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Water-susceptible primordial noble gas components in less-altered CR chondrites: A possible link to cometary materials

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Abstract

We report detailed characterizations of the petrology, mineralogy, and noble gas signatures of Renazzo-type (CR) chondrites EET 92048, MIL 090657, NWA 801, and hydrothermally treated MIL 090657 samples to investigate primordial noble gases hosted by water-susceptible materials in aqueously less-altered CR chondrites. The noble gases were extracted by a stepwise heating method. The hydrothermal treatment on MIL 090657 resulted in the alteration of amorphous silicates in the matrix and the removal of large amounts of He and Ne that were released at low temperature from the untreated sample. The ³⁶Ar/¹³²Xe ratios in EET 92048 and MIL 090657 are ~5 times higher than that of the Q noble gas component, although both meteorites do not show any evidence for solar wind noble gases. The analyses revealed that aqueously less-altered CR chondrites contain two distinct primordial noble gas components hosted by water-susceptible materials: an isotopically Q-like Ne-rich component and an Ar-rich component. The characteristics of the Q-like Ne-rich component are consistent with those of Q-like Ne in cometary materials. We propose that the CR chondrites formed at a greater heliocentric distance and share similar materials hosting the Q-like Ne-rich component with comets. The presence of the Ar-rich component in the CR chondrites, as well as in ordinary chondrites and other anhydrous carbonaceous chondrites, provides evidence of the global distribution of the Ar-rich carriers in the ordinary and carbonaceous chondrite forming regions. © 2021 Elsevier Ltd. All rights reserved.

Keywords: CR chondrites; Primitive meteorites; Noble gases; Aqueous alteration

1. INTRODUCTION

Noble gases in meteorites are essential tracers for the formation processes of materials in the early Solar System. Various noble gas isotopic anomalies associated with

https://doi.org/10.1016/j.gca.2021.08.012 0016-7037/© 2021 Elsevier Ltd. All rights reserved. nucleosynthesis in stars and supernovae have been identified in presolar grains in primitive chondrites (e.g., Lewis et al., 1987; Huss and Lewis, 1995). The HL noble gas component in presolar nanodiamonds is characterized by the enrichments in heavy (¹³⁴Xe and ¹³⁶Xe) and light isotopes (¹²⁴Xe and ¹²⁶Xe) of Xe produced by r-process and p-process, respectively. The G component in presolar SiC and graphite has isotopic anomalies produced by s-process, such as the overabundances of ²²Ne, ⁸²Kr, ⁸⁶Kr, ¹²⁸Xe, and ¹³⁰Xe. On the other hand, the Q component has the "normal" isotopic compositions (i.e., similar

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to those of the solar wind) but is characterized by the highly fractionated elemental composition in favor of the heavier noble gases relative to the solar composition (Busemann et al., 2000). The Q component dominates the heavy noble gases (Ar, Kr, and Xe) in most primitive chondrites. For example, ~95% of all ¹³²Xe in the chemically primitive Orgueil CI carbonaceous chondrite belongs to the Q component (Ott, 2002). The presolar grains mentioned above and phase Q (a carbonaceous material hosting the Q component) are acid (HF/HCl) insoluble materials. Such acidinsoluble noble gas components have been extensively studied (e.g., Lewis et al., 1975; Huss and Lewis, 1995; Busemann et al., 2000). However, the acid-soluble noble gas components that are removed during the acid treatment have received little attention so far (Ott, 2014).

Previous experiments have shown that HF/HCl-soluble materials in unequilibrated ordinary chondrites (UOCs) and some classes of anhydrous carbonaceous chondrites contain an Ar-rich noble gas component characterized by high Ar/Xe and Kr/Xe ratios relative to the Q component (Alaerts et al., 1979; Matsuda et al., 1980; Schelhaas et al., 1990; Nakamura et al., 2003b). For example, the ³⁶Ar/¹³²Xe ratios in the acid-soluble fraction of the CO chondrites are up to ~850 (Alaerts et al., 1979), which is much higher than the ${}^{36}\text{Ar}/{}^{132}\text{Xe}$ ratios of Q (40–120; Busemann et al., 2000). On the other had, closed system step etching (CSSE) of Ivuna CI chondrite (hydrous chondrite) revealed that small fractions of primordial Ne and Xe are carried by HF-soluble portion (Riebe et al., 2017), whereas there is no evidence for the presence of Ar-rich component. The hydrothermal treatment of a primitive carbonaceous chondrite Ningqiang (C3-ungrouped) resulted in a removal of the Ar-rich component, indicating that the Arrich component is hosted by water-susceptible phases (Yamamoto et al., 2006). Recently, Krietsch et al. (in press) found that the least aqueously altered CM chondrites contain the Ar-rich component, whereas most CM chondrites do not. These results suggest that aqueously altered chondrites (e.g., CI, CM, and CR chondrites) might have lost the Ar-rich component during the alteration processes on their parent bodies. Despite its early discovery of the Arrich component, its origin, radial distribution in the early Solar System, and even its host phase are poorly understood.

The aqueously altered characteristics for CI, CM, and CR chondrites suggest their formation at greater heliocentric distances than anhydrous chondrites where relatively abundant water ices had existed. Recent studies suggest that the CR chondrites have formed at a greater heliocentric distance than most of the other chondrites based on their isotopically distinct characteristics and low abundances of CAIs (calcium-rich, aluminum-rich inclusions) (Van Kooten et al., 2016, 2020; Budde et al., 2018; Desch et al., 2018). Unlike the CI and CM chondrites, some CR chondrites have avoided an extensive aqueous alteration, and the least altered CR chondrites represent some of the most pristine extraterrestrial samples available for study (e.g., Weisberg et al., 1993; Abreu and Brearley, 2010; Schrader et al., 2015; Davidson et al., 2019b). Those less-altered CR chondrites may preserve primordial

water-susceptible noble gas components. Thus, the CR chondrites provide a potential to investigate the presence of the Ar-rich component in the outer part of the chondrite-forming region in the early Solar System.

While the CR chondrites have been extensively studied by many kinds of analytical techniques (e.g., Alexander et al., 2013; Davidson et al., 2014; Floss et al., 2014; Howard et al., 2015; Schrader et al., 2015; Tenner et al., 2015, 2019; Abreu, 2016), noble gas compositions covering all noble gas elements and isotopes have never been reported in detail except for a few CR chondrites that are aqueously altered (Reynolds and Turner, 1964; Mazor et al., 1970) and solar-gas-rich CR chondrites (Bischoff et al., 1993; Scherer and Schultz, 2000). Solar-gas-rich CR chondrites containing abundant solar wind noble gases are regolith breccias that acquired solar wind noble gases by direct implantation on the surfaces of the meteorite parent bodies (Wieler, 2002a). Such solar wind noble gases often dominate He, Ne, and Ar in solar-gas-rich meteorites and hide the primordial noble gas compositions (e.g., Obase et al., 2020). Therefore, the primordial noble gas contents in less-altered CR chondrites are still not fully understood. Recently, based on total extraction noble gas analysis for ~20 bulk CR chondrites covering the whole range of aqueous alteration observed in CR chondrites, presence of the Ar-rich component in aqueously less-altered CR chondrites and water-susceptible nature of the Ar-rich component in CR chondrites were suggested (Busemann et al., 2019, 2021)

To understand the primordial noble gas signatures in less-altered CR chondrites, we analyzed the concentrations and isotopic ratios of He, Ne, Ar, Kr, and Xe extracted from three CR chondrites Elephant Moraine (EET) 92048, Miller Range (MIL) 090657, and Northwest Africa (NWA) 801 with a stepwise heating method. The degrees of aqueous alteration are also evaluated based on the characterization of petrology and mineralogy of each meteorite.

In the present study, we found high 36 Ar/ 132 Xe ratios of ~350 in the solar-gas-poor CR chondrites MIL 090657 and EET 92048, suggesting that these meteorites contain the Arrich component. Recently, Krietsch et al. (2019) and Krietsch (2020) performed the CSSE noble gas analysis on MIL 090657 in five separate etch runs. Water, acetic acid, HNO₃, HF, and HCl were used as "etching agent" for noble gas extraction. They found that large fractions of ⁴He and ²⁰Ne (\sim 25% and \sim 35% of the total ⁴He and $^{20}\ensuremath{\text{Ne}}$ in MIL 090657, respectively) were released by H_2O treatment without releasing a significant fraction of ³⁶Ar (<2%). On the other hand, most of 36 Ar (~60%) was released by acetic acid treatment. These results suggest the presence of another water-susceptible noble gas component enriched in He and Ne in MIL 090657. They also report that the Ne isotopic ratio of this component is in the range of the Q component. However, the host phase and the origin of this component is still not understood.

To constrain the host phases of the two watersusceptible noble gas components, we performed a hydrothermal treatment on MIL 090657 and examined its effect on the noble gas contents and mineral compositions. The treatment removed large fractions of He and Ne. In this paper, we discuss the possible host phases, the radial distributions in the early Solar System, and the origins of both the Ar-rich component and the new water-susceptible noble gas component enriched in He and Ne, based on the detailed characterizations of the petrology, mineralogy, and noble gas contents of the three CR chondrites and the hydrothermally treated MIL 090657 samples.

2. METHODS

2.1. Samples

The following three CR chondrites were analyzed in this study: EET 92048, MIL 090657, and NWA 801. EET 92042 and EET 92062, paired meteorites of EET 92048, have been reported as less aqueously altered CR chondrites. For example, EET 92042 has been classified as CR2.5 based on bulk H, C, and N elemental and isotopic compositions by Alexander et al. (2013), and EET 92062 has been classified as CR2.8 based on its petrographic properties by Harju et al. (2014). MIL 090657 has experienced minimal thermal and aqueous processing and is reported as one of the most pristine CR chondrites (Davidson et al., 2019b). Based on the petrographic characteristics, Furukawa et al. (2019) reported that the subtype of NWA 801 would be 3.0 to 2.8, indicating that this meteorite experienced minimal aqueous processing in its parent body. Previous noble gas studies showed that solar wind noble gases are highly concentrated in NWA 801 (Nakashima et al., 2009; Matsuda et al., 2009). NWA 801 experienced terrestrial weathering that might have lowered the presolar silicate-to-oxide abundance ratio for fine-grained rims in NWA 801 than that for minimally altered CR chondrites (Sanghani et al., 2021).

2.2. Sample preparation

Thick sections of EET 92048 (EET 92048-PS and EET 92048-NG), MIL 090657 (MIL 090657-PS, MIL 090657-NG, and MIL 090657-HT), and NWA 801 (NWA 801-NG) were prepared for analysis. The samples named "meteorite-PS" and "meteorite-NG" were prepared for detailed petrological studies and noble gas analysis, respectively. MIL 090657-HT was prepared for a hydrothermal alteration experiment.

All thick sections (0.8–1.0 mm in thickness) were mounted on glass slides with glycol phthalate and polished. EET 92048-PS, MIL 090657-PS, MIL 090657-NG, and NWA 801-PS were coated with carbon (~20 nm in thickness) for scanning electron microscope (SEM) observations. After the SEM analysis, the carbon coatings were removed by polishing and cleaned with deionized water in an ultrasonic bath. Then, small pieces of the matrix for synchrotron radiation X-ray diffraction (S-XRD) analysis were extracted from EET 92048-PS, MIL 090657-NG, and NWA 801-PS using a craft knife under a binocular microscope.

Glycol phthalate on the samples for noble gas analysis (EET 92048-NG, MIL 090657-NG, MIL 090657-HT, and NWA 801-NG) was removed by washing in boiling acetone for 5 minutes several times. Since only the surface of the

thick chip samples were washed with acetone, it is likely that the noble gas and mineral compositions of the samples were almost unaffected by this procedure. MIL 090657-HT (~150 mg) was gently crushed into small grains (a few hundred μ m in size) at low-temperature using liquid nitrogen to cause brittle fracture of metals. The low-temperature condition might have enhanced adsorption of atmospheric Kr and Xe (see Section 4.7). The aliquot of crushed MIL 090657-HT was separated into two fractions: MIL 090657-HT (42 mg) for noble gas analysis and MIL 090657-HTaq (71 mg) for a hydrothermal treatment.

MIL 090657-HTaq was heated for 20 days at 150 °C with deionized water (2 ml) in a Teflon pressurized vessel (50 cm³). The internal water vapor pressure during the treatment is estimated to be 6.2 atm. The experimental conditions such as time and water/rock ratio are similar to the previous study for Ningqiang except for the temperature heated at 200 °C (Yamamoto et al., 2006). The heating temperature was set at 150 °C based on the heat-resistant temperature of the vessel. A grain of MIL 090657-HTaq (MIL-HTaq-g1) was embedded in epoxy resin on a glass plate, then polished and coated with carbon (~20 nm in thickness) for SEM observation. Another grain of MIL 090657-HTaq ~200 μ m in diameter (MIL-HTaq-g2) was picked up for S-XRD analysis.

2.3. Electron microscopy

Initial textural and mineralogical characterization was performed with an SEM (Hitachi S3400N at Tohoku University) and a field-emission SEM (JEOL JSM 7001F at Tohoku University) operated at 15 kV. EET 92048-NG, MIL 090657-HT, and NWA 801-NG with no carbon coating were observed using the SEM with low-vacuum conditions at 40 Pa. Mosaic backscattered electron (BSE) images of all sections are given in Fig. 1 and the supplementary material (Fig. A1).

Specific portions were extracted from MIL 090657-PS and MIL-HTaq-g1 using a focused ion beam (FIB) microsampling system (Thermo Fisher Scientific, Quanta 3D 200i) at Tohoku University. The portions were cut and lifted from the polished sections and mounted on TEM grids, then thinned to thicknesses of 100–200 nm using a Ga⁺ ion beam at 30 kV. The damaged layers formed on the ultrathin section surfaces were removed using a Ga⁺ ion beam at 5 kV.

2.4. TEM analysis

The ultrathin sections of MIL 090657-PS (MIL 090657-PS-F) and MIL-HTaq-g1 (MIL-HTaq-g1-F) were analyzed using a field emission transmission electron microscope (FE-TEM) (JEOL, JEM-2100F) at Tohoku University with an energy dispersive X-ray spectrometer (EDS) operated at 200 kV. TEM images were recorded using charge-coupled device (CCD) cameras and analyzed using the software package Gatan Digital Micrograph. Crystal structures were identified based on selected area electron diffraction (SAED) patterns. The SAED patterns were analyzed using the software package ReciPro (developed by Y. Seto). The



Fig. 1. BSE images of CR chondrites. (a) Mosaic image of EET92048-NG. (b) Framboidal magnetite in the fine-grained matrix of EET 92048-PS. (c) Smooth rim on a type I chondrule in EET 92048-PS. (d) Mosaic image of MIL 090657-NG. (e) Dark inclusion (surrounded by yellow dotted line) and chondrule fragments embedded in the fine-grained matrix of MIL 090657-PS. (f) Framboidal magnetite in the dark inclusion shown in the Fig. 1e. (g), (h) Type I chondrule in MIL 090657-PS surrounded by a smooth rim from which a FIB section (MIL 090657-PS-F) was extracted. (i) Smooth rim on a type I chondrule surrounded by a fine-grained matrix of NWA 801-NG. (k) Dark inclusion (surrounded by yellow dotted line) embedded in the fine-grained matrix of NWA 801-PS. (l) Smooth rim on a type I chondrule. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

two-dimensional (2D) SAED patterns were converted to the 1D diffraction patterns using the software package IPA Analyzer (Seto et al., 2010). We also obtained high-angle annular dark field scanning TEM (HAADF-STEM) images and X-ray elemental maps using the STEM-EDS system.

2.5. Synchrotron radiation X-ray diffraction analysis

The mineral compositions of individual meteorite matrices were analyzed with the S-XRD method (Nakamura et al., 2008), especially to identify the presence of phyllosilicates. The samples (\sim 100–250 µm in diameter) were

mounted on thin carbon fibers (~5 µm in diameter) using glycol phthalate, set in a Gandolfi camera, then exposed to synchrotron radiation X-rays ($\lambda = 2.161 \pm 0.001$ Å) at beam line 3A in the Photon Factory Institute of Material Science, High Energy Accelerator Research Organization (KEK; Tsukuba, Japan). Exposure times were 10–20 min for individual samples.

