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High-Resolution Helium Isotope Imaging Reveals Solar Wind Helium Distribution in a Carbonaceous Chondrite

(ヘリウム同位体イメージングによる炭素質コンドライト
中の太陽風由来ヘリウム微細分布の解明)

A Dissertation Submitted for the Degree of Doctor of Philosophy

presented by

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ABSTRACT

Solar wind (SW) is preserved in meteorites as abundant noble gases derived from SW implantation. Noble gases are generally present in extremely low abundances in solid matter, making it possible to identify noble gases that are secondarily introduced by SW implantation. Since the projected range of typical SW particles into solid matter is less than 100 nm, SW-implantation occurs only when the grains reside in the uppermost surface of airless bodies. The SW irradiation duration is a key indicator of the timescales of geological processes on an asteroid. Furthermore, the distribution of SW noble gases in meteorites provide new insights into the mixing and sedimentation processes of the surface regolith on their parent body. In this thesis, I investigated the SW- ^4He distribution in a solar-gas-rich carbonaceous chondrite. This thesis consists of mainly two chapters as follows.

EVIDENCE OF SOLAR WIND IRRADIATION ON INDIVIDUAL MINERAL GRAINS: I performed *in situ* ^4He isotope imaging of individual mineral grains in the CR2 chondrite Northwest Africa 801 (NWA 801) using time-of-flight secondary neutral mass spectrometry with strong-field post-ionization. $^4\text{He}^+$ signals were detected along the surfaces of individual mineral grains of Fe-Ni metal, ferrihydrite, olivine, pyroxene, and troilite. The high ^4He concentrations along the surfaces indicate implantation of SW into the individual mineral grains. I determined the SW- ^4He fluence of eight individual mineral grains from the line profiles across the grain boundaries. SW- ^4He fluence ranged from 2.7×10^{16} to 5.8×10^{17} atoms cm^{-2} . I calculated the SW irradiation durations from these fluences. Assuming an irradiation occurred at 4 astronomical units, the durations translated to be between 3.8 and 82 kyr. These durations correspond to the residence time of individual mineral grains on the parent body surface.

The variation in residence time for the individual mineral grains suggests variable processes for local mixing and burial on the parent body and provides insights into the gardening rate driven by small-scale impact mixing processes on the parent body.

VISUALIZATION OF SEDIMENTARY STRUCTURE OF AN ASTEROIDAL REGOLITH IN A CARBONACEOUS CHONDRITE: By modifying the analysis program within the LIMAS operating system, I achieved ^4He imaging over an area of $100 \times 150 \mu\text{m}$, which is about 10 times larger than the previous analysis conditions. Improved *in situ* ^4He imaging technique elucidated the relationship between the heterogeneity of SW- ^4He and petrological characteristics in the NWA 801. The SW- ^4He -bearing mineral grains are predominantly distributed within the coarse grain-rich matrix. In contrast, the fine grain-rich matrix, composed of 1–5 μm grains and fine-grained groundmass, occurs as accessory rims around chondrules and an isolated olivine grain and also appears as scattered lumps ranging from several tens to hundreds of micrometers in size. No detectable ^4He was observed in the fine grain-rich matrix. Based on the similarities in occurrence, the fine grain-rich matrix observed in this study is thought to be the same material as fine-grained rims (FGRs). The partial preservation of FGRs along with the intact state of many chondrules, suggests that the daily impacts associated with gardening process primarily caused the disaggregation of loosely bound components, without sufficient energy to fracture chondrules. The heterogeneous distribution of SW- ^4He originated from low-energy impacts that caused the disaggregation of loosely bound components, which were the main processes of regolith gardening on the parent body of NWA 801. The proportions of SW- ^4He -bearing grains in opaque minerals and silicates are nearly identical (~20%), suggesting that SW irradiation occurred irrespective of mineral type, affecting both opaque minerals and silicates. If the regolith preserved in the

meteorite reflects the global surface characteristics of the NWA 801 parent body, the proportion of ^4He -bearing grains implies that approximately 20% of the unconsolidated surface layer was exposed to SW for a sufficiently long duration to accumulate detectable SW- ^4He , while the remaining 80% consisted of “immature regolith” that experienced minimal SW exposure (likely less than a few hundred years). The surface of the parent body of NWA 801 was likely composed largely of immature regolith, similar to present-day asteroids.

