A laboratory experiment on the influence of aqueous alteration on noble gas compositions in the Allende meteorite

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Abstract: We have carried out a hydrothermal experiment on the Allende CV chondrite, in order to understand the observed noble gas loss from CM and CI chondrites, possibly caused by aqueous alteration on their parent bodies (L. B. Browning *et al.*, Geochim. Cosmochim. Acta, **60**, 2621, 1996; T. Nakamura *et al.*, Geochim. Cosmochim. Acta, **63**, 257, 1999). The Allende meteorite was disaggregated and kept with liquid water in a pressurized vessel under 15 atm of water vapor at 200°C for a week. The abundances of helium, neon, argon, krypton and xenon in the samples have been measured by stepwise heating technique. The abundances of noble gases were decreased by 24% (He), 27% (Ne), 38% (Ar), 23% (Kr) and 34% (Xe) after the hydrothermal treatment, suggesting that the hot water treatment had contributed to reduce the concentrations of noble gases in Allende. This gas loss also affected the isotopic composition of xenon. For the 1200°C fraction, the amount of ^{132}Xe-HL was decreased from the sample by about 80% during the hot water treatment, while that of ^{132}Xe-Q was decreased to a lesser extent (~40%). This result seems to indicate that the carrier of Xe-HL is less resistant to the gas loss by the hot water alteration than phase Q for high temperature components. However, further studies are required as this trend becomes less obvious and rather similar if we take into consideration the xenon of 800°C fraction.

1. Introduction

Most carbonaceous chondrites have experienced aqueous alteration, resulting in changes in their texture, mineralogy and possibly chemical composition (Cohen *et al.*, 1983; Tomeoka and Buseck, 1982, 1985; Zolensky, 1984; Zolensky *et al.*, 1987). This aqueous alteration is considered to have occurred in the parent body of meteorite (Kerridge *et al.*, 1979; McSween, 1979; Richardson, 1978; Armstrong *et al.*, 1982; Tomeoka and Buseck, 1985). Water, presumably in the form of ice would have accreted along with lithic materials to form the meteorite parent bodies (Prinn and Fegley, 1987). The liquid water, which caused aqueous alteration, was derived from ice by heat generated within the parent body by the decay of short-lived radionuclides (DuFresne and Anders, 1962).

Recently, Nakamura *et al.* (1999) suggested that the aqueous alteration might

have played an important role in decreasing the abundances of noble gases in CM chondrite. They observed that the degrees of aqueous alterations in the CM chondrites were inversely correlated with the amounts of primordial noble gases trapped in phase Q (main noble gas carrier of carbonaceous material in chondrites; Lewis *et al.*, 1975). Browning *et al.* (1996) also reported similar observation in eight CM chondrites. As the elemental abundances and isotopic compositions of noble gases in chondrites are very important tracers of the processes in the early solar system, the effects of such aqueous alterations on noble gas compositions should be clarified.

There have been a few experimental works investigating the artificial aqueous alteration effect on the mineralogy and the texture of the meteorite (Imae and Shinoda, 1998; Duke and Brearley, 1998). However, how the noble gas compositions are affected by the aqueous alteration is not known. It is also of great interest how the HL component (another primordial component carried in presolar diamonds; Lewis *et al.*, 1987) will be affected by such hydrothermal processes. Therefore, we have carried out the experiments in which we artificially altered the matrix of the Allende meteorite with hot water and examined its effects on noble gas compositions. We chose Allende because it has experienced a minimal aqueous alteration after its formation, and thus should be more sensitive to the artificial alterations than CM and CI that have been altered by natural processes.

2. Sample preparation and experimental procedures

We prepared two samples to examine possible change in their noble gas compositions before and after the artificial hydrothermal alteration. The starting material was the Allende matrix (AC-1) prepared by the freeze-thaw disaggregation. The great majority of AC-1 consists of matrix minerals, but some fragments of condrules and CAIs are also included. The AC-1 is the residual material after we partially removed the floating black fine material that was very similar to the chemical residue enriched in noble gases (Matsuda *et al.*, 1999). Thus, it is likely that noble gas abundances in AC-1 are low compared to those in the bulk Allende. Parts of AC-1 were loaded into a 10 cm^3 teflon vessel together with about 5 cm^3 of pure water. This teflon vessel was then put inside the stainless-steel vessel. We heated up this sample container at 200°C for about one week. The rock to water ratio was about 1/50 by weight, where water would provide sufficient vapor to alter the sample under the pressure of about 15 atm. The altered materials (HM-1) were recovered from the vessel.