2.6. Noble gas analysis

All noble gas compositions were measured using a modified-VG5400/MS-III mass spectrometer at the Korea

Polar Research Institute (KOPRI; Incheon, Korea), following the procedure described in Nagao et al. (2008, 2016). The meteorite samples EET 92048-NG (130 mg), MIL 090657-NG (98.7 mg), NWA 801-NG (60.7 mg), MIL 090657-HT (42 mg), and MIL 090657-HTaq (41 mg) were individually wrapped with Al-foil and loaded into a sample holder in a noble gas purification line equipped with a noble gas extraction furnace using a Mo crucible placed in a Ta tube, which can be heated from the outside by radiation emitted from an electric resistance W-heater (Nagao et al., 2016). After loading the samples, the sample holder was preheated at 150 °C in ultrahigh vacuum for 24 h to remove adsorbed atmospheric gases from the sample surfaces, and the Mo crucible was degassed at 1850 °C to lower the blank level. The stepwise heating was performed from 400 °C to 1800 °C with 200 °C intervals. The heating duration was 30-40 min plus the time to reach the temperatures. The extracted gases were purified by two Ti-Zr getters and two SAES getters (NP10). He and Ne were separated from Ar, Kr, and Xe using a charcoal cold trap held at the temperature of liquid nitrogen. The He-Ne fraction was further purified using two charcoal cold traps to reduce background ⁴⁰Ar and CO₂. He and Ne were separated by a sintered stainless-steel trap at 20 K, and individually admitted into the mass spectrometer. After the He and Ne analyses, reserved Ar, Kr, and Xe were desorbed from the charcoal trap. Then Kr and Xe were adsorbed onto the sintered stainless-steel trap at 95 K. After Ar analysis, Kr was desorbed from the trap at 160 K. Xenon was finally released at 230 K.

Sensitivities and mass discrimination correction factors of the mass spectrometer were calibrated by measuring atmospheric noble gases with known isotopic ratios and amounts, and a ³He-⁴He mixture with ³He/⁴He = 1.71 \times 10⁻⁴ (Nagao et al., 2010). Blank levels for noble gases at all temperature steps were measured, and blank corrections were applied to all noble gas data. Blank contributions to sample gas amounts were <2% for ⁴He, <0.8%for ²⁰Ne, <0.5% for ³⁶Ar, <0.8% for ⁸⁴Kr, and <0.1% for ¹³²Xe for the all analysis except for the 1800 °C steps. Blank corrections for the 1800 °C steps amounted to <66% for ⁴He (except for MIL 090657-HTaq, which was indistinguishable from the blank level), <5.6% for $^{20}Ne,$ <58% for $^{36}Ar,$ <36% for $^{84}Kr,$ and <4.6% for $^{132}Xe.$ Isobaric interferences ($^{40}Ar^{++}$ on mass 20 and CO_2^{++} on mass 22) were subtracted based on the peak intensities of ${}^{40}Ar^+$ and CO_2^+ measured before and after each Ne isotopic ratio measurement, following the procedure described in Osawa (2004). The ${}^{40}\text{Ar}^{++}/{}^{40}\text{Ar}^{+}$ and CO_2^{++}/CO_2^{+} ratios for the isobaric corrections were determined before the sample analysis. The contributions of $^{40}\mathrm{Ar}^{++}$ and CO_2^{++} were less than 1.3% and 2.1% of the measured peak intensities on mass 20 and mass 22, respectively. Statistical uncertainties (1σ) are given for noble gas isotopic ratios of measured blanks, samples, and standard gases. The sensitivity errors are assumed to be 10% based on the reproducibility of the calibration measurements.

3. RESULTS

3.1. Petrologies and mineralogies of CR chondrites

3.1.1. Textural characteristics of CR chondrites

Type I chondrules with Mg# ≥90 (=molar MgO/ (MgO + FeO)%) typically 1–2 mm in diameter, chondrule fragments, and Fe-Ni metal grains are embedded in fine-grained matrices (Fig. 1a, d, e, and j). Fe-Ni metal grains are mostly located on the periphery or in the interior of the chondrules, while some metal grains are embedded in the matrices (Fig. 1a, d, and j). "Smooth rims", which are probably products of aqueous alteration (Harju et al., 2014), were identified around some chondrules in all meteorites and are up to 30 µm in thickness (Fig. 1c, g, h, i, and l). One smooth rim in MIL 090657 is surrounded by a fine-grained rim (Fig. 1i). Some dark inclusions were found in MIL 090657 (Fig. 1e and f) and NWA 801 (Fig. 1k). Alteration of mafic phenocrysts, which is commonly observed in highly altered CR chondrites (Harju et al., 2014), is not identified from any samples in this study. Framboidal magnetite produced by aqueous alteration (Harju et al., 2014) is not observed in the matrices of MIL 090657 and NWA 801 but is identified in the EET 92048 matrix (Fig. 1b). The framboidal magnetites are locally distributed in some parts of the EET 92048 matrix but are scarce in the other part. This is consistent with the observation of a paired meteorite EET 92042 (Harju et al., 2014). Some dark inclusions in MIL 090657 contain abundant framboidal magnetite (Fig. 1f). In summary, overall textural characteristics of the three CR chondrites are generally consistent with previous descriptions of CR chondrites (e.g., Harju et al., 2014; Abreu, 2016).

3.1.2. Mineral compositions of CR chondrite matrices

The mineral compositions of the CR chondrite matrices were obtained with S-XRD analyses. The analysis was performed on small pieces (\sim 100–250 µm) of representative matrix portions extracted from EET 92048, MIL 090657, and NWA 801.

The S-XRD patterns are shown in Fig. 2. The EET 92048 matrix consists of olivine, pyroxene, kamacite, and serpentine with minor magnetite and pyrrhotite (Fig. 2a). A small diffraction peak at 11.5° and a peak at 23° are probably from tochilinite, though it was not observed with SEM analysis of EET 92048-PS. The MIL 090657 matrix consists of olivine, pyroxene, kamacite with minor magnetite and pyrrhotite. Small diffraction peaks from serpentine and tochilinite-serpentine mix layers might be present (Fig. 2b). The NWA 801 matrix consists of olivine, pyroxene, kamacite, and calcite (Fig. 2c). In summary, hydrous minerals including serpentine are present in the EET 92048 matrix, whereas they are minor or not identified in the MIL 090657 and NWA 801 matrices.

3.1.3. Sub-micron structure of the matrix and a smooth rim in MIL 090657

One FIB section (MIL 090657-PS-F) was extracted from a boundary between the fine-grained matrix and a smooth



Fig. 2. Synchrotron X-ray diffraction patterns of (a) EET 92048 matrix, (b) MIL 090657 matrix, (c) NWA 801 matrix, and (d) a grain of MIL 090657-HT after the hydrothermal treatment. Abbreviations: O, olivine; Px, low-Ca pyroxene; Kam, kamacite; Mgt, magnetite; IO, iron oxide; S, serpentine; T, tochilinite; M, tochilinite-serpentine mix layer; Py, pyrrhotite; Cal, calcite.

rim of a chondrule of MIL 090657-PS (Fig. 1h and 3a). Then, the sub-micron structures of the matrix and the smooth rim were studied with TEM (Fig. 3).

Amorphous silicate grains (a few hundreds of nm in size) containing numerous sulfide nanograins were commonly observed in the matrix (Fig. 3b and c). The texture of the amorphous silicate grains is similar to that of GEMS (glass with embedded metal and sulfides) frequently found in chondritic porous interplanetary dust particles and chondritic porous micrometeorites (e.g., Bradley, 1994; Keller and Messenger, 2011; Noguchi et al., 2015). Phyllosilicates (Fig. A2; supplementary material), sulfide grains, and carbon-rich materials (Fig. 3d and e) were also identified in the matrix.

The smooth rim part consists of two layers with a sharp boundary and has vertically slipped textures (Fig. 3a and f). The chemical composition of the inner layer adjacent to the chondrule mesostasis is FeO-rich and MgO-poor compared to the outer layer (Fig. 3f and g). This is consistent with the "two-tone" rims reported in Harju et al. (2014). Based on the high-resolution TEM images, both the inner and outer layers have weakly crystallized structures (Fig. 3h and i). Fig. 4 shows the 2D SAED patterns and 1D-converted SAED patterns of the inner and outer layers of the smooth rim. Both the diffraction patterns display three broad peaks around *d* spacings of ~0.16, ~0.26, and ~0.45 nm.

3.2. Petrology and mineralogy of MIL 090657 after hydrothermal treatment

Fig. 5a and 5b show BSE images of MIL-HTaq-g1, a grain of hydrothermally treated MIL 090657. Most Fe-Ni metals were oxidized, and iron oxide shells were formed on the periphery of MIL-HTaq-g1. Alteration of mafic phenocryst is not observed.



Fig. 3. TEM images and STEM-EDS maps of a FIB section (MIL 090657-PS-F) extracted from a boundary between a fine-grained matrix and a smooth rim on a type I chondrule in MIL 090657-PS (Fig. 1h). (a) Bright-field (BF) TEM mosaic of the FIB section. (b) BF TEM image showing a GEMS-like grain in the fine-grained matrix. (c) 2D SAED pattern of the GEMS-like grain in Fig. 3b, showing amorphous halo pattern. The SAED pattern was taken from the area of the yellow circle in the small panel. (d) HAADF-STEM image and (e) combined STEM-EDS map for Fe (red), Mg (green), C (blue), and S (yellow) of the same field in the fine-grained matrix. (f) HAADF-STEM image and (g) combined STEM-EDS map for Fe (red), Mg (green), Si (blue), and S (yellow) of the same field of the smooth rim. The yellow circles represent areas from which the SAED patterns of the inner and outer rims in Fig. 4 were taken. (h) High-resolution BF TEM image of the inner layer of the smooth rim. (i) High-resolution BF TEM image of the outer layer of the smooth rim. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The mineral composition of another grain of treated MIL 090657 (MIL-HTaq-g2) was analyzed by the S-XRD method. The X-ray diffraction pattern shows that MIL-HTaq-g2 consists of iron oxide, olivine, pyroxene, serpentine, hematite, and kamacite (Fig. 2d). The sharp diffraction peaks of phyllosilicates in MIL-HTaq-g2 suggest the formation of phyllosilicates during the hydrothermal treatment.

One FIB section (MIL-HTaq-g1-F) was extracted from a boundary between a matrix and a chondrule of MIL-HTaq-g1 (Fig. 5b). A high-resolution bright-field TEM image of the fine-grained materials in Fig. 5c is shown in Fig. 5d. GEMS-like amorphous silicates are not identified in the matrix but poorly crystallized fibrous phyllosilicates are commonly distributed (Fig. 5d). Carbon-rich materials and sulfide grains were also observed in the matrix (Fig. 5e and f).

3.3. Noble gas

Concentrations and isotopic ratios of He, Ne, Ar, Kr, and Xe in the samples are given in the Tables 1–3. Amounts of ⁸⁴Kr and ¹³²Xe released in the 800 °C fraction of MIL 090657-NG were not obtained due to analytical trouble. The following indices are for denoting the noble gas components: C (cosmogenic), trap (trapped component), SW (solar wind), FSW (fractionated solar wind), and air (Earth atmosphere). The elemental and isotopic ratios of the noble gas components used in this study are summarized in Tables 4 and 5, respectively.



Two- and one-dimensional SAED patterns of smooth rim

Fig. 4. (a) and (b) 2D SAED patterns of the inner and outer layers of a smooth rim in MIL 090657-PS-F (Fig. 3h and i). Both layers show three weak rings around *d*-spacings of 0.45, 0.26, and 0.16 nm. (c) 1D-converted SAED patterns of the inner and outer layers of the smooth rim.



Fig. 5. (a) BSE image of MIL-HTaq-g1, a grain of hydrothermally treated MIL 090657-HT. (b) BSE image of an area in a yellow square in Fig. 5a. (c) Bright-field (BF) TEM mosaic of an FIB section (MIL-HTaq-g1-F) extracted from a boundary between a fine-grained matrix and a chondrule of MIL-HTaq-g1. (d) High-resolution BF TEM image of a fine-grained matrix of MIL-HTaq-g1-F. The lower right panel "d1" shows an enlarged image of poorly crystallized phyllosilicates. (e) HAADF-STEM image and (f) combined STEM-EDS map for Fe (red), Mg (green), C (blue), and S (yellow) of the same area of the fine-grained matrix of MIL-HTaq-g1-F. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Cosmogenic noble gases are produced by interactions between meteoritic materials and galactic cosmic rays (GCR) and/or solar energetic particles (also known as solar cosmic rays, SCR) via spallation and neutron-capture reactions. Such reactions mainly occur during transit to the Earth after ejection from a parent body as a meter-sized meteoroid (Wieler, 2002b).

3.3.1. Neon

Fig. 6 is the Ne three-isotope diagrams for EET 92048-NG, MIL 090657-NG, NWA 801-NG, MIL 090657-HT, and MIL 090657-HTaq.

The Ne isotopic ratios of NWA 801-NG shift from the solar wind Ne (between SW and FSW) to the cosmogenic (GCR) Ne with increasing extraction temperatures (Fig. 6a), indicating that NWA 801-NG contains solar wind Ne. On the other hand, the Ne isotopic ratios of EET 92048-NG and MIL 090657-NG shift from the primordial components (Q and HL) to the cosmogenic compositions. The absence of solar wind noble gas signature in MIL 090657 is consistent with the absence of implanted solar wind noble gases in MIL 090657 chondrules (Roth and Leya, 2018). While Ne isotopic ratios of MIL 090657-NG and MIL 090657-HT in the 400 °C fractions plot near