High-resolution ^4He isotope imaging revealed the distribution of SW- ^4He in the NWA 801 meteorite. The distribution of SW- ^4He in individual mineral grains is direct evidence of SW implantation on meteoritic grains. Quantification of SW- ^4He fluence of the individual mineral grains and the comprehensive investigation of the distribution of SW- ^4He in the interstitial matrix elucidate that gardening, which has sufficient energy to bury the SW- ^4He -enriched layer to a depth of several millimeters to centimeters, typically occurs on the parent body with a frequency of about ten thousand years.

CONTENTS

1. GENERAL INTRODUCTION.....	7
2. EVIDENCE OF SOLAR WIND IRRADIATION ON MINERAL GRAINS EMBEDDED IN MATRIX OF THE NORTHWEST AFRICA 801 CR CHONDRITE.....	10
2.1. INTRODUCTION	10
2.2. MATERIALS AND METHODS	12
2.2.1. Sample Preparation and Electron Microscopy	12
2.2.2. Helium Isotope Analysis.....	13
2.2.3. Spatial Resolution of Isotope Imaging Analysis.....	14
2.2.4. Quantification of ^4He Concentration	15
2.3. RESULTS	16
2.3.1. Petrography of Measurement Areas	16
2.3.2. ^4He Distribution of Mineral Grains	16
2.4. DISCUSSION.....	20
2.4.1. Evidence of SW Implantation.....	20
2.4.2. SW- ^4He Fluence of the Individual Mineral Grains	22
2.5. CONCLUSIONS.....	25
FIGURES.....	27
TABLES	37
3. VISUALIZATION OF SEDIMENTARY STRUCTURE OF AN ASTEROIDAL REGOLITH IN A CARBONACEOUS CHONDRITE.....	40
3.1. INTRODUCTION	40
3.2. INSTRUMENTAL MODIFICATIONS.....	42
3.3. MATERIALS AND METHODS	43
3.3.1. Isotope Imaging of ^4He -implanted Si Wafer	43
3.3.2. Sample preparation and electron microscopy for NWA 801	44

3.3.3.	Large-area Isotope Imaging of NWA 801	44
3.3.4.	Spatial Resolution of Isotope Imaging Analysis.....	45
3.3.5.	Identification of ^4He -bearing Individual Mineral Grain	45
3.3.6.	Quantification of ^4He Concentration	46
3.4.	RESULTS	46
3.4.2.	Petrography of measurement areas	47
3.4.3.	Isotope Maps	48
3.4.4.	^4He Distribution in the Interstitial Matrix	48
3.4.5.	Grain Size Distribution of Individual Mineral Grains	54
3.4.6.	SW- ^4He Concentration of Mineral Grains and Fine Grain-rich Matrix.....	54
3.5.	DISCUSSION.....	56
3.5.1.	Formation Process of the Heterogeneous Distribution of SW- ^4He	56
3.5.2.	Concentrations and Proportions of SW- ^4He -bearing Mineral Grains	59
3.6.	CONCLUSIONS.....	60
	FIGURES.....	62
	TABLES	91
4.	SUMMARY OF THIS THESIS.....	95
	REFERENCES	97
	SUPPLEMENTARY MATERIALS.....	105
	ACKNOWLEDGEMENTS	133

1. GENERAL INTRODUCTION

Noble gases are chemically inert. They are also highly volatile and were therefore overwhelmingly partitioned into the gas phase during the accretion of the early solar system. Consequently, they are extremely depleted in the solid matter due to their incompatibility in the solid phase. In fact, the terrestrial planets and their moons and asteroids are scarce in the noble gases (Wieler, 2002). The extremely low abundances of the noble gases in solid matter enable us to distinguish elementally and isotopically different noble gas components in extraterrestrial materials. These components reflect different noble gas reservoirs.