Both AC-1 and HM-1 were analyzed for the elemental abundances and the isotopic compositions of noble gases by stepwise heating gas extraction at 800, 1000, 1200, 1400 and 1600°C. The extracted gas was purified by two Ti-Zr getters heated at 700°C to remove any active species. After purification, the extracted noble gases were separated into four fractions (He + Ne, Ar, Kr and Xe) using two cold traps, and then each fraction was successively admitted to the sector-type mass spectrometer (VG5400, Osaka University) fitted with two collectors: Faraday for ^{4}He and

^{40}Ar analyses and an electron multiplier with pulse counting system for other isotopes. Its sensitivities and mass discrimination factors for noble gases were calibrated by analyzing known amounts of atmospheric noble gases. Uncertainties for noble gas sensitivities are about 5% including all the errors of the volumes of line, standard air, etc. The blank correction was applied by subtracting the pre-sample blank levels measured at 800 and 1600°C (the hot blank at 800°C was corrected for the sample gas measured at 800°C, and that at 1600°C was applied for all other temperature steps). Typical hot blanks at 1600°C were as follows: ^{22}Ne = 2.7 to 5.0×10^{-13}, ^{36}Ar = 3.3 to 12×10^{-12}, ^{84}Kr = 2.4 to 11×10^{-13} and ^{132}Xe = 3.6 to 9.0×10^{-14} cm^3 STP. A hot blank for ^{4}He was negligible by the Faraday detector, and we did not make the correction for He. We calculated the isotope ratios by assigning 100% error for the blank levels. In the case of neon isotopes, an isobaric interference on m/e = 22 from CO_2^{++} was subtracted to obtain the ^{22}Ne abundance. Our mass spectrometer can partially resolve the peak of ^{40}Ar^{++} from the ^{20}Ne, so the conventional correction by using a production ratio of ^{40}Ar^{++}/^{40}Ar^{+} was not necessary.

3. Results and discussion

3.1. Elemental abundance

Tables 1 and 2 present the results of noble gas analyses by stepwise heating from AC-1 and HM-1, respectively. Figure 1a to d show the stepped release profiles of ^{22}Ne, ^{36}Ar, ^{84}Kr and ^{132}Xe, respectively. As shown in Fig. 1, the untreated sample released large portions of noble gases at 1200°C. Also in the treated sample, we found release peaks of ^{36}Ar, ^{84}Kr and ^{132}Xe at 1200°C. However, amounts of the released noble gases from the treated sample at 1200°C became smaller than those of the untreated sample. Total amounts of He, Ne, Ar, Kr and Xe decreased by 24%, 27%, 38%, 23% and 34%, respectively, after the hydrothermal treatment. These observations suggest that the artificial alteration is responsible for the loss of noble gases from the chondritic materials. This result confirms the observation of generally lower noble gas amounts in the heavily altered CM chondrites that might have lost their noble gases by aqueous alterations on their parent body (Browning *et al.*, 1996; Nakamura *et al.*, 1999).

3.2. Isotopic compositions

Hereafter, we will discuss the observed apparent gas loss from the samples by comparing the xenon isotopic ratios before and after the hot water treatment. In the present samples, we have recognized three distinct xenon isotope components: Air-Xe, Xe-Q and Xe-HL. A ^{131}Xe/^{132}Xe *vs.* ^{130}Xe/^{132}Xe diagram (Fig. 2) is useful to detect the contribution of Air-Xe because Air-Xe, Xe-Q and Xe-HL do not fall on a single line. As shown in Fig. 2 where the data points of Xe released at 800°C are close to Air-Xe, absorbed air was released from the samples at the lowest temperature. Xenon isotopic ratios of higher temperature steps are clearly non-atmospheric and seem to be distributed around Xe-Q in Fig. 2. However, when

E. Nakasyo, T. Maruoka, T. Matsumoto and J. Matsuda

Table 1. Isotopic ratios and concentrations of AC-1 (18.01 mg).