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

EET 92048-NG	⁴ He	$^{3}\text{He}/^{4}\text{He}(10^{-4})$	²⁰ Ne	²⁰ Ne/ ²² Ne	²¹ Ne/ ²² Ne	³⁶ Ar	³⁸ Ar/ ³⁶ Ar	⁴⁰ Ar/ ³⁶ Ar
400 °C	1839	7.95 ± 0.03	8.74	9.226 ± 0.050	0.1095 ± 0.0006	3.71	0.1900 ± 0.0004	56.660 ± 0.032
600 °C	1274	35.10 ± 0.14	2.97	8.079 ± 0.042	0.1547 ± 0.0010	3.27	0.1908 ± 0.0005	174.349 ± 0.088
800 °C	1716	22.46 ± 0.08	6.71	6.385 ± 0.034	0.2605 ± 0.0011	8.09	0.1937 ± 0.0004	131.181 ± 0.068
1000 °C	811	14.14 ± 0.06	11.15	5.390 ± 0.032	0.3842 ± 0.0015	104.62	0.1891 ± 0.0004	1.829 ± 0.001
1200 °C	83	65.22 ± 0.25	2.66	4.044 ± 0.029	0.5448 ± 0.0023	63.37	0.1890 ± 0.0004	0.903 ± 0.001
1400 °C	26	70.42 ± 0.30	0.44	1.649 ± 0.010	0.8126 ± 0.0032	9.17	0.1932 ± 0.0004	3.072 ± 0.007
1600 °C	5.6	566.55 ± 2.41	0.25	0.871 ± 0.005	0.8951 ± 0.0038	0.65	0.2368 ± 0.0005	12.897 ± 0.176
1800 °C	0.03	218.19 ± 81.31	0.04	0.885 ± 0.039	0.8810 ± 0.0059	0.03	0.2573 ± 0.0031	66.517 ± 9.880
total	5755	20.81 ± 0.04	32.95	5.788 ± 0.019	0.3695 ± 0.0006	192.92	0.1897 ± 0.0002	11.029 ± 0.004
MIL 090657-NG	⁴ He	$^{3}\text{He}/^{4}\text{He}(10^{-4})$	²⁰ Ne	²⁰ Ne/ ²² Ne	²¹ Ne/ ²² Ne	³⁶ Ar	³⁸ Ar/ ³⁶ Ar	⁴⁰ Ar/ ³⁶ Ar
400 °C	3550	5.21 ± 0.03	13.80	9.879 ± 0.060	0.0548 ± 0.0002	4.49	0.1917 ± 0.0004	45.365 ± 0.020
600 °C	1710	25.93 ± 0.13	3.49	7.994 ± 0.044	0.1332 ± 0.0005	4.69	0.1904 ± 0.0004	154.660 ± 0.073
800 °C	2190	15.97 ± 0.06	9.17	7.194 ± 0.041	0.1742 ± 0.0008	9.48	0.1935 ± 0.0004	43.430 ± 0.021
1000 °C	825	14.66 ± 0.07	11.00	6.641 ± 0.037	0.2115 ± 0.0009	96.36	0.1888 ± 0.0004	0.700 ± 0.001
1200 °C	137	32.57 ± 0.24	4.44	5.832 ± 0.031	0.2798 ± 0.0013	110.09	0.1887 ± 0.0004	0.138 ± 0.001
1400 °C	33	43.69 ± 0.19	0.51	1.819 ± 0.013	0.6607 ± 0.0026	11.12	0.1905 ± 0.0004	0.533 ± 0.008
1600 °C	3.9	445.69 ± 2.05	0.10	0.843 ± 0.008	0.7582 ± 0.0033	0.84	0.2096 ± 0.0004	3.001 ± 0.187
1800 °C	0.06	19.24 ± 9.25	0.03	0.688 ± 0.070	0.7519 ± 0.0072	0.01	0.2601 ± 0.0141	n.d. \pm n.d.
total	8448	13.91 ± 0.03	42.53	7.132 ± 0.024	0.2054 ± 0.0003	237.08	0.1892 ± 0.0002	6.037 ± 0.002
NWA 801-NG	⁴ He	$^{3}\text{He}/^{4}\text{He}(10^{-4})$	²⁰ Ne	²⁰ Ne/ ²² Ne	21 Ne/ 22 Ne	³⁶ Ar	$^{38}Ar/^{36}Ar$	⁴⁰ Ar/ ³⁶ Ar
400 °C	455,591	3.24 ± 0.01	189.88	12.454 ± 0.069	0.0420 ± 0.0002	5.26	0.1897 ± 0.0004	47.517 ± 0.026
600 °C	301,433	3.38 ± 0.03	204.25	12.598 ± 0.069	0.0384 ± 0.0002	5.04	0.1934 ± 0.0004	49.270 ± 0.020
800 °C	162,906	4.23 ± 0.02	392.68	12.304 ± 0.062	0.0480 ± 0.0002	6.38	0.2001 ± 0.0004	36.589 ± 0.016
1000 °C	51,705	4.06 ± 0.04	970.02	11.914 ± 0.062	0.0541 ± 0.0002	34.39	0.1924 ± 0.0004	2.281 ± 0.005
1200 °C	1799	15.45 ± 0.09	305.71	10.828 ± 0.062	0.0915 ± 0.0004	76.63	0.1917 ± 0.0004	0.456 ± 0.002
1400 °C	56	98.76 ± 0.37	53.05	8.639 ± 0.067	0.2395 ± 0.0010	22.84	0.1950 ± 0.0004	0.658 ± 0.006
1600 °C	5.9	848.53 ± 4.10	8.91	4.693 ± 0.025	0.5247 ± 0.0021	1./6	0.2327 ± 0.0006	4.003 ± 0.145
1800 °C	0.34	81.49 ± 4.99	1.37	3.340 ± 0.031	0.6330 ± 0.0027	0.27	0.2377 ± 0.0007	52.234 ± 2.619
total	975,495	3.53 ± 0.01	2120	$11./14 \pm 0.033$	0.0689 ± 0.0001	152.0	0.1932 ± 0.0002	5.776 ± 0.005
MIL 090657-HT	⁴ He	$^{3}\text{He}/^{4}\text{He}(10^{-4})$	²⁰ Ne	²⁰ Ne/ ²² Ne	21 Ne/ 22 Ne	³⁶ Ar	$^{38}\text{Ar}/^{36}\text{Ar}$	40 Ar/ 36 Ar
400 °C	3000	2.50 ± 0.03	13.95	10.118 ± 0.031	0.0467 ± 0.0002	1.99	0.1921 ± 0.0005	41.808 ± 0.089
600 °C	1450	15.76 ± 0.19	5.80	9.233 ± 0.030	0.0826 ± 0.0008	4.23	0.1910 ± 0.0005	74.612 ± 0.141
800 °C	1776	26.55 ± 0.33	3.63	7.390 ± 0.022	0.1780 ± 0.0008	5.15	0.1923 ± 0.0005	140.782 ± 0.268
1000 °C	1318	11.47 ± 0.14	9.86	6.840 ± 0.024	0.2098 ± 0.0007	36.20	0.1891 ± 0.0005	5.856 ± 0.011
1200 °C	165	48.74 ± 0.57	6.94	6.480 ± 0.018	0.2366 ± 0.0007	128.24	0.1884 ± 0.0004	0.198 ± 0.001
1400 °C	39	46.91 ± 0.57	0.82	2.052 ± 0.008	0.6297 ± 0.0021	20.64	0.1896 ± 0.0005	0.717 ± 0.007
1600 °C	22	254.23 ± 3.06	0.14	0.863 ± 0.006	0.7530 ± 0.0028	0.74	0.2308 ± 0.0006	3.499 ± 0.194
1800 °C	1.4	1032.0 ± 34.6	0.02	0.867 ± 0.057	0.7733 ± 0.0088	0.05	0.4513 ± 0.0048	37.706 ± 4.405
total	7770	14.10 ± 0.09	41.15	7.365 ± 0.013	0.2049 ± 0.0003	197.25	0.1891 ± 0.0003	7.001 ± 0.008
MIL 090657-HTaq	⁴ He	$^{3}\text{He}/^{4}\text{He}(10^{-4})$	²⁰ Ne	²⁰ Ne/ ²² Ne	²¹ Ne/ ²² Ne	³⁶ Ar	$^{38}\text{Ar}/^{36}\text{Ar}$	⁴⁰ Ar/ ³⁶ Ar
400 °C	137	50.85 ± 0.59	0.15	5.035 ± 0.031	0.3413 ± 0.0039	0.85	0.1892 ± 0.0007	50.826 ± 0.126
600 °C	297	46.68 ± 0.55	0.17	5.210 ± 0.049	0.2841 ± 0.0047	1.06	0.1887 ± 0.0005	45.510 ± 0.108
800 °C	1252	32.16 ± 0.39	2.16	6.788 ± 0.032	0.1973 ± 0.0013	4.22	0.1911 ± 0.0005	40.132 ± 0.081
1000 °C	896	10.66 ± 0.13	6.02	5.624 ± 0.021	0.3041 ± 0.0009	17.39	0.1894 ± 0.0005	3.386 ± 0.011
1200 °C	6l	18.23 ± 0.24	2.65	4.970 ± 0.026	$0.3/82 \pm 0.0017$	55.52	0.1881 ± 0.0004	0.402 ± 0.003
1400 °C	15	$29./1 \pm 0.40$	0.5/	$2.3/6 \pm 0.010$	0.0295 ± 0.0017	22.24	0.1885 ± 0.0004	0.418 ± 0.007
1000 °C	1.1	150.46 ± 5.92	0.11	$0.81/\pm0.006$	0.7473 ± 0.0032	1.10	$0.191/\pm 0.0005$	0.707 ± 0.129
1000 °C	0	$11.0. \pm 11.0.20$	0.02	0.922 ± 0.008	0.7390 ± 0.0077	0.10	$0.1943 \pm 0.000/$	$1/.232 \pm 1.3/3$
totai	2038	27.21 ± 0.20	11.85	4.993 ± 0.013	$0.30/0 \pm 0.0006$	102.58	0.1880 ± 0.0003	3.437 ± 0.005

Concentrations are given in 10^{-8} cm³ STP g⁻¹. Uncertainties of the gas concentrations are assumed to be 10%. Uncertainties of the isotopic ratios are 1 σ .

Ne-Q, that of MIL 090657-HTaq in the same temperature fraction plot between Ne-HL and cosmogenic Ne (Fig. 6). The relatively ²²Ne-rich isotopic ratio in the 600 °C fraction suggests a presence of the G component in MIL 090657. In summary, EET 92048 and MIL 090657 are solar-gas-poor

CR chondrites, and NWA 801 is a solar-gas-rich CR chondrite. Furthermore, the hydrothermal treatment caused a loss of isotopically Q-like Ne released at low temperature.

The concentrations of 20 Ne_{trap} and 21 Ne_C in the bulk CR chondrites are calculated from the following equations:

Table 2 Concentrations and isotopic ratios of Kr.

EET 92048-NG	⁸⁴ Kr	⁷⁸ Kr/ ⁸⁴ Kr	⁸⁰ Kr/ ⁸⁴ Kr	⁸² Kr/ ⁸⁴ Kr	⁸³ Kr/ ⁸⁴ Kr	⁸⁶ Kr/ ⁸⁴ Kr
400 °C	465	0.00608 ± 0.00008	0.0398 ± 0.0003	0.2021 ± 0.0009	0.2019 ± 0.0006	0.3095 ± 0.0014
600 °C	255	0.00615 ± 0.00020	0.0401 ± 0.0004	0.2028 ± 0.0007	0.2028 ± 0.0010	0.3101 ± 0.0008
800 °C	489	0.00631 ± 0.00012	0.0423 ± 0.0007	0.2032 ± 0.0008	0.2039 ± 0.0006	0.3125 ± 0.0008
1000 °C	2984	0.00609 ± 0.00006	0.0400 ± 0.0001	0.2043 ± 0.0006	0.2044 ± 0.0005	0.3165 ± 0.0008
1200 °C	2219	0.00611 ± 0.00006	0.0400 ± 0.0002	0.2043 ± 0.0006	0.2040 ± 0.0004	0.3146 ± 0.0011
1400 °C	624	0.00616 ± 0.00009	0.0400 ± 0.0003	0.2039 ± 0.0007	0.2032 ± 0.0006	0.3129 ± 0.0009
1600 °C	57	0.00612 ± 0.00013	0.0406 ± 0.0006	0.2010 ± 0.0020	0.2039 ± 0.0017	0.3095 ± 0.0022
1800 °C	3.4	0.00643 ± 0.00081	0.0425 ± 0.0028	0.2013 ± 0.0115	0.2068 ± 0.0113	0.3153 ± 0.0120
total	7097	0.00612 ± 0.00004	0.0401 ± 0.0001	0.2040 ± 0.0003	0.2039 ± 0.0003	0.3146 ± 0.0005
MIL 090657-NG	⁸⁴ Kr	⁷⁸ Kr/ ⁸⁴ Kr	⁸⁰ Kr/ ⁸⁴ Kr	⁸² Kr/ ⁸⁴ Kr	⁸³ Kr/ ⁸⁴ Kr	⁸⁶ Kr/ ⁸⁴ Kr
400 °C	570	0.00607 ± 0.00009	0.0398 ± 0.0002	0.2019 ± 0.0011	0.2025 ± 0.0007	0.3099 ± 0.0013
600 °C	380	0.00606 ± 0.00014	0.0399 ± 0.0004	0.2022 ± 0.0015	0.2023 ± 0.0010	0.3099 ± 0.0013
800 °C	-	0.00614 ± 0.00029	0.0406 ± 0.0009	0.2012 ± 0.0022	0.2015 ± 0.0027	0.3109 ± 0.0024
1000 °C	2830	0.00607 ± 0.00010	0.0394 ± 0.0004	0.2023 ± 0.0005	0.2020 ± 0.0016	0.3110 ± 0.0013
1200 °C	3380	0.00608 ± 0.00015	0.0396 ± 0.0002	0.2035 ± 0.0012	0.2034 ± 0.0009	0.3123 ± 0.0009
1400 °C	745	0.00610 ± 0.00010	0.0396 ± 0.0005	0.2022 ± 0.0009	0.2031 ± 0.0014	0.3114 ± 0.0016
1600 °C	87	0.00612 ± 0.00019	0.0397 ± 0.0008	0.2021 ± 0.0015	0.2027 ± 0.0020	0.3100 ± 0.0023
1800 °C	1.4	0.00734 ± 0.00158	0.0443 ± 0.0061	0.2051 ± 0.0195	0.2005 ± 0.0199	0.3112 ± 0.0288
Total	_	_	_	-	_	_
NWA 801-NG	⁸⁴ Kr	⁷⁸ Kr/ ⁸⁴ Kr	⁸⁰ Kr/ ⁸⁴ Kr	⁸² Kr/ ⁸⁴ Kr	⁸³ Kr/ ⁸⁴ Kr	⁸⁶ Kr/ ⁸⁴ Kr
400 °C	464	0.00600 ± 0.00010	0.0403 ± 0.0004	0.2026 ± 0.0011	0.2012 ± 0.0007	0.3072 ± 0.0013
600 °C	118	0.00626 ± 0.00036	0.0413 ± 0.0008	0.2005 ± 0.0029	0.2018 ± 0.0011	0.3080 ± 0.0023
800 °C	111	0.00641 ± 0.00031	0.0417 ± 0.0007	0.2026 ± 0.0022	0.2020 ± 0.0021	0.3070 ± 0.0037
1000 °C	678	0.00603 ± 0.00007	0.0397 ± 0.0003	0.2020 ± 0.0006	0.2028 ± 0.0011	0.3123 ± 0.0016
1200 °C	1976	0.00598 ± 0.00010	0.0396 ± 0.0002	0.2030 ± 0.0009	0.2028 ± 0.0008	0.3113 ± 0.0011
1400 °C	872	0.00614 ± 0.00009	0.0399 ± 0.0002	0.2033 ± 0.0006	0.2034 ± 0.0009	0.3129 ± 0.0008
1600 °C	72	0.00663 ± 0.00029	0.0405 ± 0.0012	0.2030 ± 0.0022	0.2030 ± 0.0019	0.3074 ± 0.0037
1800 °C	12	0.00751 ± 0.00108	0.0429 ± 0.0022	0.2051 ± 0.0061	0.2043 ± 0.0045	0.3078 ± 0.0072
total	4303	0.00606 ± 0.00005	0.0399 ± 0.0001	0.2028 ± 0.0005	0.2027 ± 0.0005	0.3111 ± 0.0006
MIL 090657-HT	⁸⁴ Kr	⁷⁸ Kr/ ⁸⁴ Kr	⁸⁰ Kr/ ⁸⁴ Kr	82 Kr/ 84 Kr	⁸³ Kr/ ⁸⁴ Kr	⁸⁶ Kr/ ⁸⁴ Kr
400 °C	862	0.00613 ± 0.00013	0.0395 ± 0.0004	0.2023 ± 0.0011	0.2015 ± 0.0008	0.3058 ± 0.0009
600 °C	501	0.00600 ± 0.00012	0.0393 ± 0.0004	0.2005 ± 0.0014	0.2005 ± 0.0011	0.3073 ± 0.0015
800 °C	507	0.00616 ± 0.00013	0.0405 ± 0.0007	0.2019 ± 0.0013	0.2023 ± 0.0010	0.3075 ± 0.0018
1000 °C	1408	0.00603 ± 0.00010	0.0397 ± 0.0002	0.2027 ± 0.0006	0.2027 ± 0.0008	0.3127 ± 0.0010
1200 °C	3821	0.00621 ± 0.00017	0.0395 ± 0.0005	0.2031 ± 0.0010	0.2018 ± 0.0011	0.3102 ± 0.0020
1400 °C	1109	0.00613 ± 0.00011	0.0394 ± 0.0002	0.2024 ± 0.0013	0.2028 ± 0.0006	0.3118 ± 0.0012
1600 °C	90	0.00622 ± 0.00031	0.0398 ± 0.0006	0.2015 ± 0.0017	0.2038 ± 0.0018	0.3108 ± 0.0028
1800 °C	7	0.00528 ± 0.00093	0.0386 ± 0.0022	0.2019 ± 0.0097	0.2035 ± 0.0089	0.3007 ± 0.0132
total	8304	0.00615 ± 0.00008	0.0396 ± 0.0002	0.2026 ± 0.0005	0.2020 ± 0.0005	0.3100 ± 0.0010
MIL 090657-HTaq	⁸⁴ Kr	⁷⁸ Kr/ ⁸⁴ Kr	⁸⁰ Kr/ ⁸⁴ Kr	⁸² Kr/ ⁸⁴ Kr	⁸³ Kr/ ⁸⁴ Kr	⁸⁶ Kr/ ⁸⁴ Kr
400 °C	109	0.00617 ± 0.00025	0.0405 ± 0.0007	0.2046 ± 0.0019	0.2020 ± 0.0027	0.3083 ± 0.0046
600 °C	92	0.00631 ± 0.00023	0.0397 ± 0.0004	0.2037 ± 0.0030	0.2032 ± 0.0026	0.3086 ± 0.0028
800 °C	324	0.00613 ± 0.00014	0.0400 ± 0.0005	0.2026 ± 0.0018	0.2036 ± 0.0015	0.3131 ± 0.0022
1000 °C	668	0.00600 ± 0.00006	0.0393 ± 0.0004	0.2008 ± 0.0011	0.2018 ± 0.0011	0.3115 ± 0.0009
1200 °C	1657	0.00604 ± 0.00011	0.0395 ± 0.0003	0.2022 ± 0.0008	0.2019 ± 0.0007	0.3101 ± 0.0014
1400 °C	912	0.00615 ± 0.00023	0.0392 ± 0.0003	0.2014 ± 0.0017	0.2014 ± 0.0008	0.3085 ± 0.0019
1600 °C	67	0.00605 ± 0.00029	0.0389 ± 0.0013	0.2035 ± 0.0020	0.1997 ± 0.0043	0.3114 ± 0.0038
1800 °C	14	0.00584 ± 0.00057	0.0401 ± 0.0027	0.2037 ± 0.0071	0.2026 ± 0.0035	0.3101 ± 0.0088
total	3843	0.00608 ± 0.00007	0.0394 ± 0.0002	0.2019 ± 0.0006	0.2019 ± 0.0004	0.3101 ± 0.0008

Concentrations are given in 10^{-12} cm³ STP g⁻¹. Uncertainties of the gas concentrations are assumed to be 10%. Uncertainties of the isotopic ratios are 1 σ .

Table 3Concentrations and isotopic ratios of Xe.

