

TEMPERATURE DEPENDANCE OF MINERALOGICAL AND NOBLE GAS COMPOSITIONAL CHANGES DURING EXPERIMENTAL AQUEOUS ALTERATION OF NINGQIANG. Y. Yamamoto¹, T. Nakamura², T. Noguchi³, R. Okazaki² and K. Nagao¹. ¹Laboratory for Earthquake Chemistry, Graduate School of Science, University of Tokyo, Hongo, Tokyo 113-0033, Japan. ²Department of Earth and Planetary Sciences, Faculty of Sciences, Kyushu University, Hakozaki, Fukuoka 812-8581, Japan. ³Department of Materials and Biological Sciences, Faculty of Science, Ibaraki University, Bunkyo, Mito 310-8512, Japan. (y-yamam@eqchem.s.u-tokyo.ac.jp)

Introduction: Carbonaceous chondrites are one of the most primitive materials in the solar system and contain large amounts primordial noble gases. Ar-rich gas, one of the components of primordial noble gases, has $^{36}\text{Ar}/^{132}\text{Xe}$ and $^{84}\text{Kr}/^{132}\text{Xe}$ ratios higher than Q gas [1] and is contained in some anhydrous carbonaceous chondrites [e.g., 2, 3]. On the other hand, many carbonaceous chondrites experienced aqueous alteration that is thought to be the earliest chemical reaction and might have changed compositions of primordial noble gases. Carbonaceous chondrites that suffered extensive aqueous alteration are lack of Ar-rich gas, whereas they contain Q gas [4]. It would be expected that Ar-rich gas had been released during aqueous alteration.

In an earlier work, we revealed that a major part of primordial noble gases in the Ningqiang carbonaceous chondrite lost during experimental aqueous alteration [5] and the concentrations of primordial noble gases remaining in Ningqiang became lower as alteration progressed [6]. In this study, we report the temperature dependence of changes of mineralogy and noble gas compositions during experimental aqueous alteration.

Sample and experimental method: Ningqiang is classified as CV3- [7, 8] or CK3-anomalous [e.g., 9] types. This chondrite consists entirely of anhydrous minerals and contains large amount of Ar-rich gas as well as Q gas [e.g., 10]. These results suggest that this chondrite has not experienced extensive aqueous alteration in its parent body.

We prepared bulk Ningqiang powders (μm in size) weighing 600 mg and 10 g neutral liquid water. Samples were loaded into pressurized vessels and were kept at 100°C and 200°C each. After 0.5, 1, 2, 5, 10, and 20 days, 100mg of the sample was recovered and the rest of the sample was kept being soaked for another days. Natural and altered samples were analyzed for mineralogy and noble gases.

Mineral compositions before and after experimental alteration of Ningqiang samples were analyzed by powder X-ray diffraction method (Powder XRD) and synchrotron radiation X-ray diffraction method

(SR-XRD). After that, FE-SEM and TEM observation were performed.

Detailed configurations of noble gas analyses are given in an earlier work [6]. Noble gases were extracted at 300, 700, 1000, 1300 and 1850°C. The concentrations and isotopic ratios of the noble gases were determined with a mass spectrometer (modified MM5400) at Kyushu University and University of Tokyo.

Results and Discussion: Mineralogical analyses show that the natural Ningqiang consists of olivine, low-Ca pyroxene, magnetite and iron sulfide. In the sample altered at 100°C, mineralogical changes are minor through the periods of experiments and the changes could not detected by both XRD analyses and electron microscopic observations. In contrast to the 100°C altered sample, 200°C altered one greatly changed its mineralogical compositions. Olivine, low-Ca pyroxene and iron sulfide were decomposed, and serpentine and hematite were formed in the samples altered at 200°C. Serpentine appears to have formed from the elements supplied by the decomposition of olivine and pyroxene. The serpentine / (olivine + low-Ca pyroxene) ratio, determined from integrated intensities of the powder XRD diffraction peaks, increases from the 0.5-day altered sample to the 5-day one, but remains constant after the 5 days. These results indicate that a large portion of serpentine formed in the early stage of experimental aqueous alteration at 200°C. It was confirmed by the electron microscopic observation that serpentine grows around olivine and pyroxene grains (Fig. 1). In the sample altered over 10 days, all the olivine and pyroxene grains smaller than $1\mu\text{m}$ were completely replaced by serpentine.

Some amounts of noble gases in the Ningqiang were removed by the experimental aqueous alteration. In the sample altered at 100°C, 12%, 43% and 30% of ^{36}Ar , ^{84}Kr and ^{132}Xe , respectively, were removed from natural Ningqiang during only the 0.5-day alteration. The $^{36}\text{Ar}/^{132}\text{Xe}$ and $^{84}\text{Kr}/^{132}\text{Xe}$ ratios of removed gases during experimental aqueous alteration at 100°C are toward to atmospheric ratios. This suggests that major part of the noble gases removed during experimental

aqueous alteration at 100°C were atmospheric noble gases absorbed in Ningqiang.