Temp °C	^{4}He 10^{-6}cm^{3}STP/g	^{3}He/^{4}He 10^{-4}	^{22}Ne 10^{-9}cm^{3}STP/g	^{20}Ne/^{22}Ne	^{21}Ne/^{22}Ne	^{36}Ar 10^{-8}cm^{3}STP/g	^{38}Ar/^{36}Ar	^{40}Ar/^{36}Ar	^{84}Kr 10^{-10}cm^{3}STP/g	^{78}Kr/^{84}Kr	^{80}Kr/^{84}Kr	^{82}Kr/^{84}Kr	^{83}Kr/^{84}Kr	^{86}Kr/^{84}Kr
800	20	29.7	8.7	2.85	0.684	1.0	0.2045	84.4	4.6	0.00422	0.0399	0.1991	0.2043	0.3156
		±0.4		±0.29	±0.070		±0.0009	±2.6		±0.00061	±0.0006	±0.0010	±0.0024	±0.0041
1000	5.7	25.4	5.2	1.97	0.806	2.2	0.2063	81.8	3.0	0.0064	0.0437	0.1976	0.2023	0.3101
		±0.1		±0.21	±0.015		±0.0009	±2.9		±0.0025	±0.0015	±0.0017	±0.0022	±0.0029
1200	0.9	8.6	5.6	2.19	0.773	7.4	0.2033	11.2	7.0	0.00617	0.0429	0.1983	0.2047	0.3080
		±0.4		±0.23	±0.079		±0.0008	±0.8		±0.00037	±0.0007	±0.0017	±0.0017	±0.0027
1400	N.D.		2.3	1.91	0.831	1.4	0.1963	27.0	1.9	N.D.	0.0512	0.197	0.208	0.310
				±0.22	±0.085		±0.0009	±4.3			±0.0024	±0.014	±0.011	±0.010
1600	N.D.		(0.2)	7.4	0.089	0.1	0.1970	182	(0.2)	N.D.	0.055	N.D.	0.220	0.341
				±1.4	±0.032		±0.0050	±17			±0.024		±0.030	±0.049
Total	27	28.1	22	2.41	0.747	12	0.2031	23.0	17	0.00560	0.0434	0.1982	0.2049	0.3114
		±1.5		±0.14	±0.035		±0.0005	±1.0		±0.00051	±0.0007	±0.0023	±0.0017	±0.0023

Temp °C	^{132}Xe 10^{-10}cm^{3}STP/g	^{124}Xe/^{132}Xe	^{126}Xe/^{132}Xe	^{128}Xe/^{132}Xe	^{129}Xe/^{132}Xe	^{130}Xe/^{132}Xe	^{131}Xe/^{132}Xe	^{134}Xe/^{132}Xe	^{136}Xe/^{132}Xe
800	3.0	0.00452	0.00403	0.07619	1.1039	0.1506	0.7994	0.4140	0.3697
		±0.00065	±0.00048	±0.00087	±0.0045	±0.0007	±0.0073	±0.0010	±0.0018
1000	2.3	0.00523	0.00494	0.08376	1.3081	0.1634	0.8150	0.3895	0.3306
		±0.00045	±0.00052	±0.00097	±0.0069	±0.0012	±0.0083	±0.0035	±0.0029
1200	6.1	0.00467	0.00381	0.08420	1.5448	0.1629	0.8174	0.3872	0.3261
		±0.00032	±0.00034	±0.00080	±0.0068	±0.0009	±0.0077	±0.0017	±0.0015
1400	1.6	0.00403	0.00409	0.0839	2.041	0.161	0.8250	0.3930	0.3354
		±0.00055	±0.00080	±0.0014	±0.015	±0.013	±0.0081	±0.0050	±0.0040
1600	0.1	N.D.	N.D.	0.053	2.55	0.095	0.820	0.47	0.400
				±0.016	±0.38	±0.012	±0.040	±0.11	±0.090
Total	13	0.00466	0.00410	0.08207	1.4691	0.1596	0.8138	0.3949	0.3384
		±0.00024	±0.00024	±0.00051	±0.0078	±0.0017	±0.0044	±0.0014	±0.0013

The datum with parenthesis means >20% contributions from the procedural blank

N.D. = not determined due to small amount of gas released.

Table 2. Isotopic ratios and concentrations of HM-1 (20.12 mg).