The elemental and isotopic ratios of noble gases in meteorites are diverse, indicating that these gases are a mixture resulting from different physical processes. Noble gas components include components that originate from in situ production, trapped components, and terrestrial atmospheric noble gases. In situ components include radiogenic noble gases and cosmogenic noble gases. Radiogenic noble gases are formed in situ by the decay of both long-lived radionuclides (e.g., ^4He , ^{40}Ar) and short-lived radionuclides (e.g., ^{129}Xe). Cosmogenic noble gases are produced in situ due to the interaction of galactic cosmic ray (GCR) (>0.1 GeV) particles with solid material. Trapped components are generally divided into primordial noble gases and solar wind (SW) noble gases. Meteorites contain various components of primordial noble gases. Among these are noble gases of nucleosynthetic origin found in presolar grains (nanodiamonds, silicon carbide) (e.g., Lewis et al., 1994; Huss and Lewis, 1994). The primordial noble gas called Q (P1) was probably trapped from the early solar nebula,

which is the most abundant reservoir for heavy noble gases (Ar, Kr, and Xe). The phase-Q is probably situated in carbonaceous phases of chondrites (Ott et al., 1981). SW noble gases are classified as trapped components, but their origin is known to be due to ion implantation by SW irradiation after the formation of the parent body surface as regolith (Rajan, 1974).

The present SW is mainly composed of H (~95 atom%) and He (5 atom%) and has an average energy of ~1 keV/nucleon corresponding to a continuous plasma stream of ~400 km s⁻¹ (Reisenfeld et al., 2007). Airless bodies such as the Moon and asteroids have been exposed to the SW. The SW irradiation contributes a significant amount of H and He to regolith materials on the uppermost surface of airless bodies. Such implanted SW noble gases have been identified from gas-rich meteorites, lunar regolith (e.g., Signer et al., 1977), and asteroidal samples collected from Itokawa (Nagao et al., 2011) and Ryugu (Okazaki et al., 2023). Among meteorites, those that contain significant amounts of noble gases are referred to as gas-rich meteorites. The existence of gas-rich meteorite was initially documented in 1956 (Gerling and Levsky, 1956). Suess et al. (1964) proposed that the origin of light noble gases in gas-rich meteorites is derived from SW. Eberhardt et al. (1965, 1966) have shown that a strong negative correlation exists between the trapped gas content and the grain size in the Khor Temiki aubrite and, moreover, that dissolution of a few percent of a pyroxene fraction removes almost all the trapped gases. From these experimental results they have concluded that the trapped gases are concentrated on the surface of the individual grains and that the gases represent most likely trapped low energy ions, for instance, SW particles. Based on these results and the observations of implanted SW in lunar regolith, it is widely accepted that the light noble gases found in gas-rich meteorites originate from SW.

The projected range of typical SW particles into solid matter is less than 100 nm (Futagami et al., 1990; Kuhlman et al., 2015; Wiens et al., 2004; Grimberg et al., 2008; Bajo et al. 2015). SW-implantation occurs only when the grains reside in the uppermost surface of airless bodies. Thus, the SW irradiation timescale of individual meteoritic grain is valuable for establishing the actual surface residence times and is a key indicator of the timescales of geological processes of the regolith on their parent body. Furthermore, the distribution of SW noble gases in meteorites provide new insights into the mixing and sedimentation processes of the surface regolith on their parent body.

In this thesis, I investigated the SW- ^4He distribution in a solar-gas-rich carbonaceous chondrite. In Chapter 2, I conducted the quantitative ^4He isotope imaging of individual mineral grains in the NWA 801 CR2 chondrite at sub-micrometer spatial resolution to elucidate the distribution of SW- ^4He on the surface regions of individual mineral grains and SW- ^4He irradiation duration for each mineral. In Chapter 3, I conducted comprehensive ^4He isotope imaging of NWA 801 at micrometer spatial resolution to elucidate the lithology of the surface regolith of their parent body. The instrumental modifications were performed to observe the ^4He distribution. The summary of this thesis is described in Chapter 4. It is noted that Chapter 2 is from a paper published in the *Meteoritics and Planetary Science* (Wada et al., 2025), and Chapter 3 is from a manuscript to be prepared for submitting to the *Meteoritics and Planetary Science*.

