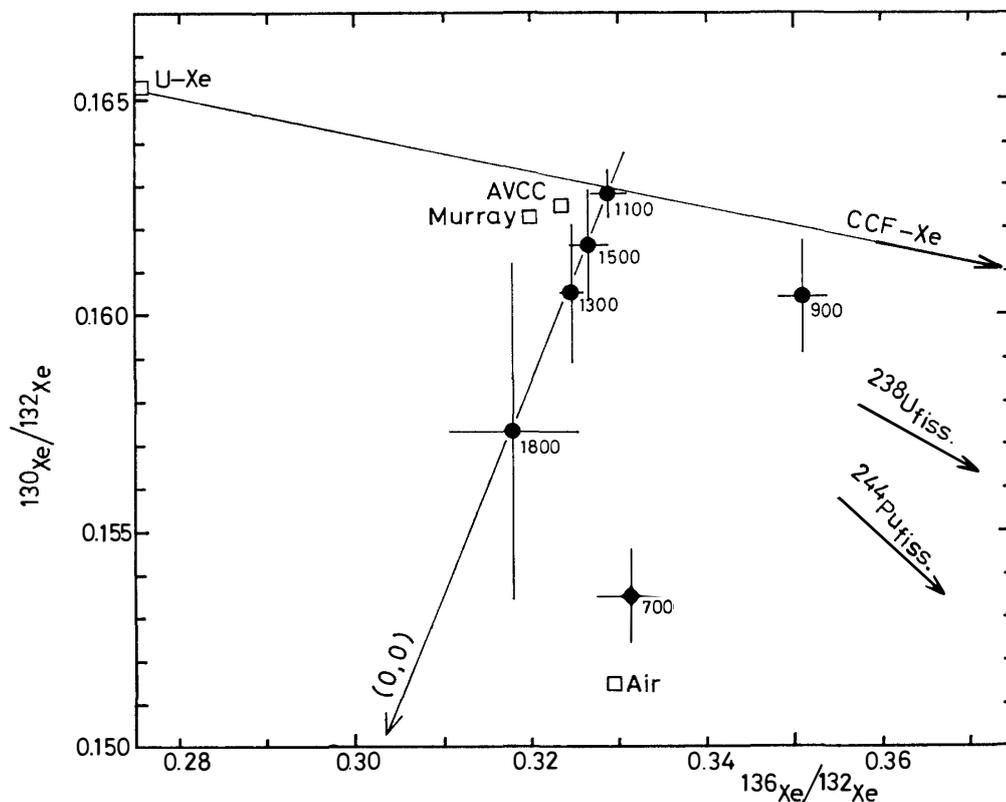


Fig. 6. The $^{130}\text{Xe}/^{132}\text{Xe}$ ratios are plotted against the $^{136}\text{Xe}/^{132}\text{Xe}$ ratios. Four points of higher temperature fractions lie on a straight line which passes the origin. The correlation can be accounted for by an admixture of pure ^{132}Xe .

The $^{130}\text{Xe}/^{132}\text{Xe}$ ratio is plotted against the $^{136}\text{Xe}/^{132}\text{Xe}$ ratio in Fig. 6. The point for 700°C is plotted near the terrestrial atmospheric Xe, which indicates the atmospheric contamination as noted before. Xe for the 900°C fraction shifts rightward from AVCC-Xe. In contrast to these points for two fractions of low temperatures (700 and 900°C), the four points for higher temperature fractions (1100, 1300, 1500 and 1800°C) lie on a straight line through the origin (0, 0) which indicates an admixture of pure ^{132}Xe . For the 1800°C fraction the deviation of the ratios from those of AVCC-Xe is about 3%. The point moves to an opposite direction from the data field reported for s-process Xe found in the etched residues of Murchison C2 chondrite by ALAERTS *et al.* (1980). The s-process Xe is plotted above the mixing line between AVCC-Xe and CCF-Xe.