EET 92048-NG	¹³² Xe	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
400 °C	380	0.00446 ± 0.00014	0.00395 ± 0.00005	0.0811 ± 0.0003	1.048 ± 0.005	0.1611 ± 0.0008	0.8153 ± 0.0021	0.3797 ± 0.0013	0.3197 ± 0.0012
600 °C	163	0.00466 ± 0.00014	0.00411 ± 0.00005	0.0831 ± 0.0004	1.058 ± 0.005	0.1621 ± 0.0012	0.8198 ± 0.0031	0.3802 ± 0.0018	0.3221 ± 0.0012
800 °C	293	0.00479 ± 0.00013	0.00412 ± 0.00008	0.0833 ± 0.0005	1.041 ± 0.004	0.1627 ± 0.0010	0.8217 ± 0.0026	0.3921 ± 0.0015	0.3389 ± 0.0013
1000 °C	1868	0.00472 ± 0.00013	0.00408 ± 0.00008	0.0835 ± 0.0004	1.032 ± 0.003	0.1635 ± 0.0009	0.8196 ± 0.0020	0.3860 ± 0.0009	0.3291 ± 0.0011
1200 °C	1650	0.00462 ± 0.00012	0.00410 ± 0.00007	0.0831 ± 0.0003	1.031 ± 0.004	0.1632 ± 0.0008	0.8206 ± 0.0026	0.3812 ± 0.0010	0.3224 ± 0.0010
1400 °C	744	0.00464 ± 0.00013	0.00410 ± 0.00010	0.0830 ± 0.0006	1.034 ± 0.003	0.1626 ± 0.0009	0.8203 ± 0.0025	0.3788 ± 0.0011	0.3193 ± 0.0015
1600 °C	91	0.00460 ± 0.00015	0.00409 ± 0.00012	0.0830 ± 0.0007	1.040 ± 0.005	0.1623 ± 0.0009	0.8212 ± 0.0058	0.3797 ± 0.0032	0.3197 ± 0.0023
1800 °C	4.9	0.00473 ± 0.00073	0.00431 ± 0.00050	0.0833 ± 0.0029	1.062 ± 0.014	0.1610 ± 0.0044	0.8263 ± 0.0008	0.3816 ± 0.0068	0.3266 ± 0.0046
total	5193	0.00466 ± 0.00006	0.00408 ± 0.00004	0.0831 ± 0.0002	1.035 ± 0.002	0.1630 ± 0.0005	0.8199 ± 0.0012	0.3830 ± 0.0005	0.3251 ± 0.0006
MIL 090657-NG	¹³² Xe	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
400 °C	526	0.00442 ± 0.00013	0.00394 ± 0.00007	0.0812 ± 0.0005	1.031 ± 0.003	0.1608 ± 0.0009	0.8146 ± 0.0022	0.3799 ± 0.0014	0.3205 ± 0.0008
600 °C	261	0.00459 ± 0.00013	0.00408 ± 0.00009	0.0826 ± 0.0007	1.039 ± 0.003	0.1620 ± 0.0011	0.8208 ± 0.0030	0.3791 ± 0.0020	0.3213 ± 0.0011
800 °C	-	0.00496 ± 0.00020	0.00418 ± 0.00034	0.0827 ± 0.0014	1.027 ± 0.011	0.1617 ± 0.0035	0.8214 ± 0.0080	0.3977 ± 0.0068	0.3435 ± 0.0024
1000 °C	1660	0.00476 ± 0.00018	0.00408 ± 0.00009	0.0832 ± 0.0006	1.041 ± 0.004	0.1624 ± 0.0008	0.8198 ± 0.0043	0.3842 ± 0.0019	0.3283 ± 0.0015
1200 °C	2210	0.00468 ± 0.00020	0.00409 ± 0.00008	0.0831 ± 0.0005	1.035 ± 0.004	0.1623 ± 0.0011	0.8215 ± 0.0024	0.3807 ± 0.0010	0.3211 ± 0.0018
1400 °C	883	0.00460 ± 0.00013	0.00405 ± 0.00012	0.0830 ± 0.0005	1.034 ± 0.004	0.1629 ± 0.0009	0.8204 ± 0.0031	0.3804 ± 0.0016	0.3205 ± 0.0009
1600 °C	162	0.00467 ± 0.00015	0.00413 ± 0.00011	0.0827 ± 0.0005	1.036 ± 0.005	0.1624 ± 0.0010	0.8232 ± 0.0047	0.3806 ± 0.0011	0.3205 ± 0.0014
1800 °C	2.3	0.00471 ± 0.00124	0.00417 ± 0.00090	0.0826 ± 0.0052	1.079 ± 0.027	0.1619 ± 0.0037	0.8073 ± 0.0016	0.3931 ± 0.0093	0.3319 ± 0.0132
total	_	_	-	_	_	_	_	_	_
NWA 801-NG	¹³² Xe	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
400 °C	239	0.00381 ± 0.00014	0.00356 ± 0.00051	0.0740 ± 0.0006	1.016 ± 0.005	0.1531 ± 0.0012	0.7946 ± 0.0034	0.3839 ± 0.0019	0.3258 ± 0.0016
600 °C	48	0.00438 ± 0.00033	0.00378 ± 0.00029	0.0802 ± 0.0009	1.056 ± 0.007	0.1585 ± 0.0016	0.8158 ± 0.0056	0.3792 ± 0.0023	0.3220 ± 0.0018
800 °C	39	0.00404 ± 0.00030	0.00400 ± 0.00026	0.0820 ± 0.0011	1.068 ± 0.014	0.1588 ± 0.0018	0.8148 ± 0.0086	0.3928 ± 0.0029	0.3436 ± 0.0040
1000 °C	389	0.00475 ± 0.00014	0.00410 ± 0.00005	0.0825 ± 0.0005	1.041 ± 0.005	0.1610 ± 0.0009	0.8194 ± 0.0026	0.3893 ± 0.0017	0.3369 ± 0.0014
1200 °C	1197	0.00467 ± 0.00017	0.00408 ± 0.00007	0.0835 ± 0.0004	1.036 ± 0.004	0.1627 ± 0.0012	0.8209 ± 0.0033	0.3868 ± 0.0016	0.3229 ± 0.0019
1400 °C	719	0.00463 ± 0.00016	0.00410 ± 0.00011	0.0824 ± 0.0003	1.034 ± 0.003	0.1622 ± 0.0009	0.8195 ± 0.0024	0.3806 ± 0.0012	0.3205 ± 0.0011
1600 °C	78	0.00459 ± 0.00019	0.00421 ± 0.00021	0.0830 ± 0.0014	1.041 ± 0.007	0.1616 ± 0.0013	0.8187 ± 0.0062	0.3808 ± 0.0037	0.3221 ± 0.0027
1800 °C	13	0.00443 ± 0.00060	0.00401 ± 0.00029	0.0831 ± 0.0021	1.049 ± 0.019	0.1611 ± 0.0050	0.8163 ± 0.0011	0.3807 ± 0.0055	0.3221 ± 0.0059
total	2723	0.00458 ± 0.00009	0.00404 ± 0.00006	0.0821 ± 0.0002	1.035 ± 0.002	0.1613 ± 0.0006	0.8177 ± 0.0017	0.3850 ± 0.0009	0.3248 ± 0.0009
MIL 090657-HT	¹³² Xe	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
400 °C	1655	0.00368 ± 0.00007	0.00338 ± 0.00007	0.0730 ± 0.0003	0.989 ± 0.003	0.1537 ± 0.0006	0.7936 ± 0.0017	0.3865 ± 0.0016	0.3276 ± 0.0009
600 °C	881	0.00386 ± 0.00007	0.00356 ± 0.00006	0.0748 ± 0.0004	1.006 ± 0.004	0.1548 ± 0.0006	0.8014 ± 0.0022	0.3841 ± 0.0014	0.3259 ± 0.0012
800 °C	971	0.00375 ± 0.00007	0.00347 ± 0.00006	0.0736 ± 0.0003	0.997 ± 0.004	0.1536 ± 0.0006	0.7969 ± 0.0026	0.3864 ± 0.0016	0.3276 ± 0.0012
1000 °C	1659	0.00402 ± 0.00009	0.00368 ± 0.00009	0.0761 ± 0.0004	1.010 ± 0.004	0.1563 ± 0.0006	0.8039 ± 0.0028	0.3894 ± 0.0018	0.3326 ± 0.0010
1200 °C	2652	0.00452 ± 0.00007	0.00401 ± 0.00007	0.0813 ± 0.0003	1.028 ± 0.003	0.1617 ± 0.0007	0.8175 ± 0.0025	0.3824 ± 0.0014	0.3227 ± 0.0009
1400 °C	1240	0.00445 ± 0.00007	0.00397 ± 0.00007	0.0811 ± 0.0004	1.027 ± 0.003	0.1612 ± 0.0007	0.8170 ± 0.0019	0.3822 ± 0.0014	0.3212 ± 0.0012
1600 °C	215	0.00437 ± 0.00011	0.00396 ± 0.00014	0.0807 ± 0.0008	1.032 ± 0.004	0.1594 ± 0.0011	0.8176 ± 0.0043	0.3806 ± 0.0021	0.3216 ± 0.0026
1800 °C	15.3	0.00427 ± 0.00060	0.00373 ± 0.00044	0.0793 ± 0.0014	1.028 ± 0.015	0.1598 ± 0.0027	0.8187 ± 0.0009	0.3831 ± 0.0049	0.3200 ± 0.0067
total	9288	0.00412 ± 0.00003	0.00373 ± 0.00003	0.0774 ± 0.0002	1.012 ± 0.001	0.1577 ± 0.0003	0.8071 ± 0.0010	0.3849 ± 0.0007	0.3259 ± 0.0004

MIL 090657-HTaq	132 Xe	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
400 °C	92	0.00429 ± 0.00016	0.00383 ± 0.00011	0.0794 ± 0.0009	1.036 ± 0.006	0.1601 ± 0.0015	0.8189 ± 0.0040	0.3777 ± 0.0026	0.3179 ± 0.0022
600 °C	78	0.00429 ± 0.00022	0.00392 ± 0.00022	0.0795 ± 0.0010	1.038 ± 0.009	0.1594 ± 0.0014	0.8145 ± 0.0039	0.3750 ± 0.0020	0.3157 ± 0.0031
800 °C	239	0.00462 ± 0.00012	0.00410 ± 0.00020	0.0802 ± 0.0008	1.036 ± 0.005	0.1585 ± 0.0010	0.8173 ± 0.0036	0.3872 ± 0.0029	0.3313 ± 0.0011
1000 °C	593	0.00460 ± 0.00009	0.00406 ± 0.00007	0.0814 ± 0.0006	1.030 ± 0.004	0.1605 ± 0.0009	0.8174 ± 0.0022	0.3905 ± 0.0018	0.3371 ± 0.0013
1200 °C	1102	0.00449 ± 0.00012	0.00396 ± 0.00010	0.0811 ± 0.0004	1.034 ± 0.005	0.1609 ± 0.0010	0.8180 ± 0.0025	0.3783 ± 0.0018	0.3194 ± 0.0019
1400 °C	768	0.00453 ± 0.00011	0.00407 ± 0.00009	0.0822 ± 0.0004	1.034 ± 0.003	0.1621 ± 0.0009	0.8208 ± 0.0025	0.3810 ± 0.0014	0.3193 ± 0.0010
1600 °C	72	0.00453 ± 0.00024	0.00398 ± 0.00027	0.0806 ± 0.0009	1.031 ± 0.006	0.1601 ± 0.0022	0.8152 ± 0.0060	0.3808 ± 0.0043	0.3201 ± 0.0029
1800 °C	16.6	0.00439 ± 0.00070	0.00387 ± 0.00039	0.0811 ± 0.0025	1.044 ± 0.013	0.1623 ± 0.0027	0.8236 ± 0.0006	0.3824 ± 0.0091	0.3218 ± 0.0079
total	2960	0.00452 ± 0.00006	0.00402 ± 0.00005	0.0813 ± 0.0002	1.033 ± 0.002	0.1609 ± 0.0005	0.8184 ± 0.0013	0.3821 ± 0.0009	0.3237 ± 0.0008
Concentrations are g	iven in 10	$^{-12}$ cm ³ STP g ⁻¹ . Unc	certainties of the gas co	incentrations are ass	sumed to be 10%.	Uncertainties of the	isotopic ratios are 1	lа.	

$${}^{20}\mathrm{Ne}_{\mathrm{trap}} = {}^{22}\mathrm{Ne} \times \frac{\left({}^{21}\mathrm{Ne}/{}^{22}\mathrm{Ne}\right) - \left({}^{21}\mathrm{Ne}/{}^{22}\mathrm{Ne}\right)_{\mathrm{C}}}{\left({}^{21}\mathrm{Ne}/{}^{22}\mathrm{Ne}\right)_{\mathrm{trap}} - \left({}^{21}\mathrm{Ne}/{}^{22}\mathrm{Ne}\right)_{\mathrm{C}}} \times \left({}^{20}\mathrm{Ne}/{}^{22}\mathrm{Ne}\right)_{\mathrm{trap}}$$
(1)

$${}^{21}\text{Ne}_{\text{C}} = {}^{22}\text{Ne} \times \frac{\left({}^{21}\text{Ne}/{}^{22}\text{Ne}\right)_{\text{trap}} - \left({}^{21}\text{Ne}/{}^{22}\text{Ne}\right)}{\left({}^{21}\text{Ne}/{}^{22}\text{Ne}\right)_{\text{trap}} - \left({}^{21}\text{Ne}/{}^{22}\text{Ne}\right)_{\text{C}}} \times \left({}^{21}\text{Ne}/{}^{22}\text{Ne}\right)_{\text{C}}$$
(2)

Here, (²⁰Ne/²²Ne)_{trap} and (²¹Ne/²²Ne)_{trap} are coordinates of the intersection of two straight lines in the 20 Ne/ 22 Ne vs. 21 Ne/ 22 Ne diagram (not shown in Fig. 6). The first line passes through sample Ne and cosmogenic Ne. The second line passes through Ne-HL and the average of Ne-Q (for EET 92048-NG, MIL 090657-NG, MIL 090657-HT, and MIL 090657-HTaq), or Ne-SW and Ne-FSW (for NWA 801-NG). The (²¹Ne/²²Ne)_C ratio of 0.811 ± 0.027 for NWA 801-NG is obtained from the regression line of the Ne isotopic ratios in the hightemperature fractions of 1400 °C to 1800 °C, assuming that the $({}^{20}\text{Ne}/{}^{22}\text{Ne})_{\text{C}}$ ratios are 0.8 (Eugster, 1988). The slope and intercept values (1σ) of the regression line are calculated using the ordinary least squares model of IsoplotR (Vermeesch, 2018). Likewise, the regression lines of the 1200 °C to 1600 °C fractions yield the (²¹Ne/²²Ne)_C ratios of 0.904 \pm 0.012, 0.760 \pm 0.008, and 0.752 \pm 0.027 for EET 92048-NG, MIL 090657-NG, and MIL 090657-HT, respectively. The regression line of the 1000 °C to 1600 °C fractions yields the $({}^{21}\text{Ne}/{}^{22}\text{Ne})_{\text{C}}$ ratio of 0.761 \pm 0.040 for MIL 090657-HTaq.

Fig. 7b shows the ²⁰Ne release patterns for EET 92048-NG, MIL 090657-NG, MIL 090657-HT, and MIL 090657-HTaq, along with Renazzo (Reynolds and Turner, 1964). EET 92048-NG, MIL 090657-NG, and MIL 090657-HT have two release peaks in the 400 °C and 1000 °C fractions, while MIL 090657-HTaq and Renazzo have single peaks in the 1000 °C and the 800 °C fractions, respectively. MIL 090657-HTaq released significantly smaller amounts of ²⁰Ne in the 400 °C and 600 °C fractions than MIL 090657-HT (Table 1), indicating that the hydrothermal treatment caused a loss of most Ne released at low temperature.

3.3.2. Helium

The total ³He/⁴He ratios of the EET 92048-NG, MIL 090657-NG, MIL 090657-HT, and MIL 090657-HTaq range from 1.4×10^{-3} to 2.7×10^{-3} (Table 1). These values are much higher than (³He/⁴He)_Q of 1.23×10^{-4} (Ott, 2014) and (³He/⁴He)_{HL} of $<1.70 \times 10^{-4}$ (Huss and Lewis, 1994a), suggesting cosmogenic He contributions to the total He in EET 92048 and MIL 090657. However, these values are much lower than the typical cosmogenic ³He/⁴He ratio of 0.2 (Wieler, 2002b), indicating that amounts of cosmogenic ⁴He are negligible in the total ⁴He of the samples (less than 2% of the total ⁴He). The lowest ³He/⁴He ratios of EET 92048-NG, MIL 090657-NG, and MIL 090657-HT were observed in the 400 °C fractions, while that of MIL

Table 4 Elemental ratios of noble gas components (atom ratios).