2. EVIDENCE OF SOLAR WIND IRRADIATION ON MINERAL GRAINS EMBEDDED IN MATRIX OF THE NORTHWEST AFRICA 801 CR CHONDRITE

2.1. INTRODUCTION

Evidence for solar wind (SW) implantation was first observed in the Khor Temiki aubrite meteorite (Eberhardt et al., 1965), classified as a gas-rich meteorite. Analysis of this meteorite revealed a negative correlation between noble gas concentrations and grain sizes for minerals in the matrix. Noble gas concentrations in different grain-size fractions of the aubrite were proportional to the surface-to-volume ratio, except for cosmogenic ^3He and ^{21}Ne . Similar correlations were observed in lunar samples (e.g., Hintenberger et al., 1970, 1971), suggesting that individual grains are enriched with SW noble gases on their surfaces.

Another approach to confirming SW implantation in grain surfaces is closed system step etching (CSSE) gas extraction, which employs acid leaching to extract noble gases from the surface layers of minerals. CSSE-based noble-gas analysis of gas-rich chondrites has shown that He- and Ne-rich components were released in the early etching steps, indicating their presence in the surface layers (Becker et al., 1998; Wieler et al., 2000; Riebe et al., 2017), and similar isotopic compositions to SW for He and Ne (Geiss et al., 1972; Heber et al., 2009). The $^4\text{He}/^{36}\text{Ar}$ and $^{20}\text{Ne}/^{36}\text{Ar}$ ratios in most gas-rich meteorites are lower than the SW ratios, indicating escape of He and Ne from the meteorites after SW implantation (Wieler et al., 1989; Lipschutz et al., 1993). On the

other hand, the $^4\text{He}/^{36}\text{Ar}$ and $^{20}\text{Ne}/^{36}\text{Ar}$ ratios of Fe-Ni metal in the gas-rich chondrites are nearly identical to the SW ratios, indicating that SW-He and -Ne in Fe-Ni metal are particularly retentive than those in silicates (Murer et al., 1997). These CSSE results provide evidence that SW noble gases are trapped in the near-surface layers of minerals via ion implantation.

In situ analysis can clarify the location and concentration of SW noble gases, and their distribution can be compared with petrographic observations. The conventional *in situ* noble gas analysis approach with laser ablation has achieved sub-millimeter-scale analysis of SW noble gases in meteorites. Nakamura et al. (1999) showed that SW noble gases are enriched in the clastic matrix but not in the pristine matrix of CM chondrites. *In situ* analysis with secondary neutral mass spectrometry (SNMS) achieved micrometer-scale ^4He analysis in solid materials (Bajo et al., 2015, 2016). Through laser ionization mass nanoscope (LIMAS) analysis, Bajo et al. (2024) showed that SW- ^4He was highly heterogeneously distributed grain by grain in the clastic matrix of a gas-rich meteorite, Northwest Africa 801 (NWA 801) CR2 chondrite. However, *in situ* analyses have not yet demonstrated that the SW-implanted ^4He resides at the very grain surface. NWA 801 contains a large amount of SW noble gases, about one order of magnitude higher than in other solar-gas-rich CR chondrites (Bischoff et al., 1993; Scherer and Schultz, 2000; Obase et al. 2021). Bajo et al. (2024) showed high ^4He concentration spots in the polished section of NWA 801. Nakashima et al. (2009) mentioned that NWA 852, the paired meteorite of NWA 801, was exposed to galactic cosmic rays on the parent body for several tens of million years. The previous studies suggest that NWA 801 preserves the regolith environments of the CR chondrite parent body.

In this study, I conducted *in situ* analysis of the ^4He isotope distribution using

LIMAS in individual mineral grains embedded in the NWA 801 section at a sub-micrometer spatial resolution, revealing the distribution of SW-⁴He on the surface region of individual mineral grains. Furthermore, the SW irradiation duration for each mineral and the implications for the history of regolith formation on the parent body are discussed.