Temp °C	^{4}He 10^{-6}cm^{3}STP/g	^{3}He/^{4}He 10^{-4}	^{22}Ne 10^{-9}cm^{3}STP/g	^{20}Ne/^{22}Ne	^{21}Ne/^{22}Ne	^{36}Ar 10^{-8}cm^{3}STP/g	^{38}Ar/^{36}Ar	^{40}Ar/^{36}Ar	^{84}Kr 10^{-10}cm^{3}STP/g	^{78}Kr/^{84}Kr	^{80}Kr/^{84}Kr	^{82}Kr/^{84}Kr	^{83}Kr/^{84}Kr	^{86}Kr/^{84}Kr
800	14	26.7 ±0.7	6.4	3.21 ±0.33	0.657 ±0.067	1.1	0.1974 ±0.0009	167.2 ±3.8	6.0	0.00589 ±0.00018	0.03852 ±0.00049	0.1988 ±0.0011	0.2016 ±0.0012	0.3071 ±0.0017
1000	4.9	20.6 ±0.1	3.8	3.08 ±0.35	0.680 ±0.014	1.7	0.1993 ±0.0010	(*)8.6	(2.0)	0.00539 ±0.00078	0.0377 ±0.0020	0.1955 ±0.0020	0.1964 ±0.0021	0.3067 ±0.0033
1200	0.7	9.5 ±0.5	3.1	2.18 ±0.29	0.787 ±0.081	3.4	0.1986 ±0.0009	2.4 ±3.8	4.3	0.0063 ±0.0059	0.04224 ±0.00064	0.2014 ±0.0046	0.2023 ±0.0048	0.3113 ±0.0049
1400	0.2	N.D.	2.2	1.81 ±0.32	0.822 ±0.084	1.2	0.1946 ±0.0011	10 ±22	(1.4)	0.00642 ±0.00085	0.0489 ±0.0032	0.2080 ±0.0070	0.2026 ±0.0027	0.3111 ±0.0041
1600	N.D.		0.1	3.7 ±9.7	0.61 ±0.11	0.1	0.199 ±0.011	48 ±112	(0.2)	0.008 ±0.016	0.06 ±0.12	0.23 ±0.31	0.22 ±0.20	0.30 ±0.12
Total	20	20.8 ±2.1	16	2.78 ±0.19	0.711 ±0.034	7.5	0.1979 ±0.0054	36.1 ±4.6	13	0.0061 ±0.0018	0.0416 ±0.0052	0.201 ±0.013	0.2017 ±0.0086	0.3086 ±0.0055

Temp	^{132}Xe 10^{-10}cm^{3}STP/g	^{124}Xe/^{132}Xe	^{126}Xe/^{132}Xe	^{128}Xe/^{132}Xe	^{129}Xe/^{132}Xe	^{130}Xe/^{132}Xe	^{131}Xe/^{132}Xe	^{134}Xe/^{132}Xe	^{136}Xe/^{132}Xe
800	1.5	0.00531 ±0.00033	0.00366 ±0.00027	0.0802 ±0.0014	1.331 ±0.016	0.1493 ±0.0083	0.8077 ±0.0026	0.4260 ±0.0023	0.3896 ±0.0027
1000	2.0	0.00519 ±0.00031	0.00392 ±0.00039	0.0835 ±0.0015	1.120 ±0.005	0.1623 ±0.0033	0.828 ±0.013	0.3935 ±0.0015	0.3366 ±0.0022
1200	3.4	0.00489 ±0.00017	0.00471 ±0.00039	0.0832 ±0.0014	1.376 ±0.007	0.1648 ±0.0017	0.8262 ±0.0076	0.3811 ±0.0010	0.3205 ±0.0019
1400	1.4	0.00457 ±0.00026	0.00455 ±0.00050	0.0842 ±0.0015	1.928 ±0.025	0.1676 ±0.0038	0.837 ±0.018	0.3825 ±0.0022	0.3243 ±0.0029
1600	0.2	0.0095 ±0.0037	0.0086 ±0.0037	0.0820 ±0.0050	2.19 ±0.27	0.188 ±0.036	0.96 ±0.18	0.374 ±0.012	0.315 ±0.017
Total	8.6	0.00508 ±0.00015	0.00436 ±0.00022	0.08287 ±0.00074	1.416 ±0.010	0.1624 ±0.0021	0.8279 ±0.0064	0.3922 ±0.0009	0.3372 ±0.0013

The datum with parenthesis means >20% contributions from the procedural blank.