Such an enrichment is also seen in the Kr isotopic ratios. In Fig. 7 a point for the 1800°C fraction is shifted to the origin (0, 0). Because the excessive ^{132}Xe and ^{84}Kr are released in the 1800°C fraction, the component enriched in ^{132}Xe and ^{84}Kr is retentively held in high temperature host phases. Since the thermal neutron cross sections are large for ^{131}Xe and ^{83}Kr , ^{132}Xe and ^{84}Kr enrichments may occur by *in situ* neutron capture. However, the definite enrichments are found only in the highest temperature fraction

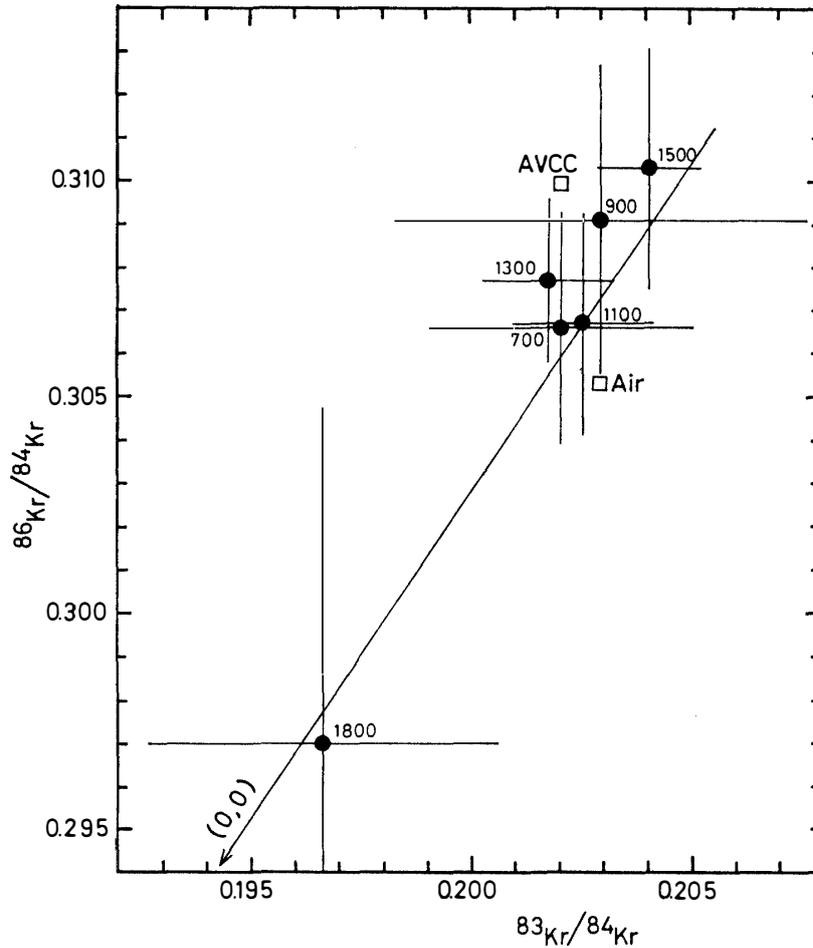


Fig. 7. The $^{86}\text{Kr}/^{84}\text{Kr}$ ratios are plotted against the $^{83}\text{Kr}/^{84}\text{Kr}$ ratios. Kr in the 1800°C fraction is shifted toward the origin which indicates ^{84}Kr excess.

(1800°C). The *in situ* neutron capture should produce not only the enrichments in ^{132}Xe and ^{84}Kr for all the temperature fractions but also the ^{128}Xe , ^{80}Kr and ^{82}Kr enrichments by ($n, \gamma\beta$) reaction from I and Br. The result with no definite enrichments in ^{128}Xe , ^{80}Kr and ^{82}Kr seems to suggest that the *in situ* neutron capture is not a cause of the enrichments. If only a high temperature condensates with no I and Br were irradiated with neutron in the early stage of the solar system, the ^{84}Kr and ^{132}Xe enrichments in the 1800°C fraction might be produced. Another possibility for the enrichments in ^{84}Kr and ^{132}Xe is a stellar nucleosynthesis in a supernova. HEYMANN and DZICZKANIEC (1980) reported that the ^{84}Kr and ^{132}Xe enrichments are produced in the Ne- and C-shells of a supernova of 25 solar mass where the neutron density is low and the photodisintegration-dominated nucleosynthesis occurs.

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