2.2. MATERIALS AND METHODS

2.2.1. Sample Preparation and Electron Microscopy

A polished section of the NWA 801 meteorite was used in this study. A chip of the chondrite, embedded in an epoxy resin disk, was polished using an automatic polishing machine (MA-200e, Musashino Denshi) at Hokkaido University. The polished section was then cut into a $5 \times 7 \times 1 \text{ mm}^3$ cuboid using a low-speed diamond saw (IsoMet Low Speed, Buehler) for placement in a sample holder. To minimize degassing, the cut specimen was immersed in acetone for 1 day to remove the epoxy resin. Finally, the specimen surface was coated with an approximately 10 nm thick gold film via vacuum deposition.

Backscattered electron (BSE) images of the sample were obtained using a field emission scanning electron microscope (FE-SEM; JSM-7000F, JEOL) at Hokkaido University to characterize the petrography prior to isotope-imaging analysis. Quantitative elemental analysis and X-ray elemental mapping were performed using an energy-dispersive X-ray spectrometer (EDS; X-Max 150, Oxford) attached to the FE-SEM. A 15 keV electron beam probe was used with a current of 4 nA.

In this study, LIMAS analysis was conducted with a focus on individual mineral grains of the main constituent minerals in the NWA 801 meteorite (olivine, pyroxene, ferrihydrite, Fe-Ni metal, and troilite). Detailed analyses were performed on

the grains in which ^4He was detected.

2.2.2. Helium Isotope Analysis

Helium isotope analysis was conducted using an SNMS (LIMAS, JEOL) at Hokkaido University (Bajo et al., 2024). A primary ion beam of $^{69}\text{Ga}^+$ (30 keV and ~ 1 nA) was focused onto the sample surface using an aberration correction system (Bajo et al. 2016), and pulsed in 200 ns using a beam blanker. The sample surface was inclined at 55° to the primary beam, resulting in a vertical beam diameter 1.7 times larger than the horizontal diameter.

He isotope analysis was conducted using an SNMS (LIMAS, JEOL) at Hokkaido University (Bajo et al., 2024). In this study, we used a finer primary $^{69}\text{Ga}^+$ beam of 1 nA than that of Bajo et al. (2024) of 40 nA to achieve *in situ* analysis with a sub-micrometer spatial resolution. The primary ion beam with an energy of 30 keV was pulsed in 200 ns and focused onto the sample surface. The sample surface was inclined at 55° to the primary beam, resulting in a vertical beam diameter 1.7 times larger than the horizontal diameter.

Sputtered neutrals were photoionized under strong-field ionization conditions using a focused femtosecond laser (~ 5.5 mJ, Astrella, Coherent) installed in LIMAS and operating at a 1 kHz repetition rate. The laser was focused at a 30 μm diameter spot approximately ~ 50 μm above the sample surface. Ions with m/z 4 ($^4\text{He}^+$), 12 ($^{24}\text{Mg}^{2+}$), 16 ($^{32}\text{S}^{2+}$), 18.6 ($^{56}\text{Fe}^{3+}$), and 28 ($^{56}\text{Fe}^{2+}$, $^{28}\text{Si}^+$) were measured using a multi-turn time-of-flight (TOF) mass spectrometer, “MULTUM II” (Okumura et al., 2004; Totonani et al., 2016), with an acceleration voltage of -5 kV. The ion-injection system parameters for MULTUM II were optimized for high ion-transmission efficiency based

on the study by Tono et al. (2016). The flight time was adjusted to achieve a mass resolving power of $\sim 20,000$ (calculated using $T/2\Delta T$) for m/z 4. Interference ions, such as $^{12}\text{C}^{3+}$ and $^{16}\text{O}^{4+}$ for $^4\text{He}^+$, were removed using ion gates in the MULTUM II ion trajectory. A typical TOF mass spectrum from the sample is shown in Fig. 1. To obtain a 2D isotope image, the primary beam was scanned horizontally and vertically at 100 nm intervals. Each point in the image represents a pixel, and 600–6000 TOF mass spectra were collected for each pixel. The analytical conditions for the isotope-imaging analysis are summarized in Table 1.