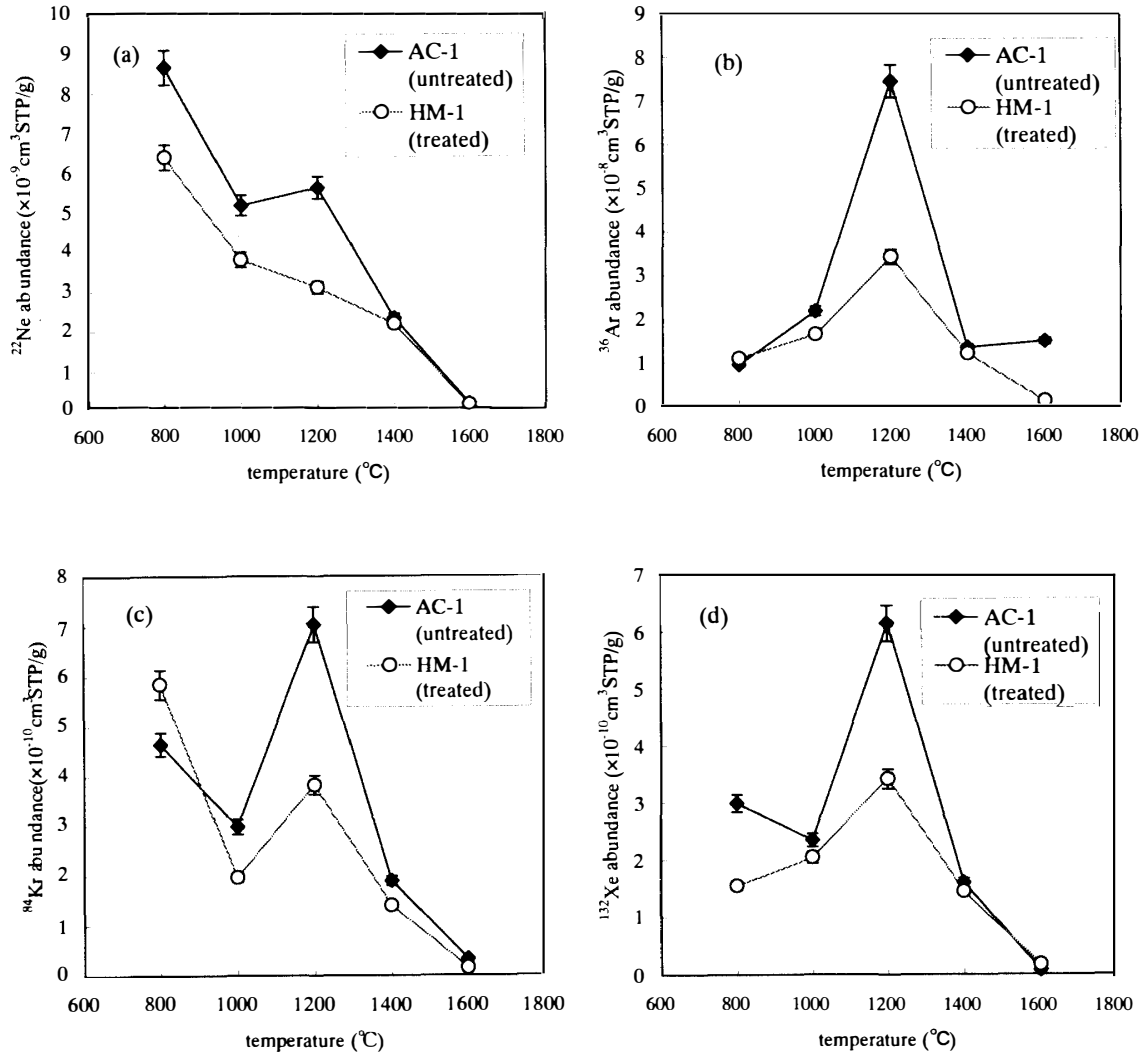N.D. = not determined due to small amount of gas released.