Noble gas compositions in the 200°C altered samples drastically changed compared with those in the 100°C altered samples. 84%, 73% and 70% of ^{36}Ar , ^{84}Kr and ^{132}Xe , respectively, were removed from natural Ningqiang during only the 5-day alteration, while the losses of ^4He and ^{20}Ne were 42% and 41%, respectively. These indicate that the altered phases contain greater amounts of Ar, Kr and Xe than those remaining in the 5-day sample. Altered portions of both olivine and pyroxene lose cosmogenic ^{21}Ne because alteration products such as serpentine are formed via reconstruction of atoms in olivine and pyroxene. Therefore, ^{21}Ne concentrations in the altered samples could be a measure of the extent of the aqueous alteration reactions.

As the alteration proceeds, the concentrations of primordial noble gases decrease steeply while those of ^{21}Ne decreases slowly (Fig. 2), suggesting that even low degrees of aqueous alteration removes great amounts of primordial noble gases. In the sample altered over 5 days, noble gas concentrations are basically similar to those of the 5-day sample, thus indicating that the loss of primordial noble gases completed within the 5-day alteration. Among primordial noble gases, Ar-rich gas was lost in the largest amounts by the experimental aqueous alteration. The $^{36}\text{Ar}/^{132}\text{Xe}$ and $^{84}\text{Kr}/^{132}\text{Xe}$ ratios of the noble gases remained in altered samples fall gradually as the alteration progresses from 0.5 to 5 days. The calculated elemental ratios of the noble gases lost from samples are higher than those of the natural Ningqiang, indicating that the noble gases removed during the alteration are enriched in Ar-rich gas. This result suggests that Ar-rich gas is located in materials that are very susceptible to aqueous alteration. In contrast, primordial noble gases remaining in the samples after aqueous alteration are close to Q gas based on elemental and isotope compositions, indicating that the trapping phase of Q gas is much more resistant to aqueous alteration than the host phases of Ar-rich gas. The results of our study confirm that chondrites experienced aqueous alteration, like CM chondrites, do not have Ar-rich noble gases, although it has large amounts of Q gas.

As well as primordial noble gases, radiogenic noble gases were also significantly removed during the experimental aqueous alteration. Almost all of ^{129}Xe , which is generated by the β decay of ^{129}I , was lost by the 10-day alteration (Fig. 2). This result suggests that not

only thermal metamorphism but also aqueous alteration could disturb radiometric ages, and it is therefore thought that the ^{129}I - ^{129}Xe age reflects the timing when the aqueous alteration has finished in hydrous asteroids.

References: [1] Wacker J. F. and Marti K. (1983) *EPSL*, 62, 147-158. [2] Schelhaas N. et al. (1990) *GCA* 54, 2869-2882. [3] Miura Y. N. et al. (2002) *Proc. NIPR Symp. Antarct. Meteorites.*, 27, 96-97. [4] Nakamura T. et al. (1999) *GCA*, 63, 241-255. [5] Yamamoto Y. et al. (2004) *Meteorit. Planet. Sci.*, 39, A116. [6] Yamamoto Y. et al. (2005) *Proc. NIPR Symp. Antarct. Meteorites*, 29, 98-99. [7] Rubin A. E. et al. (1988) *Meteoritics*, 23, 13-23. [8] Koeberl C. et al. (1987) *LPS XVIII*, 499-500. [9] Kallemeyn G. W. (1996) *LPS XXVII*, 635-636. [10] Nakamura T. et al. (2003) *Meteorit. Planet. Sci.*, 38, 243-250.

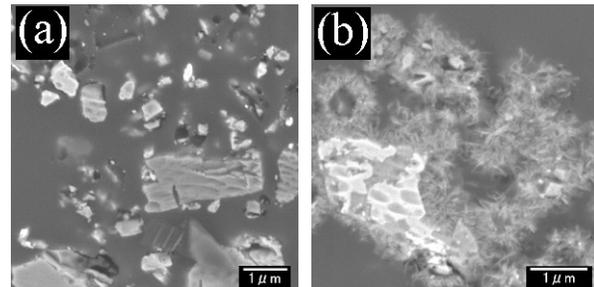


Fig. 1. The BSE images of (a) Natural Ningqiang. Small particles are mainly olivine. (b) 20-day altered sample at 200°C. Fibrous materials around silicate grains are serpentine.

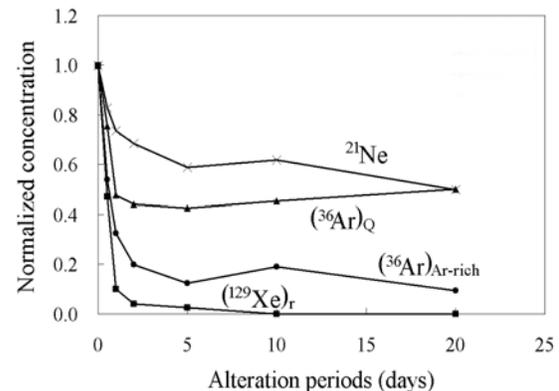


Fig. 2. The changes of ^{21}Ne , $(^{36}\text{Ar})_{\text{Q}}$, $(^{36}\text{Ar})_{\text{Ar-rich}}$ and radiogenic ^{129}Xe , $(^{129}\text{Xe})_{\text{r}}$, concentrations in the 200°C altered sample. Concentrations are normalized to those in the natural Ningqiang.