Component	⁴ He/ ¹³² Xe	²⁰ Ne/ ¹³² Xe	³⁶ Ar/ ¹³² Xe	⁸⁴ Kr/ ¹³² Xe	References
Q	374 ± 72	3.2 ± 0.5	76 ± 7	0.81 ± 0.05	[1]
HL	$(3.00 \pm 0.30) \times 10^5$	485 ± 26	50 ± 20	0.48 ± 0.04	[2, 3]
Subsolar	_	7.59 ± 1.07	2660 ± 376	5.86 ± 0.84	[4]
Ar-rich gas ^a	_	4.4	429	2.0	[5]
Solar wind	$(6.5 \pm 0.4) \times 10^8$	$(1.0 \pm 0.6) \times 10^{6}$	$(2.37 \pm 0.14) \times 10^4$	9.9 ± 0.3	[6, 7]
Air	216	679	1298	26.8	[8]
Air-saturated water (25 °C)	20	72	428	15.9	[9]

References: [1] Busemann et al. (2000); [2] Ott (2014); [3] Huss and Lewis (1994b); [4] Crabb and Anders (1981), South Oman; [5] Nakamura et al. (2003a); [6] Heber et al. (2009), uncertainties are 1σ ; [7] Vogel et al. (2011), uncertainties are 1σ ; Ozima and Podosek (2002); [8] Sano et al. (2013); [9] Ozima and Podosek (2002).

^a Estimated ratios of the Ar-rich noble gas component in a dark inclusion in the Ningqiang carbonaceous chondrite.

Table 5 Isotopic ratios of He, Ne, Ar, Kr, and Xe of noble gas components.

Component	$^{3}\text{He}/^{4}\text{He}(10^{-4})$	²⁰ Ne/ ²² Ne	²¹ Ne/ ²² Ne	³⁸ Ar/ ³⁶ Ar	References
Q	1.23 ± 0.02	10.11-10.67	0.0294 ± 0.0010	0.187 ± 0.001	[1, 2]
ĤL	$<\!\!1.70 \pm 0.10$	8.500 ± 0.057	0.036 ± 0.001	0.227 ± 0.003	[2, 3]
G		<0.1	< 0.0015		[2, 4]
Solar wind	4.645 ± 0.008	13.777 ± 0.010	0.0329 ± 0.0007	0.183 ± 0.001	[5]
FSW		10.67	0.0294		[6]
Air	0.01399 ± 0.00013	9.8	0.029	0.188	[7]
Cosmogenic	1900 ± 100	0.8	0.8-0.95	1.54	[8, 9]
Component	⁸² Kr/ ⁸⁴ Kr	⁸⁶ Kr/ ⁸⁴ Kr	¹³⁰ Xe/ ¹³² Xe	¹³⁶ Xe/ ¹³² Xe	References
Q	0.2018 ± 0.0002	0.3095 ± 0.0005	0.1619 ± 0.0003	0.3164 ± 0.0008	[1]
P3	0.2023	0.3128 ± 0.0006	0.1589 ± 0.0003	0.3096	[2, 3]
HL	0.1590	0.3623 ± 0.0018	0.1542 ± 0.0003	0.6991	[2, 3]
G	0.4167	0.454-1.176	0.4826 ± 0.0042	0.00343	[2, 4]
Subsolar	0.2045 ± 0.0009	0.3073 ± 0.0017	0.1649 ± 0.0010	0.3095 ± 0.0020	[10]
Solar wind	0.2054 ± 0.0002	0.3012 ± 0.0004	0.1650 ± 0.0004	0.3001 ± 0.0006	[11]
Air	0.20217 ± 0.00004	0.30524 ± 0.00025	0.1514 ± 0.0001	0.3294 ± 0.0004	[7]
Cosmogenic	1.13 ± 0.28	0			[12]

References: [1] Busemann et al. (2000); [2] Ott (2014); [3] Huss and Lewis (1994a), uncertainties are 1σ ; [4] Lewis et al. (1994); [5] Heber et al. (2012); [6] Benkert et al. (1993); [7] Ozima and Podosek (2002); [8] Eugster (1988); [9] Wieler (2002b); [10] Crabb and Anders (1981), South Oman; [11] Meshik et al. (2014); [12] Lavielle and Marti (1988).

090657-HTaq was observed at the 1000 °C fraction (Table 1).

The amount of radiogenic ⁴He produced in CR chondrites in the past 4.56 Gyr is estimated to be 1.8×10^{-5} cm³-STP g^{-1} using the median concentrations of U and Th of CR chondrites (0.0101 ppm and 0.0404 ppm, respectively; Braukmüller et al., 2018) and the equation given in Farley (2002). The ⁴He concentrations in EET 92048-NG $(5.8 \times 10^{-5} \text{ cm}^3 \text{ STP g}^{-1})$, MIL 090657-NG $(8.4 \times 10^{-5} \text{ cm}^3 \text{ str}^{-1})$ cm³ STP g⁻¹), and MIL 090657-HT $(7.8 \times 10^{-5} \text{ cm}^3 \text{ STP g}^{-1})$ indicate that radiogenic ⁴He contributes less than 31% and 23% of the total ⁴He in EET 92048 and MIL 090657, respectively. Therefore, the major fractions (more than two-thirds) of ⁴He in EET 92048 and MIL 090657 are from primordial components. The ⁴He concentration in the NWA 801-NG solar-gas-rich CR chondrite (9.7 \times 10⁻³ $cm^3 STP g^{-1}$) is two orders of magnitude higher than those in EET 92048 and MIL 090657.

Fig. 7a shows the ⁴He release patterns for EET 92048-NG, MIL 090657-NG, MIL 090657-HT, MIL 090657-HTaq, and Renazzo (Reynolds and Turner, 1964). The ⁴He release patterns are similar to the ²⁰Ne release patterns that EET 92048-NG, MIL 090657-NG, and MIL 090657-HT have two release peaks in the 400 °C fractions and the 800–1000 °C fractions, and MIL 090657-HTaq has a single peak in the 800 °C fraction. MIL 090657-HTaq released significantly smaller amounts of ⁴He in the 400 °C and 600 °C fractions than MIL 090657-HT (Table 1), indicating that the hydrothermal treatment caused a loss of most He released in the 400 °C and 600 °C fractions.

3.3.3. Xenon

The contributions of cosmogenic Xe to the total Xe in the three CR chondrites are negligible as the Xe isotopic



Fig. 6. (a) Ne isotopic ratios of EET 92048-NG, MIL 090657-NG, and NWA 801-NG. (b) Ne isotopic ratios of MIL 090657-HT and MIL 090657-HTaq (hydrothermally treated MIL 090657-HT). The total isotopic ratios of each meteorite are shown as triangles of the same colors. The colored solid straight lines are the regression lines for high temperature steps of each sample (see text for details). The curved black line is the range of theoretically calculated Ne isotopic ratios produced by the interaction of solar cosmic rays (SCRs) in ordinary chondrites (Garrison et al., 1995). The isotopic ratios of each component are listed in Table 5. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

ratios of all the samples plot around Q (and the Earth atmosphere for MIL 090657-HT) in 124 Xe/ 126 Xe vs. 130 Xe/ 126 Xe diagrams in supplementary materials (Fig. A3).

Fig. 8a–c show the Xe three-isotope diagrams of 130 Xe/ 132 Xe vs. 136 Xe/ 132 Xe for EET 92048-NG, MIL 090657-NG, and NWA 801-NG. Atmospheric Xe is detected at the lower temperature fractions (400–800 °C) of NWA 801-NG. Since most data points plot around Q, Xe-Q is the dominant component in all three CR chondrites. Xe-HL is also detected in all three samples, and most significant in the 800 °C fractions. The data points of EET

92048-NG above the Q-HL mixing line suggest the presence of Xe-G, consistent with the Kr isotopic ratios (Section 3.3.4).

Fig. 8d–e show the Xe three-isotope diagrams for MIL 090657-HT and MIL 090657-HTaq. A significant contribution of atmospheric Xe is observed in the 400 °C to 1000 °C fractions of MIL 090657-HT, although which is not observed in MIL 090657-NG (Fig. 8b). The Xe isotopic ratios of MIL 090657-HTaq are much closer to the Q-HL mixing line than those of MIL 090657-HT, but there is also a small contribution of atmospheric Xe.



Fig. 7. Extraction temperatures vs. fractions relative to the totals of extracted gases from EET 92048-NG, MIL 090657-NG, MIL 090657-HT, and MIL 090657-HTaq (hydrothermally treated MIL 090657-HT) along with Renazzo (Reynolds and Turner, 1964) for (a) 4 He, (b) 20 Ne, (c) 36 Ar, (d) 84 Kr, and (e) 132 Xe.

The air-corrected 132 Xe concentrations (132 Xe_{corr}) in MIL 090657-HT and MIL 090657-HTaq are calculated based on the assumption that 130 Xe and 132 Xe are two-component mixtures of Xe-Q and Xe-air. The 132 Xe_{corr} fractions of the total 132 Xe in MIL 090657-HT and MIL 090657-HTaq are calculated to be (60 \pm 3)% and (90 \pm 5)% of 132 Xe, which correspond to 132 Xe_{corr} of (5.6 \pm 0.4) \times 10⁻⁹ cm³ STP g⁻¹ and (2.7 \pm 0.2) \times 10⁻⁹ cm³ STP g⁻¹, respectively. In summary, Xe in the three meteorites mainly consists of Xe-Q and Xe-HL with a small fraction of Xe-G for EET 92048.

3.3.4. Krypton

The contributions of cosmogenic Kr and neutron capture process (Marti et al., 1966) to the total Kr in the three CR chondrites are negligible since the isotopic ratios of total Kr are neither significantly affected by cosmogenic Kr nor neutron capture process (Fig. 9 and Fig. A4). However, the high 80 Kr/ 83 Kr ratios relative to Q in the 800 °C fraction of EET 92048-NG, and the 400–800 °C fractions of NWA 801-NG (Fig. A4) are attributable to cosmogenic Kr and/or neutron capture process.

Fig. 9a–c show the Kr three-isotope diagrams of 82 Kr/ 84 Kr vs. 86 Kr/ 84 Kr for EET 92048-NG, MIL 090657-NG, and NWA 801-NG. Since the 86 Kr/ 84 Kr ratio

of Kr-G varies widely (0.454–1.176; Lewis et al., 1994), the two mixing lines of Q and G with the lowest and highest ⁸⁶Kr/⁸⁴Kr ratios are shown in the diagrams. The most data points of NWA 801-NG and MIL 090657-NG are close to Q but show small contributions of Kr-G. On the other hand, the ⁸²Kr/⁸⁴Kr and ⁸⁶Kr/⁸⁴Kr ratios in EET 92048-NG are high, indicating a larger contribution of Kr-G than the other two CR chondrites. The contribution of Kr-G for EET 92048-NG is most significant in the 1000 °C fraction, which is consistent with the highest ¹³⁰Xe/¹³²Xe ratio in the 1000 °C fraction of EET92048-NG.

Fig. 9d–e show the same diagrams for MIL 090657-HT and MIL 090657-HTaq. The data points show small contributions of Kr-G in some extraction steps but mostly plot around Q except for the 400 °C fraction of MIL 090657-HT close to the air. Except for the atmospheric Kr contribution, the two samples are similar in that Kr is almost composed of Kr-Q. In summary, Kr in the three meteorites mainly consists of Kr-Q with small fractions of Kr-G.

3.3.5. Argon

The total 38 Ar/ 36 Ar ratios of the five samples from the three meteorites range from 0.189 to 0.193 (Table 1), which are intermediate between the typical (38 Ar/ 36 Ar)_{trap} ratio of 0.188 (Ozima and Podosek, 2002) and the (38 Ar/ 36 Ar)_C



Fig. 8. Xe three-isotope diagrams of 130 Xe/ 132 Xe vs. 136 Xe/ 132 Xe. (a) EET 92048-NG, (b) MIL 090657-NG, (c) NWA 801-NG, (d) MIL 090657-HT, and (e) MIL 090657-HTaq (hydrothermally treated MIL 090657-HT). The total composition for MIL 090657 is not determined because the amount of Xe released in the 800 °C fraction is missing. The isotopic ratios of each component are listed in Table 5.

ratio of 1.54 (Wieler, 2002b). Assuming two-component mixing of trapped and cosmogenic Ar, the cosmogenic contributions to the total 36 Ar are estimated to be less than 0.4% for all the samples. Thus, cosmogenic 36 Ar is negligible.

The total ⁴⁰Ar/³⁶Ar ratios of EET 92048 (11.029 \pm 0.004), MIL 090657-NG (6.037 \pm 0.002), and NWA 801 (5.776 \pm 0.005) are much higher than primordial components (<0.12; Ott, 2014, and references therein) and solar wind Ar (<0.01; Eugster et al., 2001), suggesting that the radiogenic ⁴⁰Ar (produced by radioactive decay of ⁴⁰K) dominates ⁴⁰Ar of the three meteorites. The total ⁴⁰Ar/³⁶Ar ratios of MIL 090657-HT (7.001 \pm 0.008) and MIL 090657-HTaq (3.457 \pm 0.005) are close to that of MIL 090657-NG and much lower than the atmospheric value of 296 (Ozima and Podosek, 2002). Hence, the contribution of atmospheric ³⁶Ar adsorbed on MIL 090657-HT and MIL 090657-HTaq is negligible. In summary, except for radiogenic ⁴⁰Ar, Ar in all the samples mainly consists of trapped components with small fractions of cosmogenic Ar.

3.3.6. ³⁶Ar/¹³²Xe vs. ⁸⁴Kr/¹³²Xe

Since ³⁶Ar, Kr, and Xe are dominated by trapped components, we assume that ³⁶Ar, ⁸⁴Kr, and ¹³²Xe in the three meteorites are equal to ³⁶Ar_{trap}, ⁸⁴Kr_{trap}, and ¹³²Xe_{trap}. Fig. 10 shows diagrams of trapped ³⁶Ar/¹³²Xe vs. ⁸⁴Kr/¹³²Xe for EET 92048-NG, NWA 801-NG, MIL

090657-HT, and MIL 090657-HTaq. The totals of EET 92048-NG and MIL 090657-HTaq plot near the mixing line of Q and subsolar. The highest 36 Ar/ 132 Xe ratios are observed in the 1000 °C fraction for EET 92048-NG and the 1200 °C fractions for the two MIL 090657 samples. The Xe-enriched compositions in the 400–800 °C fractions for MIL 090657-HT are due to large contributions of atmospheric Xe.

The total $({}^{36}\text{Ar}/{}^{132}\text{Xe})_{\text{trap}}$ ratio of EET 92048-NG is 371 \pm 30. The total ${}^{36}\text{Ar}_{\text{trap}}{}^{132}\text{Xe}_{\text{corr}}$ (${}^{132}\text{Xe}_{\text{corr}}$: aircorrected ${}^{132}\text{Xe}_{\text{trap}}$) ratios of MIL 090657-HT and MIL 090657-HTaq are 353 \pm 34 and 385 \pm 38, respectively. The almost consistent ${}^{36}\text{Ar}_{\text{trap}}{}^{132}\text{Xe}_{\text{corr}}$ ratios between MIL 090657-HT and MIL 090657-HTaq indicate that most of the ${}^{36}\text{Ar}$ in MIL 090657 was not released by the hydrothermal treatment.

The $({}^{36}\text{Ar}/{}^{132}\text{Xe})_{\text{trap}}$ and ${}^{36}\text{Ar}_{\text{trap}}/{}^{132}\text{Xe}_{\text{corr}}$ ratios in these solar-gas-poor samples are much higher than those of Q (76 \pm 7; Busemann et al., 2000) and HL (50 \pm 20; Huss and Lewis, 1994b), indicating the presence of the Ar-rich noble gas component in EET 92048 and MIL 090657. Assuming that all ${}^{132}\text{Xe}$ (or ${}^{132}\text{Xe}_{\text{corr}}$ for the two MIL 090657 samples) is Xe-Q, it is estimated that the ~80% fractions of ${}^{36}\text{Ar}_{\text{trap}}$ in EET 92048 and MIL 090657 are from the Ar-rich component based on the $({}^{36}\text{Ar}/{}^{132}\text{Xe})_{\text{trap}}$ and ${}^{36}\text{Ar}_{\text{trap}}/{}^{132}\text{Xe}_{\text{corr}}$ ratios.



Fig. 9. Kr three-isotope diagrams of 82 Kr/ 84 Kr vs. 86 Kr/ 84 Kr. (a) EET 92048-NG, (b) MIL 090657-NG, (c) NWA 801-NG, (d) MIL 090657-HT, and (e) MIL 090657-HTaq (hydrothermally treated MIL 090657-HT). The total composition for MIL 090657 is not determined because the amount of Kr released in the 800 °C fraction is missing. The isotopic ratios of each component are listed in Table 5.

The total $({}^{36}\text{Ar}/{}^{132}\text{Xe})_{trap}$ ratio of NWA 801-NG (560 ± 44) is higher than those of the other three samples. The total $({}^{84}\text{Kr}/{}^{132}\text{Xe})_{trap}$ ratio of NWA 801-NG is higher than the mixing line between Q and SW.

4. DISCUSSION

4.1. Degree of aqueous alteration

MIL 090657 has experienced minimal thermal and aqueous processing and is reported as one of the most pristine CR chondrites (Davidson et al., 2019b). The presence of the GEMS-like grains in the MIL 090657 matrix (Fig. 3b and c) supports its pristine characteristics since GEMSlike grains in chondrites are highly susceptible to aqueous alteration and have been identified only in some minimally altered carbonaceous chondrites (e.g., Leroux et al., 2015; Matsumoto et al., 2019). Thus, MIL 090657 is one of the best CR chondrites to study the water-susceptible noble gas components potentially lost from aqueously altered CR chondrites.