(*) means upper limit

Fig. 1. *Release profiles of ^{22}Ne, ^{36}Ar, ^{84}Kr and ^{132}Xe in the untreated and treated Allende matrix. Isotopic ratios of $^{20}Ne/^{22}Ne$ and $^{38}Ar/^{36}Ar$ indicate the contributions from spallogenic ^{22}Ne and ^{36}Ar in each temperature step (see Tables 1 and 2), but they probably do not affect the overall trends shown in (a) and (b); because spallogenic ^{22}Ne occurs in fairly constant proportion in each step and the contribution on ^{36}Ar should apparently be small in comparison to the original components (Q and HL).*

these data are plotted in a $^{134}Xe/^{132}Xe$ vs. $^{136}Xe/^{132}Xe$ diagram (Fig. 3), the data points of high temperature fractions form a clear linear array extending from Xe-Q to Xe-HL, suggesting that xenon released at higher temperature steps are explained by variable mixtures of Xe-Q and Xe-HL. As these two components are very close each other in Fig. 2, such a mixing trend is less clear in Fig. 2 than in Fig. 3. Figure 3 is more useful than Fig. 2 to examine the relationship of Xe-Q and Xe-HL because these two components are more separable from each other. Note that, although there are some indications of Q and HL component for Ne, Ar and Kr in the samples (Tables 1 and 2), the presence of additional cosmogenic or atmospheric components makes it difficult to identify the Q and HL mixing relationship in those isotope systems.
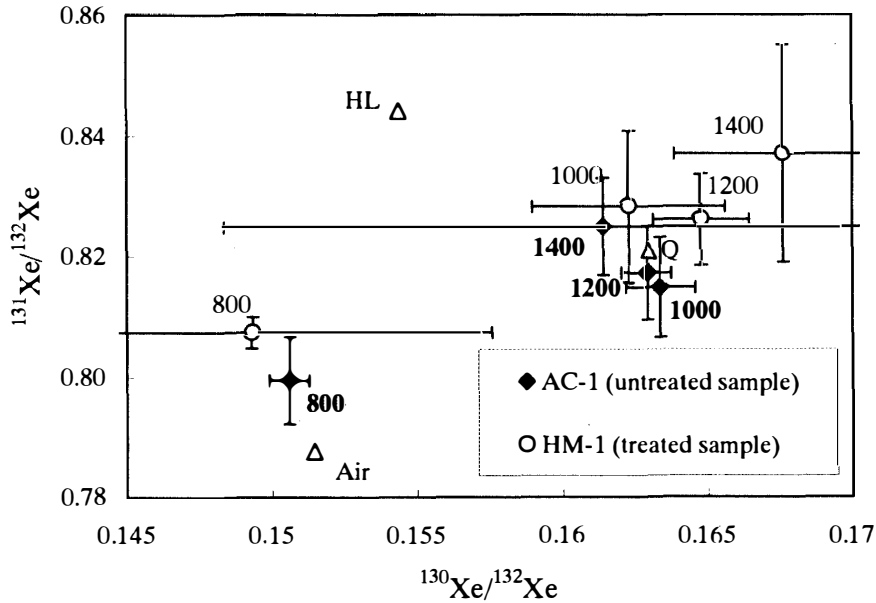
Fig. 2. A $^{131}Xe/^{132}Xe$ versus $^{130}Xe/^{132}Xe$ diagram. The data of 1600°C are excluded because of larger uncertainties. The data source of Xe-Q is from Huss et al. (1996), HL from Huss and Lewis (1994) and Air from Nier (1950).
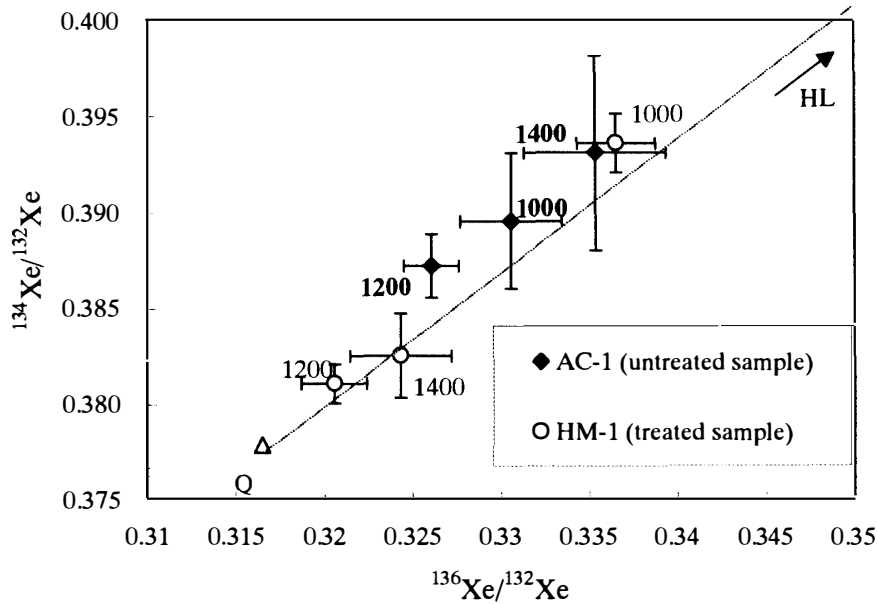