The background signal of ^4He (hereafter referred to as blank ^4He) originates from residual ^4He gas above the sample surface. This was measured without the primary beam pulse after each isotope-imaging run. The $(1\text{--}2) \times 10^6$ TOF spectra were collected to determine the blank ^4He intensity, which varied with analytical conditions, ranging from 1×10^{-4} to 1×10^{-5} ion counts per mass spectrum scan (cpms). The background signals of ^{56}Fe , ^{24}Mg , and ^{32}S are negligible compared to their signals from the sample.

2.2.3. Spatial Resolution of Isotope Imaging Analysis

The spatial resolution for individual ion images was determined using line scan profiles of $^{56}\text{Fe}^{2+}$ and $^{24}\text{Mg}^{2+}$ (Figs. S1–S6) and $^4\text{He}^+$ (Figs. 3–8) across the interface between individual mineral grain and fine-grained groundmass, defined by the 16–84% criterion (Nagata et al., 2019). This is equivalent to the $\pm 1\sigma$ criterion for a Gaussian beam. The spatial resolutions for the individual analyses are listed in Table 2. Because the spatial resolutions calculated from the $^{56}\text{Fe}^{2+}$ and $^{24}\text{Mg}^{2+}$ profiles are broadened by the interface–surface angle, and the ^4He layer is narrower than 100 nm (see Section 4.2), the spatial resolution of the image in this study is defined as the width of the ^4He layer. The

resolutions varied with individual images: 100 nm for Areas 1, 2 and 3, 150 nm for Area 6 and 240 nm for Areas 4 and 5.

2.2.4. Quantification of ^4He Concentration

To determine the ^4He concentration in the sample, a ^4He -implanted Fe-Ni alloy ($\text{Fe}_{0.55}\text{Ni}_{0.45}$) plate was prepared as a standard, with an implantation dose of 3×10^{15} atoms cm^{-2} at an energy of 20 keV. The ^4He -implanted Fe-Ni standard was analyzed using LIMAS with a 30 keV and 32 nA $^{69}\text{Ga}^+$ primary ion beam, delivered in 400 ns pulses. A 30×30 pixel area ($12 \times 20 \mu\text{m}^2$) was scanned, and 100 TOF mass spectra were collected per pixel. Depth profiles of $^4\text{He}^+$, $^{56}\text{Fe}^{2+}$, and $^{58}\text{Ni}^{2+}$ were obtained from the central 5×5 pixels to minimize crater edge effects. Other measurement conditions were identical to those used for sample analysis. Following depth profiling, the crater depth was measured using a confocal laser microscope (VK-X200, Keyence).

Since the ion intensity is proportional to the primary beam current, I calculate the ^4He concentration using the ratio of the primary beam currents between the isotope imaging and the Fe-Ni standard measurement to correct for the ^4He concentration calculations in the sample. Because photoionization by a focused fs laser is less sensitive to matrix effects caused by differences in ionization efficiency depending on the sample material, the ^4He concentration was calculated using the following equation, adapted from Bajo et al. (2024):

$$[^4\text{He}] = (^4\text{He}^+)_{\text{meas}} / I_{p1} \times SF^{\text{He}} \quad (1)$$

where $[^4\text{He}]$, $^4\text{He}^+_{\text{meas}}$, I_{p1} , and SF^{He} denote the ^4He concentration in the measuring volume of a single pixel (atoms cm^{-3}), measured ^4He intensities (cpms), primary beam currents for sample, and sensitivity factor for ^4He (atoms cm^{-3} cpms $^{-1}$), respectively. The SF^{He} is

derived from the standard analysis as follows:

$$SF^{\text{He}} = \frac{\phi \times n}{\sum_{k=1}^n ((^4\text{He}^+)_{\text{meas}} - (^4\text{He}^+)_{\text{bg}})_k / I_{p2} \times d} \quad (2)$$

where Φ , k , and I_{p2} , d denote the ^4He fluence (atoms cm^{-2}) for the standard, number of measurement layer (from 1 to n), primary beam currents for standard, and sputtered crater depth (cm), respectively. Additionally, $(^4\text{He}^+)_{\text{bg}}$ denotes the blank ^4He .