Framboidal magnetites were observed in some parts of the EET 92048 matrix (Fig. 1b), but not identified in MIL 090657 and NWA 801 by electron microscope observations. The S-XRD analysis displays that phyllosilicates are present in EET 92048 matrix (Fig. 2a), but those are minor or not identified in MIL 090657 and NWA 801. Those characteristics indicates that EET 92048 is not as pristine as the other two meteorites. However, the absence of mafic phenocryst alteration indicates that EET 92048 is less altered than significantly altered CR chondrites such as Renazzo (CR2.4) and Al Rais (CR2.3) reported in Harju et al. (2014). Although the absence of framboidal magnetites and phyllosilicates in the NWA 801 matrix suggests that NWA 801 is less altered than EET 92048, NWA 801 is not suitable for studying the water-susceptible noble gas components in CR chondrite since the meteorite contains a large amount of solar wind noble gases.

4.2. Smooth rim in MIL 090657

Harju et al. (2014) considered that smooth rims in CR chondrites mainly consist of phyllosilicates based on the low analytical weight totals (~82–86 wt%) on EPMA analysis, and suggested that the smooth rims are products of aqueous alteration. On the contrary, based on TEM analysis, Abreu (2016) reported that a smooth rim in the LaPaz Icefield (LAP) 02342 CR chondrite is composed of amorphous silicates, and phylosilicates are absent in the smooth rim.



Fig. 10. Elemental ratio diagrams of trapped 36 Ar/ 132 Xe vs. 84 Kr/ 132 Xe. (a) EET 92048-NG, (b) NWA 801-NG, (c) MIL 090657-HT, and (d) MIL 090657-HT (hydrothermally treated MIL 090657-HT). The elemental ratios of Renazzo (Reynolds and Turner, 1964), Ningqiang (C-ung.), and a dark inclusion in Ningqiang (Nakamura et al., 2003b) are also shown in the figures. The elemental ratios of each component are listed in Table 4. Abbreviation: SH, stepwise heating.

In contrast to the smooth rim in LAP 02342, our TEM analysis revealed that the smooth rim in MIL 090657-PS-F has weakly crystallized structures. The d-values of the smooth rim (~0.45, ~0.26, and ~0.16 nm) are similar to those of synthesized magnesium silicate hydrate (M-S-H), a weakly crystalized phyllosilicate-like phase, around 0.45, 0.33, 0.26, and 0.15 nm (Roosz et al., 2015), suggesting M-S-H-like structure of the smooth rim. The occurrence of M-S-H in the natural environment has been reported as the precipitation on ultramafic rocks with interaction of alkaline water (Nishiki et al., 2020). The formation of M-S-H by hydration of amorphous silicates in a primitive chondrite has also been proposed (Matsumoto et al., 2019). Thus, the M-S-H-like phase that constitutes the smooth rim on the chondrule might have been formed by precipitation with interaction of alkaline water, or hydration of an amorphous rim on the chondrule. In either case, the smooth rim in MIL 090657-PS-F is probably a product of aqueous alteration.

4.3. Noble gas concentrations in MIL 090657-HT and MIL 090657-HTaq

The noble gas concentrations in hydrothermally untreated (-HT) and treated (-HTaq) MIL 090657 samples are compared in Fig. 11. Again, the low-temperature condition during sample preparation probably enhanced adsorption of atmospheric Kr and Xe for MIL 090657-HT (see Section 4.7). All noble gas concentrations in MIL 090657-HTaq are lower than those in MIL 090657-HT. As shown in Fig. 11f–g, the concentrations of ³⁶Ar, ⁸⁴Kr, and ¹³²Xe_{corr} (air-corrected ¹³²Xe) in MIL 090657-HTaq are similarly ~50% of those in MIL 090657-HT. The ⁸⁴Kr concentration in MIL 090657-HTaq is slightly more depleted than ³⁶Ar and ¹³²Xe_{corr}. This is attributable to atmospheric ⁸⁴Kr in MIL 090657-HT mainly released at 400 °C step, which is ~10% of the total ⁸⁴Kr in MIL 090657-HT (Figs. 7 and 9). On the other hand, ⁴He and ²⁰Ne are much more depleted in the treated sample that ⁴He and ²⁰Ne concentra-



Fig. 11. Noble gas concentrations in MIL 090657-HT and MIL 090657-HTaq (hydrothermally treated MIL 090657-HT). (a) ⁴He concentrations, (b) ²⁰Ne concentrations, (c) ³⁶Ar concentrations, (d) ⁸⁴Kr concentrations, and (e) ¹³²Xe concentrations. The ¹³²Xe concentrations are divided into ¹³²Xe-air and ¹³²Xe_{corr} (air-corrected ¹³²Xe). (f) and (g) Concentration ratios of MIL 090657-HTaq to MIL 090657-HT for each element and ¹³²Xe_{corr}.

tions in MIL 090657-HTaq are ${\sim}30\%$ of those in MIL 090657-HT.

The $\sim 50\%$ lower noble gas concentrations for 36 Ar, ⁸⁴Kr, and ¹³²Xe_{corr} of MIL 090657-HTaq can be explained by (1) gas loss caused by the hydrothermal treatment, or (2)lower matrix/chondrule ratio. The latter is because primordial noble gases in chondrites mostly reside in matrix and are poor in chondrules (Vogel et al., 2004). The possibility (1) is less likely because significant amounts of gas loss would result in different depletion factors for each element. For example, a release of the Ar-rich component results in preferential loss of ³⁶Ar. Similarly, loss of the Q component, which has the lower ${}^{36}\text{Ar}/{}^{132}\text{Xe}$ ratio (76 \pm 7; Busemann et al., 2000) than MIL 090657, preferentially reduces the ¹³²Xe_{corr} abundance. This is consistent with the previous study that H₂O treatment of MIL 090657 resulted in almost no release of ³⁶Ar (Krietsch et al., 2019; Krietsch, 2020). Therefore, the lower concentrations for ³⁶Ar, ⁸⁴Kr, and ¹³²Xe_{corr} are likely due to a lower matrix/chondrule ratio for MIL 090657-HTag than MIL 090657-HT. The increase in sample weight caused by oxidation and hydration of MIL 090657-HTaq materials might have also contributed to the lower concentrations. On the contrary, the more significant depletion for ⁴He and ²⁰Ne

is consistent with the changes in release patterns between MIL 090657-HT and MIL 090657-HTaq, suggesting removal of low temperature He and Ne by the hydrothermal treatment (Fig. 7). In summary, the almost consistent relative concentrations for ³⁶Ar, ⁸⁴Kr, and ¹³²Xe_{corr} suggest that most of the primordial Ar, Kr, and Xe in MIL 090657 were not affected by the hydrothermal treatment.

4.4. Water-susceptible primordial noble gas components in less-altered CR chondrites

The Ar-rich noble gas component characterized by high Ar/Xe ratios has been found from UOCs (Schelhaas et al., 1990), some anhydrous carbonaceous chondrites such as CO chondrites (Alaerts et al., 1979), CV chondrites (Matsuda et al., 1980), the Ningqiang carbonaceous chondrite (Nakamura et al., 2003b), and some least-altered CM chondrites (Krietsch et al., in press). The Ar-rich component in Ningqiang is water-susceptible since it disappears after 10-days hydrothermal treatment at 200 °C (Yamamoto et al., 2006). Therefore, the Ar-rich component might have been lost by aqueous alteration processes on the meteorite parent bodies.

The high (³⁶Ar/¹³²Xe)_{trap} ratios in MIL 090657 and EET 92048 indicate that the Ar-rich component exists in the lessaltered CR chondrites (Fig. 10), consistent with the previous reports (Busemann et al., 2019, 2021). The deficit of the Ar-rich component in the Renazzo aqueously altered CR chondrite (Reynolds and Turner, 1964) inferred from its Q-like Ar/Kr/Xe elemental composition (Fig. 10a) supports the water-susceptible nature of the Ar-rich component in CR chondrites (Busemann et al., 2021). However, the Ar-rich component in MIL 090657 survived the 20days hydrothermal treatment at 150 °C. Instead, large fractions of He and Ne in MIL 090657 were lost during the treatment (see Section 3.3). This suggests the presence of another noble gas component enriched in He and Ne that is more susceptible to water than the Ar-rich component in MIL 090657.

Here, we discuss the possible carrier phases and the origins of the two water-susceptible noble gas components found in MIL 090657 based on the results of the noble gas and petrological analyses. We also discuss the distributions of these components in the early Solar System based on the noble gas compositions in various extraterrestrial materials reported in the literature.

4.4.1. Ar-rich component in less-altered CR chondrites

Based on the trapped ${}^{36}\text{Ar}/{}^{132}\text{Xe}$ ratios and ${}^{132}\text{Xe}$ concentrations, ~80% of the all ${}^{36}\text{Ar}_{\text{trap}}$ in EET 92048 and MIL 090657 is estimated to be from the Ar-rich component (see Section 3.3.6). Therefore, the single release peaks of ${}^{36}\text{Ar}$ for the stepwise heating analysis on EET 92048 and MIL 090657 (Fig. 7c) indicate that the Ar-rich component was mainly released at 1000–1200 °C. This is consistent with the release temperature of the Ar-rich component in Ningqiang (Nakamura et al., 2003b).

Nakamura et al. (2003b) found that the Ar-rich component in a dark inclusion in Ningqiang is removed by light HF-HCl etching. They suggested that the amorphous silicate rims (0.1 μ m in thickness on average) around finegrained silicates (Zolensky et al., 2003) carry the Ar-rich component based on TEM observation that revealed removal of the amorphous rims by the etching. These are probably different from the smooth rims in CR chondrites because the smooth rims are not amorphous (Figs. 3 and 4) and more than 10 times thicker (5–30 μ m; Harju et al., 2014) than the amorphous rims in the dark inclusion.

The absence of GEMS-like grains and predominance of poorly crystallized phyllosilicates in the fine-grained matrix of the hydrothermally treated MIL 090657 sample (MIL-HTaq-g1; Fig. 5) indicate that most GEMS-like amorphous silicate grains in MIL 090657 had been altered to phyllosilicates during the treatment. This observation suggests that amorphous silicate rims on fine-grained silicates, if present in MIL 090657, had also been destroyed by the treatment. On the other hand, the consistent ${}^{36}\text{Ar}_{\text{trap}}/{}^{132}\text{Xe}_{\text{corr}}$ (${}^{132}\text{Xe}_{\text{corr}}$: air-corrected ${}^{132}\text{Xe}_{\text{trap}}$) ratios between MIL 090657-HT (353 ± 34) and MIL 090657-HTaq (385 ± 38) indicate that the Ar-rich component mostly survived the treatment. Therefore, the carrier phase of the Ar-rich component in CR chondrites might not be amorphous rims on fine-grained silicates.

The 10-days hydrothermal treatment on the bulk Ningqiang sample at 200 °C resulted in removing the Ar-rich component (Yamamoto et al., 2006), in contrast to the 20-days treatment on MIL 090657 at 150 °C. The different experimental conditions (e.g., temperature and water-rock ratios) may have resulted in different outcomes in the two experiments. Another possibility is that the carrier phase of the Ar-rich component in MIL 090657 may be different from that in Ningqiang and less susceptible to water.

Krietsch et al. (2019) and Krietsch (2020) reported that the treatment with acetic acid released most of 36 Ar and 38 Ar in MIL 090657, suggesting that the Ar-rich component carrier is acetic-acid soluble materials. In summary, the Arrich component carrier phase in the CR chondrites is acetic acid-soluble materials that survive hydrothermal treatment at 150 °C but is destroyed by the severe aqueous alteration on the parent body, indicating it is probably not amorphous silicates.

In the ${}^{36}\text{Ar}/{}^{132}\text{Xe}$ vs. ${}^{84}\text{Kr}/{}^{132}\text{Xe}$ diagram (Fig. 10a), EET 92048 plots near the mixing line between Q and subsolar. The subsolar component has been observed in some enstatite chondrites with high petrologic types (e.g., Okazaki et al., 2001, 2010; Patzer and Schultz, 2002), and ureilites may also contain a similar component (Broadley et al., 2020). Okazaki et al. (2001) reported that the subsolar component in enstatite chondrites is concentrated in porphyritic pyroxene chondrules and suggested that this component is the solar noble gas implanted in chondrule precursors. The higher release temperature of the subsolar component (1300-1600 °C; Okazaki et al., 2010) than that of the Ar-rich component in the CR chondrites (1000-1200 °C) indicates that the host phase of the Ar-rich component in the CR chondrites is different from the subsolar component in enstatite chondrites. Since MIL 090657-HT and MIL 090657-HTaq are contaminated by the adsorbed atmospheric Kr and Xe (Section 4.7), the ³⁶Ar/¹³²Xe vs. ⁸⁴Kr/¹³²Xe compositions of MIL 090657-HT and MIL 090657-HTaq are not discussed here.

The elemental ratios of the Ar-rich component in the dark inclusion in Ningqiang have been estimated by the difference between the noble gas compositions of the etched and unetched samples (Nakamura et al., 2003b). In the ³⁶Ar/¹³²Xe vs. ⁸⁴Kr/¹³²Xe diagram (Fig. 10a), the Ningqiang host (Nakamura et al., 2003b) plots near the mixing line of Q and the Ar-rich component in the dark inclusion (DI). Because EET 92048-NG plots above this mixing line, the Ar-rich component in EET 92048 has a higher ³⁶Ar/⁸⁴-Kr ratio than that in Ningqiang. Nakamura et al. (2003b) proposed that the origin of the Ar-rich component in Ninggiang is the implantation of noble gas ions with a composition fractionated relative to the solar abundance formed by the incomplete ionization by high-temperature electrons under plasma conditions at the spouting point of the ancient solar wind or the T Tauri disk wind. The incompletely ionized noble gases are enriched in the heavier elements due to the lower ionization potentials (Jokipii, 1964). In this process, electrons with higher temperatures produce less-fractionated noble gas compositions. If the Ar-rich component in EET 92048 had been produced by the similar incomplete ionization process, the higher ³⁶Ar/⁸⁴Kr ratio of the Ar-rich component than that in Ningqiang suggests that the temperature of electrons that induced the ionization of the Ar-rich component in EET 92048 was higher than that induced the ionization of the Ar-rich component in Ningqiang.

Because the petrologic properties suggest that NWA 801 is aqueously less altered, this solar-gas-rich CR chondrite may also contain the Ar-rich component. The higher total (³⁶Ar/¹³²Xe)_{trap} ratio of NWA 801-NG than the other two meteorites is probably due to abundant implanted solar wind Ar. However, the higher (84Kr/132Xe)trap ratios of NWA 801-NG than the SW-Q mixing line in the ${}^{36}\text{Ar}/{}^{132}\text{Xe}$ vs. ${}^{84}\text{Kr}/{}^{132}\text{Xe}$ diagram (Fig. 10b) suggests the presence of the Ar-rich component in NWA 801 because the Ar-rich component is characterized by a higher Kr/Xe ratio than solar wind (e.g., Nakamura et al., 2003b). NWA 801 experienced terrestrial weathering (Sanghani et al., 2021) that often enhances atmospheric contamination into chondrites (Scherer et al., 1994). However, it is less likely that the high total (Kr/Xe)_{trap} ratio is due to atmospheric contamination because the total Kr and Xe isotopic compositions of NWA 801-NG are not close to the air (Fig. 8c, 9c, A3a, and A4c). Thus, NWA 801 probably contains the Ar-rich component. The terrestrial weathering effect on the Ar-rich component is not vet known, but it may reduce the Ar-rich component abundance since the Ar-rich component is water-susceptible. This should be investigated in future studies.

4.4.2. Ar-rich component in other primitive chondrites

Fig. 12 shows $(^{20}\text{Ne}/^{132}\text{Xe})_{\text{trap}}$ vs. $(^{36}\text{Ar}/^{132}\text{Xe})_{\text{trap}}$ diagrams for EET 92048-NG, MIL 090657-HT, and MIL

090657-HTaq (MIL 090657-HT after hydrothermal treatment) along with various primitive carbonaceous chondrites and UOCs reported in the literature (Mazor et al., 1970; Schelhaas et al., 1990; Bischoff et al., 1993; Scherer and Schultz, 2000; Nakamura et al., 2003a; Okazaki and Nagao, 2017; Riebe et al., 2017; Davidson et al., 2019a; Krietsch et al., in press). The meteorites shown in the diagrams are summarized in the supplementary materials (Table A1). The meteorites with 20 Ne/ 22 Ne ratios smaller than the arbitrarily determined value of 1.2 are excluded because of the difficulty in correcting cosmogenic Ne. The CM chondrites in Krietsch et al. (in press) showing atmospheric contributions on Xe are excluded. The concentrations of air-corrected ¹³²Xe (¹³²Xe_{corr}) are applied to MIL 090657-HT and MIL 090657-HTag. For the deconvolution of Ne in the meteorites from the literature, the ratios of $({}^{20}\text{Ne}/{}^{22}\text{Ne})_{\text{C}}$, $({}^{21}\text{Ne}/{}^{22}\text{Ne})_{\text{C}}$, and $({}^{21}\text{Ne}/{}^{22}\text{Ne})_{\text{trap}}$ are assumed to be 0.8, 0.9 (average chondritic values; Eugster, 1988), and 0.03 (a typical value for trapped Ne), respectively. the For deconvolution of Ar, $({}^{38}Ar/{}^{36}Ar)_{C}$ and $({}^{38}Ar/{}^{36}Ar)_{trap}$ ratios are assumed to be 1.54 (Wieler, 2002b) and 0.188 (Ozima and Podosek, 2002), respectively. The 132 Xe in the meteorites are assumed to be entirely trapped. The errors of the $(^{20}\text{Ne}/^{132}\text{Xe})_{\text{trap}}$ and $({}^{36}\text{Ar}/{}^{132}\text{Xe})_{\text{trap}}$ ratios in the meteorites are less than 21% (Table A1).