Fig. 3. A $^{134}Xe/^{132}Xe$ versus $^{136}Xe/^{132}Xe$ diagram. The data of 800°C are excluded because of air contamination. The data of 1600°C are excluded because of the large uncertainty. The data source of Q is from Huss et al. (1996), HL is from Huss and Lewis (1994).

3.3. Nature of the lost xenon by artificial hot water alteration

An interesting point to be noted in Fig. 3 is that the xenon isotopic ratios observed in the treated sample are systematically closer to Xe-Q compared to those of the untreated sample. This feature is most clearly seen in xenon released at 1200 and 1400°C where we observed the major gas loss during the hot water treatment (Fig. 1). The shifts of data points towards Xe-Q after the hot water treatment
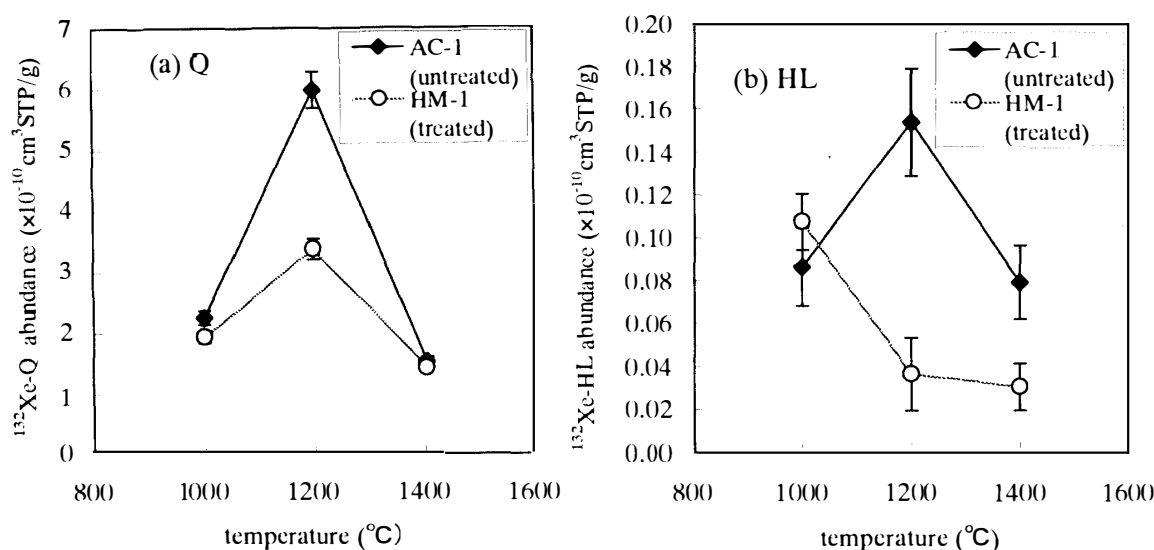
Fig. 4. The release profiles of ^{132}Xe-Q and ^{132}Xe-HL from AC-1 and HM-1. The data of 800 and 1600°C are excluded because of air contamination and large uncertainty, respectively. ^{132}Xe-Q and ^{132}Xe-HL concentrations are determined by the following equations;

$$\alpha(^{136}Xe/^{132}Xe)_Q + (1-\alpha)(^{136}Xe/^{132}Xe)_{HL} = (^{136}Xe/^{132}Xe)_{measured}$$

$$^{132}Xe\text{-}Q = \alpha(^{132}Xe)_{measured}$$

$$^{132}Xe\text{-}HL = (1-\alpha)(^{132}Xe)_{measured}$$

Table 3. The percentages of the concentrations of ^{132}Xe-Q and ^{132}Xe-HL in HM-1 (the sample after the hydrothermal treatment) relative to those in AC-1 (the starting material).