2.3. RESULTS

2.3.1. Petrography of Measurement Areas

NWA 801 is classified as a CR2 chondrite (Connolly et al., 2007) and is highly enriched in SW noble gases (Obase et al., 2021). BSE and X-ray elemental images of the polished section of NWA 801 are shown in Fig. 2. SEM analysis reveals that the NWA 801 section contains millimeter-sized chondrules and their fragments embedded in matrix. Pervasive rust veins, primarily composed of ferrihydrite, which are typical terrestrial weathering products, are evident throughout the sample. The matrix contains mineral grains of Fe-Ni metal, ferrihydrite, olivine, pyroxene, and troilite, ranging from approximately 1 to 50 μm . These mineral grains are surrounded by a fine-grained groundmass typically less than 0.1 μm in size (Fig. 2c). Fe-Ni metal and troilite also occur within chondrule interiors and rims. Some Fe-Ni metal grains have been partially or completely replaced by ferrihydrite. In this study, we examined the mineral grains and the surrounding fine-grained groundmass in Areas 1–6, as shown in Fig. 2a.

2.3.2. ^4He Distribution of Mineral Grains

Enlarged BSE and $^4\text{He}^+$ images of Areas 1–6 are shown in Figs. 3–8. The ion images reveal that the mineral grains (<50 μm in size) of Fe-Ni metal (Figs. 3–5),

ferrihydrite (Figs. 4–7), olivine (Figs. 3, 6, and 8), pyroxene (Figs. 3, 6, and 7), and troilite (Fig. 3–8) are enriched in ^4He along part of their grain-surface contours. These mineral grains are embedded in a fine-grained groundmass. Line scans of $^{56}\text{Fe}^{2+}$ and $^{24}\text{Mg}^{2+}$ across a few selected grain boundaries illustrate the relationships between the boundaries and ^4He -concentrated regions. These line scans were obtained from a location with a high $^4\text{He}^+$ intensity to determine the largest fluence of SW irradiation for the grain.

Fe-Ni metal

Three Fe-Ni metal grains can be observed (Figs. 3–5) in Areas 1–3 (Fig. 2). In Area 1, the Fe-Ni metal grain is approximately $20 \times 20 \mu\text{m}^2$ (Fig. 3a). $^4\text{He}^+$ signals were detected along the edge of the grain, except for the lower right edge (Fig. 3b). Intensity changes in the line profiles for $^{56}\text{Fe}^{2+}$ and $^{24}\text{Mg}^{2+}$ delineate the grain boundary (Fig. 3c). Similar profile changes are observed for the other Fe-Ni metal grains (Figs. 4c, 4d, and 5c). The $^4\text{He}^+$ signal appears as a peak at the metal–groundmass interface with a width of 100 nm, defined as the full width at $e^{-\frac{1}{2}}$ height of the $^4\text{He}^+$ peak maximum, equivalent to $\pm 1\sigma$ for a Gaussian distribution. The peak ^4He concentration is 6.4×10^{21} atoms cm^{-3} (Fig. 3c), and the $^4\text{He}^+$ signals are below the detection limit in the metal's interior and the surrounding groundmass.

In Area 2, the Fe-Ni metal grain is approximately $5 \times 7 \mu\text{m}^2$ (Fig. 4a). $^4\text{He}^+$ signals are detected along the grain edge, except for the center-left edge (Fig. 4b). The $^4\text{He}^+$ profile exhibits a single peak with a 100 nm width ($\pm 1\sigma$) at the narrow distribution of the metal–groundmass interface. The peak ^4He concentration is 3.1×10^{21} atoms cm^{-3} (Fig. 4c), and the $^4\text{He}^+$ signals are below the detection limit in the metal's interior and the surrounding groundmass. The $^4\text{He}^+$ peak width in Line C–D (Fig. 4d) is wider than that in Line A–B (Fig. 4c), and a ^4He peak is observed within the Fe-Ni metal. This wider

$^4\text{He}^+$ distribution in Line C–D is attributed to the diagonal polishing of the grain edge.