If Q and HL dominate trapped noble gases in a meteorite, the meteorite should be on the Q-HL mixing line in the $({}^{20}\text{Ne}/{}^{132}\text{Xe})_{\text{trap}}$ vs. $({}^{36}\text{Ar}/{}^{132}\text{Xe})_{\text{trap}}$ diagram (Fig. 12). And, if a meteorite contains solar wind noble gases, it should be near the Q-SW mixing line that is very close to the Q-HL mixing line when the $({}^{20}\text{Ne}/{}^{132}\text{Xe})_{\text{trap}}$ ratio is less



SW

Fig. 12. Elemental ratio diagrams of trapped ²⁰Ne/¹³²Xe vs. ³⁰Ar/¹³²Xe. EET 92048-NG, MIL 090657-HT, and MIL 090657-HTaq (hydrothermally treated MIL 090657-HT), and NWA 801-NG are shown along with various primitive chondrites in the literatures (Mazor et al., 1970; Schelhaas et al., 1990; Bischoff et al., 1993; Scherer and Schultz, 2000; Nakamura et al., 2003a; Okazaki and Nagao, 2017; Riebe et al., 2017; Davidson et al., 2019b; Krietsch et al., in press). The ¹³²Xe_{trap} concentrations in MIL 090657-HT and MIL 090657-HTaq are corrected for atmospheric contributions. The noble gas deconvolution procedures are described in Section 4.4.2. The elemental ratios of the meteorites shown in the diagrams are summarized in the supplementary materials (Table A1). The dashed lines represent mixing lines between Q and each of the other components. The elemental ratios of each component are listed in Table 4.

than ~100 (see the Q-SW mixing line in Fig. 12). Solar-gaspoor EET 92048 and MIL 090657 plot away from the Q-HL mixing line because of the excess ³⁶Ar from the Arrich component. As shown in the diagram, some primitive chondrites also have excess (³⁶Ar/¹³²Xe)_{trap} ratios, suggesting they contain the Ar-rich component. We assume that these meteorites do not contain the subsolar component because the subsolar (or similar) component has only been found from enstatite chondrites and ureilites (e.g., Okazaki et al., 2001, 2010; Patzer and Schultz, 2002; Broadley et al., 2020). The $({}^{84}Kr/{}^{132}Xe)_{trap}$ ratios of the meteorites (0.7–1.7) are much lower than those of air (26.8; Sano et al., 2013) and the air-saturated water (15.9; Ozima and Podosek, 2002) but close to Q (0.81 \pm 0.5; Busemann et al., 2000) (Fig. A5), indicating that air contamination is insignificant for the $\binom{^{20}\text{Ne}/^{132}\text{Xe}}{_{\text{trap}}}$ and $\binom{^{36}\text{Ar}/^{132}\text{Xe}}{_{\text{trap}}}$ ratios. Here, we discuss the presence of the Ar-rich component in the various primitive chondrites.

The two CR chondrites Al Rais and Renazzo, both experienced aqueous alteration (Harju et al., 2014), plot near the Q-HL mixing line. This indicates the absence of the Ar-rich component in the two CR chondrites. As already discussed in the previous section, the aqueous alteration processes on the CR chondrite parent body probably removed the Ar-rich component (Busemann et al., 2021). The other three CR chondrites GRA 95229, Acfer 097, and El Djouf 001 are solar-gas-rich meteorites (Bischoff et al., 1993; Scherer and Schultz, 2000) but show higher ($^{36}Ar/^{132}Xe$)_{trap} ratios than the Q-SW mixing line, suggesting the presence of the Ar-rich component. The presence of the Ar-rich component with their petrologic subtypes of 2.7–2.8, indicating aqueously less altered characteristics (Harju et al., 2014).

Most of the CI and CM chondrites and the Tagish Lake (C2-ungrouped) carbonaceous chondrite, which have experienced aqueous alteration (e.g., Weisberg et al., 2006), plot near the Q-HL mixing line or the Q-SW mixing line, suggesting they do not contain the Ar-rich component. Some CM chondrites such as NWA 10827, LEW 85312, and Paris have high $({}^{36}\text{Ar}/{}^{132}\text{Xe})_{trap}$ ratios (179–241). Krietsch et al. (in press) found that these meteorites are some of the least aqueously altered CM chondrites, and they concluded that CM chondrites contained the Ar-rich component originally but aqueous alteration causes loss of the Ar-rich component.

The CV chondrites are divided into oxidized (CV_{oxA} and CV_{oxB}) and reduced (CV_{red}) subgroups (Weisberg et al., 1997). The CV_{red} chondrites are less aqueously altered than the CV_{ox} chondrites (Weisberg et al., 2006). The elevated ($^{36}Ar/^{132}Xe$)_{trap} ratios (~300) in the CV_{red} chondrites Efremovka and Leoville, and the higher ($^{36}Ar/^{132}Xe$)_{trap} ratios than the Q-SW mixing line for the Arch CV_{red} chondrite indicate the presence of the Ar-rich component. On the other hand, most CV_{ox} chondrites plot near the Q-HL or Q-SW mixing lines. This implies that CV chondrites had primarily obtained the Ar-rich component in the early Solar System, but subsequent aqueous alteration removed it from some CV chondrites.

The CO chondrites are minimally affected by aqueous alteration (Brearley, 2006) and escaped from high-

temperature metamorphism higher than 500–600 °C (Sears et al., 1991a). The Acfer 094 carbonaceous chondrite (C2-ungrouped) is extremely primitive and has experienced minimal secondary alteration processes (e.g., Greshake, 1997; Matsumoto et al., 2019). The CO chondrites, Acfer 094, Ningqiang, and the UOCs show high $({}^{36}\text{Ar}/{}^{132}\text{Xe})_{trap}$ ratios (181–281 for CO chondrites, 624 for Acfer 094, 263 for Ningqiang, and 163–247 for UOCs), suggesting the presence of the Ar-rich component in these primitive chondrites.

Although some evidence for aqueous alteration has been identified in the Semarkona LL3.00 chondrite (Brearley, 2006), the presence of the Ar-rich component indicates that the alteration process was not sufficient to destroy all Ar-rich component carriers. This is consistent with a recent TEM study on Semarkona that demonstrates the aqueous alteration has affected Semarkona heterogeneously, and regions of much more pristine matrix that have escaped extensive alteration are still preserved within the meteorite (Dobrică and Brearley, 2020).

In summary, the Ar-rich component commonly exists in UOCs and anhydrous primitive carbonaceous chondrites, including the less-altered CR chondrites analyzed in this study. This indicates that the acquisition of the Ar-rich component is a common process in the ordinary and carbonaceous chondrite forming regions of the early Solar System. The absence of the Ar-rich component in most of the aqueously altered chondrites suggests that the Ar-rich component in most chondrites is hosted by similar watersusceptible phases. However, we cannot exclude the possibility that some aqueously altered chondrites have not obtained the Ar-rich component from the beginning.

Jokipii (1964) proposed that ions would concentrate in the solar magnetic field near the Sun, and such ions would fractionate relative to the primordial solar nebula composition depend on the ionization potentials of each element. The possible origin of the Ar-rich component is that the primordial solar nebula noble gas fractionated to the heavier elements due to the lower ionization potentials were implanted into water-susceptible materials near the protosun (Nakamura et al., 2003b). Then, the Ar-rich component carriers were subsequently transported over a wide range of orbital distances, possibly by outward flows around the midplane of the protoplanetary disk (Ciesla, 2007).

4.4.3. He and Ne rich water-susceptible materials in lessaltered CR chondrites

The significant changes in the ⁴He and ²⁰Ne release patterns between MIL 090657-HT and MIL 090657-HTaq (Fig. 7a and b) indicate that the hydrothermal treatment removed large fractions of He and Ne in MIL 090657. This is consistent with the previous report in Krietsch et al. (2019) and Krietsch (2020) that the H₂O treatment of MIL 090657 released large amounts of He and Ne, which correspond to the ~25% of total ⁴He and ~35% of total ²⁰Ne in MIL 090657.

The relative abundances of ³⁶Ar, ⁸⁴Kr, and ¹³²Xe_{corr} were not changed after the hydrothermal treatment (Fig. 11), suggesting that most ¹³²Xe_{corr} in MIL 090657-HTaq survived the treatment. Therefore, assuming that the ⁴He/¹³²Xe_{corr} ratio is entirely homogeneous in MIL 090657, the differences in the $^{4}\text{He}/^{132}\text{Xe}_{corr}$ ratios of MIL 090657-HT (13900 \pm 1200) and MIL 090657-HTaq (10000 \pm 980) indicate that the treatment removed 28 \pm 11% of total ^{4}He in MIL 090657. Likewise, the $^{20}\text{Ne}/^{132}\text{Xe}_{corr}$ ratios of MIL 090657-HT (73.7 \pm 6.2) and MIL 090657-HTaq (44.6 \pm 4.4) indicate that 40 \pm 10% of ^{20}Ne was lost from MIL 090657. These estimates are in agreement with the results in Krietsch et al. (2019) and Krietsch (2020). The concentrations of the lost fractions of ^{4}He and ^{20}Ne in MIL 090657-HT are (2.2 \pm 0.9) \times 10⁻⁵ cm³ STP g⁻¹ and (1.6 \pm 0.4) \times 10⁻⁷ cm³ STP g⁻¹, respectively.

The ⁴He and ²⁰Ne release peaks at the 400–600 °C fractions of MIL 090657-HT are not present in MIL 090657-HTaq (Fig. 7a and b), indicating that the watersusceptible fractions of He and Ne in MIL 090657 are released at relatively low temperature. Since the first extraction temperature is 400 °C, the release of the He and Ne hosted by water-susceptible materials probably started at a temperature lower than 400 °C.

The amount of ²⁰Ne released in the 400 °C fraction of MIL 090657-HTaq is two orders of magnitude less than that of MIL 090657-HT. This suggests that ²⁰Ne released in the 400 °C fraction of MIL 090657-NG is dominated by Ne in the water-susceptible carrier. Assuming that all the Ne in MIL 090657-NG is a mixture of the cosmogenic Ne with isotopic ratios estimated in Section 3.3.1 and trapped Ne with a (²¹Ne/²²Ne)_{trap} ratio of 0.0294 \pm 0.001 0 (typical value for Q; Ott, 2014), the (²⁰Ne/²²Ne)_{trap} ratio of Ne released in the 400 °C fraction of MIL 090657-NG is calculated to be 10.21 \pm 0.06 (Fig. 13).

The similar release patterns and the Ne isotopic ratios for EET 92048-NG (Fig. 6a and 7) suggest that the same water-susceptible noble gas component is also present in EET 92048. The $({}^{20}\text{Ne}/{}^{22}\text{Ne})_{trap}$ ratio of Ne released in the 400 °C fraction of EET 92048-NG is calculated to be 10.09 ± 0.05 based on the same assumptions for MIL 090657-NG. The estimated (²⁰Ne/²²Ne)_{trap} ratios are in the range of Q-Ne (10.11-10.67; Busemann et al., 2000) within uncertainties, indicating that the Ne isotopic composition of the water-susceptible component in the less-altered CR chondrites is Q-like. This is also consistent with the previous report by Krietsch et al. (2019) and Krietsch (2020) that the isotopic composition of the Ne released by the H₂O treatment is in the range of Q. Thus, this watersusceptible noble gas component is neither implanted solar wind nor terrestrial noble gas but a primordial noble gas component.

If this water-susceptible component has the same ${}^{20}\text{Ne}/{}^{132}\text{Xe}$ ratio as Q (3.2 ± 5; Busemann et al., 2000), the ${}^{132}\text{Xe}$ concentration of this water-susceptible component in MIL 090657-HT should be ~5 × 10⁻⁸ cm³ STP g⁻¹. However, this is one order of magnitude higher than the concentration of ${}^{132}\text{Xe}_{corr}$ in MIL 090657-HT ((5.6 ± 0.4) × 10⁻⁹ cm³ STP g⁻¹), although ${}^{132}\text{Xe}_{corr}$ is the total of all primordial components including Xe-Q, Xe-HL, and Xe of the Ar-rich component. Therefore, the noble gas component removed by the hydrothermal treatment has a much higher Ne/Xe ratio than Q.

Fig. 13. Ne three-isotope diagram showing the Ne isotopic ratios released in the 400 °C fractions of EET 92048-NG and MIL 090657-NG. The Ne isotopic ratios in a refractory amorphous interplanetary dust particle "Manchanito" (Ogliore et al., 2020) and cometary dusts (St-1 and Thera-2) captured by the Stardust spacecraft (Marty et al., 2008) are also shown. The ²⁰Ne/²²Ne ratios of the trapped components released in the 400 °C fractions are calculated to be 10.08 \pm 0.06 and 10.21 \pm 0.06 for EET 92048-NG and MIL 090657-NG, respectively. Assumptions for the calculation are: (1) Ne in the samples are mixtures of cosmogenic and trapped components, and (2) the ²¹Ne/²²Ne ratios of the trapped components are 0.0294 \pm 0.0010 (typical value for the Q component; Busemann et al., 2000).

This Q-like Ne-rich noble gas component released at low temperature (<400-600 °C) is probably not present in the Renazzo aqueously altered CR chondrite because this meteorite releases very small fractions of ²⁰Ne at 200-600 °C (Fig. 7b) (Reynolds and Turner, 1964). This suggests that the Q-like Ne-rich component had been lost by the aqueous alteration process in the CR chondrite parent body. On the other hand, the Q-like Ne-rich component is present in EET 92048 that has been more aqueously altered than MIL 090657. This indicates that the aqueous alteration for EET 92048 was not sufficient to cause a complete loss of the Q-like Ne-rich component.

The terrestrial contamination to Ne released at low temperature from EET 92048 and MIL 090657 is unlikely because of the following two reasons. First, adsorption of terrestrial Ne is not observed even in heavily weathered chondrites from hot deserts (Scherer et al., 1994). Thus, it is less likely that the amount of adsorbed terrestrial ²⁰Ne is comparable to the ²⁰Ne released at low temperature (400–600 °C), which are more than one-third of the total ²⁰Ne in the two meteorites. Second, significant atmospheric contaminations are not observed in any noble gas isotopic ratios of EET 92048-NG and MIL 090657-NG. For example, all Xe isotopic ratios plot near the Q-HL mixing line (Fig. 8a and b). Moreover, the ⁴⁰Ar/³⁶Ar ratios are much





Fig. 14. Extraction temperatures vs. 20 Ne/ 22 Ne (black) and 40 Ar/ 36 Ar (red) ratios. (a) EET 92048-NG, (b) MIL 090657-NG, (c) MIL 090657-HT, and (d) MIL 090657-HTaq (hydrothermally treated MIL 090657-HT). The isotopic ratios of each component are listed in Table 5. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

lower than the atmospheric Ar composition (Fig. 14). Considering that Ar is more efficiently adsorbed onto chondritic materials (Fanale and Cannon, 1972), the terrestrial Ne contamination for EET 92048 and MIL 090657 is unlikely. Contamination of terrestrial He is much less likely because the amounts of ⁴He in both meteorites are more than 100 times larger than ²⁰Ne, whereas the ⁴He/²⁰Ne ratio of air is very low (0.319; Sano et al., 2013).

MIL 090657-HT and MIL 090657-HTaq contain nonnegligible amounts of atmospheric Xe (~40% and ~10% of 132 Xe, respectively) that are probably adsorbed onto the samples during the crushing procedure in liquid nitrogen at ~77 K (see Section 4.7). Given that Xe is more than 10^5 times efficiently adsorbed on a carbonaceous chondrite than Ne at 113 K (Allende, CV chondrite; Fanale and Cannon, 1972), atmospheric ²⁰Ne adsorbed onto the samples are negligibly low (less than 0.01% of the total ²⁰Ne). This is consistent with the ⁴⁰Ar/³⁶Ar ratios much lower than the atmospheric value (Fig. 14c and d). Adsorption of atmospheric He is also negligible since He is less adsorbable than Ne. In summary, the aqueously less-altered CR chondrites contain the water-susceptible noble gas component enriched in He and Q-like Ne, and this component is released at a relatively low temperature of <400–600 °C.