^{132}Xe-Q	^{132}Xe-HL	Note
57±4	23±11	Only for the 1200°C fraction
69±3	54±10	For the 1000–1400°C fractions
	77± 7	Only for the 800°C fraction*
69±3	66± 6	Xenon-Q for the 1000–1400°C fractions and Xe-HL for the 800–1400°C fractions*

* The concentration of ^{132}Xe-HL for the 800°C fraction was calculated on the assumption that Xe in the 800°C fraction was the mixture of Air and Xe-HL.

indicate that Xe-HL had been lost from the samples more significantly than Xe-Q for these temperatures. For the quantitative discussion, we have calculated the fractions of ^{132}Xe-Q and ^{132}Xe-HL components in each temperature step (see caption to Fig. 4 for the calculation). Figure 4 shows the release profiles of the deconvolved ^{132}Xe-Q and ^{132}Xe-HL for temperatures between 1000 to 1400°C. As shown in Fig. 4, the hot water treatment had removed 77% of ^{132}Xe-HL from AC-1, whereas this treatment had removed 43% of ^{132}Xe-Q at 1200°C fraction. This feature is also obvious when we compare the amount of ^{132}Xe-Q and that of ^{132}Xe-HL for the 1000 to 1400°C steps (Table 3). About 70% of ^{132}Xe-Q observed in AC-1 at these temperatures had been survived the hot water treatment while the HM-1 preserved 54% of the original ^{132}Xe-HL released between 1000 to 1400°C.

It has been reported that ordinary chondrites lost their Xe-HL component more significantly than Xe-Q by thermal metamorphism, which indicates that the

carrier of Xe-HL is less resistant to thermal metamorphism than that of Xe-Q (Huss *et al.*, 1996). Although the conditions of pressure, temperatures and duration time of hot water alteration experiment were quite different from those of thermal metamorphism, the present results indicate that the HL carrier is indeed less retentive than phase-Q during thermal and hydrothermal alterations as far as the results of high temperature steps are concerned.

We have excluded the data of $800°C$ fraction in the above discussion. They were largely air-contaminated, so the deconvolution with Xe-HL and Xe-Q was impossible for the $800°C$ data. However, it should also be noted that $^{136}Xe/^{132}Xe$ ratios in $800°C$ fractions are highest among those in other temperature steps (Tables 1 and 2), indicating that considerable amounts of ^{132}Xe-HL were released at $800°C$. Thus, we calculated the amounts of ^{132}Xe-HL by assuming that Xe for the $800°C$ fractions was the mixture of only Air and Xe-HL. This would be reasonable as the $800°C$ data plot close to an Air and Xe-HL mixing trend in Fig. 2. The calculated amounts of ^{132}Xe-HL in the $800°C$ fractions are the largest among those estimated in all temperature steps in AC-1 and HM-1. We also observed the loss of ^{132}Xe-HL after the hot water treatment in the $800°C$ fraction, but its degree (23%) is apparently smaller than those observed in higher temperature fractions. As a result, when we include the $800°C$ fraction results for ^{132}Xe-HL, the suggested difference in retentivities of Xe-Q and Xe-HL became less clear (last row in Table 3). Therefore, although the high temperature fractions do suggest the higher gas retentivity for Xe-Q than Xe-HL upon the alteration as observed in the actual chondrites, further studies are necessary to clarify this feature because both components appeared to have been lost from the samples to a similar extent as a whole by the hot water treatment.

4. Summary

We have carried out a laboratory experiment that simulates the aqueous alteration of carbonaceous chondrites on their parent body. Elemental abundances and isotopic compositions of noble gas were measured in Allende before and after the alteration, and we found that:

1) The total abundance of each noble gas in Allende was decreased after the hot water treatment. This result confirms the observation of generally lower noble gas amounts in the CM chondrites that might have lost their noble gases by aqueous alterations on their parent body (Browning *et al.*, 1996; Nakamura *et al.*, 1999).

2) The xenon isotopic ratios suggest that the host phase of Xe-Q is less susceptive to gas loss by the hydrothermal alteration than that of Xe-HL for high temperature fractions. However, this feature is not clear when we compare the data for the total fraction. Further experiments should be necessary to examine the difference of the retentivity of Xe-Q and Xe-HL in more detail.

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