4.4.4. Carrier of Q-like Ne-rich component in less-altered CR chondrites

The different reactions of the Ar-rich component (almost unaffected) and the Q-like Ne-rich component (lost) in MIL 090657 to the hydrothermal treatment indicate that their host phases are different. The Q-like Ne in the lessaltered CR chondrites are characterized by: (1) its Q-like isotopic ratios; (2) removal by hydrothermal treatment at 150 °C; (3) low release temperature (<400–600 °C); and (4) coexistence with a large amount of He. Here, we discuss the possible host phase of the Q-like Ne-rich component.

The phase Q hosting the Q component is an acidinsoluble carbonaceous phase (Ott, 2014) and releases the Q gas at relatively high temperatures (mainly at >1000 °C) (Huss et al., 1996). Since these characteristics are different from the Q-like Ne in the CR chondrites, phase Q is not the host material of this component.

Recently, highly concentrated Ne $(0.0065 \pm 0.0016 \text{ cm}^3 \text{STP g}^{-1})$ with Q-like isotopic ratio is found in an amorphous refractory interplanetary dust particle (IDP) "Manchanito" (Ogliore et al., 2020). Most fraction of ²⁰Ne in Manchanito was released at 400–600 °C. The Q-like isotopic ratio and the low release temperature are consistent with the Q-like Ne in MIL 090657. Therefore, the Manchanito-like amorphous materials are one of the most promising candidates for the carrier of the Q-like Ne-rich component in the less-altered CR chondrites. This is supported by the hydrothermal treatment experiment on MIL 090657 that resulted in removing the Q-like Ne and destruction of amorphous silicates in the matrix, as discussed in Section 4.4.1.

Since the chemical composition of Manchanito (Ca- and Al-rich amorphous silicate) is not similar to the typical Feand Mg-rich amorphous silicate in CR chondrite matrices (Ogliore et al., 2020), the GEMS-like amorphous silicate grains might not be the host phase of the Q-like Ne-rich component. If the Manchanito-like amorphous materials are the carrier of the Q-like Ne-rich component, the abundance of the materials in MIL 090657 is estimated to be 25 ± 8 ppm based on the ²⁰Ne concentration in Manchanito (Ogliore et al., 2020) and the concentration of the ²⁰Ne of the Q-like Ne in MIL 090657-HT.

Most Fe-Ni metals were also altered by the hydrothermal treatment, as demonstrated by the S-XRD analysis and SEM observation (Figs. 2b, d, and 5a). However, the high retentivity of meteoritic metals for He and Ne (Nyquist et al., 1972) is not consistent with the low release temperature of the Q-like Ne in MIL 090657. Therefore, Fe-Ni metals are less promising as the host phase of the Q-like Ne-rich component in CR chondrites.

4.4.5. Does *Q*-like Ne-rich component exist in other pristine chondrites?

The low release temperature (<400–600 °C) of the Q-like Ne found in the less-altered CR chondrites suggests that this noble gas component can be lost not only by aqueous alteration but also by high-temperature metamorphism. Most anhydrous chondrites have experienced hightemperature metamorphism after their accretion (e.g., 500–600 °C for type 3.5 chondrites; Sears et al., 1991b) that might have resulted in a loss of the Q-like Ne-rich component. Several indicators imply that Dominion Range (DOM) 08006 (CO3.00) is the most pristine CO chondrite in our collections (Davidson et al., 2019a), suggesting that DOM 08006 had avoided both severe aqueous alteration and high-temperature metamorphism. Here, we discuss if there is a Q-like Ne-rich component in DOM 08006 based on the noble gas data reported in Davidson et al. (2019a).

As there is no evidence for solar wind implantation in DOM 08006 (Davidson et al., 2019a), the $({}^{20}Ne/{}^{132}Xe)_{trap}$ ratio is almost identical to the total of the primordial noble gases in DOM 08006. The Xe isotopic composition is similar to Q with a small addition of HL, suggesting that Xe in DOM 08006 is dominated by Q and HL (Davidson et al., 2019a). Assuming two-component mixture of Q and HL

(Table 2) for Xe, the ¹³⁶Xe/¹³²Xe ratio of 0.3299 \pm 0.0039 in DOM 08006 (Davidson et al., 2019a) yields the ¹³²Xe-Q/(¹³²Xe-Q + ¹³²Xe-HL) ratio of 0.977 \pm 0.003 in this meteorite. Thus, the (²⁰Ne/¹³²Xe)_{trap} ratio of this Q + HL mixture should be 20 \pm 5 based on the ²⁰Ne/¹³²Xe ratios of Q (3.2 \pm 0.5; Busemann et al., 2000) and HL (485 \pm 26; Huss and Lewis, 1994b). If DOM 08006 contains the Q-like Ne-rich component, the (²⁰Ne/¹³²Xe)_{trap} ratio should be higher than 20 \pm 5 since this component has a much higher ²⁰Ne/¹³²Xe ratio than that of Q (Section 4.4.3). However, the (²⁰Ne/¹³²Xe)_{trap} ratio in DOM 08006 is ~14 (Fig. 12), indicating that the (²⁰Ne/¹³²Xe)_{trap} ratio does not show positive evidence for the presence of the Q-like Ne-rich component in DOM 08006.

The deficit of the Q-like Ne-rich component in DOM 08006 suggests that the carrier materials of this component were more efficiently accreted on the CR chondrite parent body than the CO chondrite parent body. Otherwise, the Q-like Ne-rich component had been lost by mild heating in DOM 08006, which is probably ~200 °C for the least-metamorphosed CO3 chondrites (Huss et al., 2006, and references therein).

Some of the most primitive CM chondrites may contain an unknown Ne-rich component since these meteorites show elevated $^{20}Ne_{trap}$ concentrations without evidence for solar wind noble gases (Krietsch et al., in press). The relationship between this Ne-rich component and the Qlike Ne-rich component is unclear, and further study is needed.

4.4.6. Possible link to cometary materials: implication to the forming region of the CR chondrite parent body

Manchanito containing highly concentrated O-like Ne was found in a fragile cluster IDP (Ogliore et al., 2020). Highly concentrated Ne with a Q-like isotopic composition is also found in the Stardust aerogel samples extracted from the Wild 2 comet coma collection tray (Marty et al., 2008; Palma et al., 2019) (Fig. 13). Because of some reasons such as (1) Manchanito's noble gas signatures are similar to those of the Stardust samples, (2) cluster IDPs are possibly derived from comets (e.g., Joswiak et al., 2017), and (3) the relatively late formation of Manchanito (Ogliore et al., 2020) is consistent with other cometary objects, Ogliore et al. (2020) concluded that the most likely parent body of Manchanito is a comet. As the carrier of the Q-like Ne-rich component in MIL 090657 is probably Manchanito-like amorphous materials (Section 4.4.4), the less-altered CR chondrites and comets may share similar Manchanito-like materials.

If so, the deficit of the Q-like Ne-rich component in DOM 08006 (CO3.00) suggests that the Manchanito-like materials existed in the outer region of the early Solar System were more efficiently provided to the CR chondrite forming region than the CO chondrite forming region. This is consistent with the recent implications that the CR chondrites were accreted at a greater heliocentric distance than the CO chondrites based on isotopic studies (Van Kooten et al., 2016, 2020; Budde et al., 2018) and a model prediction (Desch et al., 2018). Another supporting evidence for the link between CR chondrites and comets is that CR

chondrule-like silicates have been identified in cometary materials, suggesting that the CR chondrite forming region and the comet forming region were not physically isolated (Ogliore et al., 2012; Nakashima et al., 2012; Frank et al., 2014; Defouilloy et al., 2017).

4.4.7. Distribution of water-susceptible noble gas components in the early Solar System

It has been suggested that the ordinary chondrites formed closer to the Sun than the carbonaceous chondrites (e.g., Desch et al., 2018). In this view, the radial distributions of the two water-susceptible noble gas components (the Ar-rich component and the Q-like Ne-rich component) in the early Solar System are illustrated in Fig. 15 based on the above discussions. The Ar-rich component was commonly distributed in the ordinary chondrite and carbonaceous chondrite forming regions. However, its presence beyond the carbonaceous chondrite forming region is not yet known. On the other hand, the Q-like Ne-rich component was present in the outer region of the solar nebula where comets were formed. The distribution of this component extends to the CR chondrite forming region.

4.5. Cosmogenic Ne

A cosmic-ray exposure (CRE) age for EET 92048-NG is calculated from the ²¹Ne_C concentration and ²¹Ne_C production rates (P₂₁). The P₂₁ value is determined using the model for carbonaceous chondrites given by Leya and Masarik (2009) and the average elemental abundances in CR chondrites given in Alexander (2019). The pre-atmospheric radius of the meteorite is assumed to be <100 cm since the pre-atmospheric radius of stony meteorites rarely exceeds a meter and mostly less than ~40 cm (Wieler, 2002b). The possible P₂₁ value for EET 92048-NG with the (²¹Ne/²²Ne)_C ratio of 0.904 ± 0.012 is in the range of (0.259–0.321) × 10⁻⁸ cm³ STP g⁻¹ Ma⁻¹, depending on the pre-atmospheric radius and depth below the surface of the meteorite. Then, the CRE age for EET 92048 should be in the range of 6.3–7.8 Ma.

The GCR-induced ²¹Ne/²²Ne ratios are mostly related to shielding depths, where samples with lower shielding have smaller ²¹Ne/²²Ne ratios (e.g., Wieler, 2002b). The lowest GCR-induced ²¹Ne/²²Ne ratios for CR chondrites are estimated to be 0.824 (in 0–1 cm depth of 10 cm radius meteoroid) using the model for carbonaceous chondrites given by Leya and Masarik (2009) and the average elemental abundances in CR chondrites (Alexander, 2019).

The $({}^{21}\text{Ne}/{}^{22}\text{Ne})_{\text{C}}$ ratio of 0.760 \pm 0.008 in MIL 090657-NG is below the estimated lowest GCR-induced ²¹Ne/²²Ne ratio in CR chondrites. The low (²¹Ne/²²Ne)_C ratio suggests the presence of SCR-induced Ne that has a low ²¹Ne/²²Ne ratio of <0.65 (Garrison et al., 1995). The lower energy of the SCRs (<100 MeV) compared to the GCRs (~100 MeV to several GeV for most nucleons) (Vainio et al., 2009; Schwadron et al., 2017) leads to SCR nuclide production exclusively within a few centimeters of the exposed silicate surface (Garrison et al., 1995). Therefore, the low $(^{21}\text{Ne}/^{22}\text{Ne})_{\text{C}}$ ratio indicates that MIL 090657-NG resided within a few centimeters of its parent meteoroid. Otherwise, Na-rich chemical composition may also produce the low cosmogenic 21 Ne/ 22 Ne ratio due to its efficient production of ²²Ne (Leya et al., 2000). However, this is much less likely because exceptionally high Na (more than 10 wt%) content (e.g., 0.36 wt% in the mean CR chondrite; Kallemeyn et al., 1994) is required to reproduce the low $(^{21}\text{Ne}/^{22}\text{Ne})_{\text{C}}$ in MIL 090657-NG.

The production of cosmogenic noble gases also occurs near the surface of the air-less meteorite parent body. Solar-gas-rich regolith breccias were once exposed to the solar wind on the surface of the parent bodies and obtained solar wind noble gases via implantation. Such solar-gas-rich meteorites also obtained cosmogenic noble gases simultaneously with solar wind irradiation (e.g., Wieler et al., 1989; Meier et al., 2014; Obase et al., 2020). The solar-gas-rich composition indicates that NWA 801 had been exposed to GCR on the parent body regolith prior to the exposure in the parent meteoroid during the transit to the Earth. The $({}^{21}\text{Ne}/{}^{22}\text{Ne})_{\text{C}}$ ratio of 0.811 \pm 0.027 in NWA 801-NG is also below the estimated lowest GCR-induced ²¹Ne/²²Ne ratio in CR chondrites, suggesting the presence of SCR-induced Ne produced during the exposure on the parent body or in the parent meteoroid. The CRE ages for MIL 090657-NG and NWA 801-NG are not calculated because of the difficulties in correcting the SCR-induced Ne.

4.6. Solar-gas-poor composition for brecciated CR chondrite

Some CR chondrites display evidence for brecciation (e.g., Weisberg et al., 1993; Bischoff et al., 2006; Harju et al., 2014). MIL 090657 shows a brecciated texture that the meteorite contains abundant chondrule fragments and



Fig. 15. Schematic overview of the radial distributions of the Ar-rich noble gas component and the Q-like Ne-rich noble gas component in the early Solar System. PS is the protosun. OC, CC, and comets are the forming regions of ordinary chondrites, carbonaceous chondrites, and comets, respectively. The CO and CR chondrite forming regions are inside of CC. The Ar-rich component was commonly present in the OC and CC chondrite forming regions. The Q-like Ne-rich component was distributed in the outer region of the early Solar System where comets and the CR chondrites were formed.

some dark inclusions that are more aqueously altered than the host lithology (Fig. 1e and f).

The solar-gas-rich noble gas composition indicates that NWA 801 is a regolith breccia exposed to the solar wind for a long time on the surface regolith of the parent body ($\sim 10^7$ years; Nakashima et al., 2009). Hence, the brecciation for NWA 801 is attributable to cosmic dust impacts during the residence in the surface regolith. On the other hand, MIL 090657 does not contain detectable amounts of solar wind noble gases.

A lot of brecciated ordinary chondrites lack solar wind noble gases, and some of which might have lost noble gases by significant heating following large impacts (Bogard, 1995; Bischoff et al., 2018). However, the significant heating is ruled out for MIL 090657 because it also results in depletion of primordial noble gases found in this meteorite, including the Q-like Ne-rich component with a low release temperature. A possible explanation for the brecciated texture in solar-gas-poor MIL 090657 is that the brecciation occurred before the dissipation of the nebular gas that is opaque to the solar wind (Pascucci and Tachibana, 2010). Another possibility is that MIL 090657 did not acquire detectable amounts of solar wind noble gases due to very rapid brecciation.

4.7. Adsorbed atmospheric noble gases on MIL 090657-HT and MIL 090657-HTaq

The Xe isotopic ratios of MIL 090657-NG plot along the Q-HL mixing line in the Xe three-isotope diagram of ¹³⁰Xe/¹³²Xe vs. ¹³⁶Xe/¹³²Xe (Fig. 8b), indicating that the atmospheric Xe in MIL 090657 is negligible. However, the Xe isotopic ratios of MIL 090657-HT (especially in the 400-1000 °C fractions) are significantly affected by atmospheric Xe (Fig. 8d), which contributes $\sim 40\%$ of the total ¹³²Xe. In contrast, Kr isotopic ratios of MIL 090657-HT are less affected by the atmospheric component (Fig. 9d), and the isotopic ratios of He, Ne, and Ar in MIL 090657-HT and MIL 090657-HTaq do not show any significant atmospheric contribution. These isotopic ratios indicate that the Xe-enriched elemental ratios in the 400-800 °C fractions of MIL 090657-HT (Fig. 10c) are attributable to the adsorbed atmospheric noble gases significantly fractionated to the heavier elements.

Fanale and Cannon (1972) reported that heavier noble gases are efficiently adsorbed on a carbonaceous chondrite in a low-temperature condition. This suggests that the crushing procedure in liquid nitrogen (\sim 77 K) enhanced the adsorption of the atmospheric Xe onto MIL 090657-HT.

The atmospheric 132 Xe contribution on the total 132 Xe in MIL 090657-HTaq (~10%) is much less than that in MIL 090657, indicating that the hydrothermal treatment removed a large fraction of the adsorbed atmospheric Xe (Fig. 11e).

5. CONCLUSIONS

The detailed characterizations of petrology, mineralogy, and noble gas contents of the three CR chondrites EET

92048, MIL 090657, NWA 801, and the hydrothermally treated MIL 090657 samples revealed that aqueously less-altered CR chondrites contain two distinct watersusceptible noble gas components: the Ar-rich component and the Q-like Ne-rich component. Both components had been lost from aqueously altered CR chondrites.

The presence of the Ar-rich component in the CR chondrites, as well as in ordinary chondrites and other anhydrous carbonaceous chondrites (Alaerts et al., 1979; Matsuda et al., 1980; Schelhaas et al., 1990; Nakamura et al., 2003b; Krietsch et al., in press), indicates a global distribution of this component in the ordinary chondrite and carbonaceous chondrite forming regions in the early Solar System.

The characteristics of the Q-like Ne-rich component in MIL 090657 match the noble gases in cometary materials (Marty et al., 2008; Ogliore et al., 2020). The CR chondrites might have formed at a great heliocentric distance close to the comet forming region that results in the less-altered CR chondrites and comets sharing similar amorphous materials hosting this noble gas component.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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APPENDIX A. SUPPLEMENTARY MATERIAL

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