In Area 3, the Fe-Ni metal grain is approximately $20 \times 30 \mu\text{m}^2$ (Fig. 5a). $^4\text{He}^+$ signals can be partially observed along the grain edge (Fig. 5b), whereas other areas are below the detection limit. The $^4\text{He}^+$ profile exhibits a single 100 nm wide peak ($\pm 1\sigma$). The peak ^4He concentration is $2.2 \times 10^{21} \text{ atoms cm}^{-3}$ (Fig. 5c).

Ferrihydrite

Two ^4He -rich ferrihydrite grains can be observed (Figs. 6 and 7) in Areas 4 and 5 (Fig. 2). In Area 4, the grain is approximately $10 \times 15 \mu\text{m}^2$ (Fig. 6a), and $^4\text{He}^+$ signals were detected along its edge (Fig. 6b). Line profiles of $^4\text{He}^+$, $^{56}\text{Fe}^{2+}$, and $^{24}\text{Mg}^{2+}$ across the grain boundary exhibit significant intensity changes at the boundary (Fig. 6c). The $^4\text{He}^+$ profile shows a single 250 nm wide peak ($\pm 1\sigma$), and the peak ^4He concentration is $17.0 \times 10^{21} \text{ atoms cm}^{-3}$ (Fig. 6c). The $^4\text{He}^+$ signals are below the detection limit in the ferrihydrite's interior, whereas weak ^4He intensities are detectable in the groundmass, with small ^4He -enriched spots (Fig. 6b).

In Area 5, the ferrihydrite grain is approximately $7 \times 8 \mu\text{m}^2$ (Fig. 7a), and $^4\text{He}^+$ signals are detected along its edge (Fig. 7b). Line profiles of $^{56}\text{Fe}^{2+}$ and $^{24}\text{Mg}^{2+}$ (Fig. 7c) show similar changes at the grain boundary as those observed in the ferrihydrite grain in Area 4. The $^4\text{He}^+$ profile shows a single 240 nm wide peak ($\pm 1\sigma$), and the peak ^4He concentration is $0.6 \times 10^{21} \text{ atoms cm}^{-3}$ (Fig. 7g). The $^4\text{He}^+$ signals in the ferrihydrite's interior are below the detection limit.

Although ferrihydrite grains were also present in Areas 2 and 3, detectable $^4\text{He}^+$ were not observed at the grain boundaries or within the grains (Figs. 4 and 5).

Olivine and Pyroxene

^4He -rich olivine and pyroxene grains were observed in the $^4\text{He}^+$ images of Areas 4 and 5, as shown in Figs. 6 and 7, respectively. In Area 4, the olivine grain is approximately $3 \times 2 \mu\text{m}^2$ (Fig. 6a), and $^4\text{He}^+$ signals are detected along its edge (Fig. 6b). The line profiles show a ^4He peak at the grain boundary (Fig. 6d) with a width of 230 nm ($\pm 1\sigma$) and a peak concentration of 0.9×10^{21} atoms cm^{-3} (Fig. 6d). The $^4\text{He}^+$ signals in the olivine's interior were below the detection limit, whereas the pyroxene grains in this area exhibited no detectable $^4\text{He}^+$ signals.

In Area 5, the pyroxene grain is approximately $2 \times 1 \mu\text{m}^2$ (Fig. 7a), and ^4He is observed along its edge (Fig. 7b). The $^4\text{He}^+$ peak in the line profile appears at the grain boundary with a width of 240 nm ($\pm 1\sigma$) (Fig. 7c) and a peak concentration of 0.7×10^{21} atoms cm^{-3} (Fig. 7h). The $^4\text{He}^+$ signals in the pyroxene's interior were below the detection limit and no ^4He was detected within other pyroxene grains in Area 5.

Troilite

Four ^4He -rich troilite grains were observed in the $^4\text{He}^+$ images of Areas 2, 4, 5, and 6, as shown in Figs. 4, 6, 7, and 8. The largest troilite grain of approximately $5 \times 5 \mu\text{m}^2$ (Fig. 8a) was observed in Area 6, and $^4\text{He}^+$ signals were detected along its edge, except for the upper and left edges (Fig. 8b). Line scans of $^4\text{He}^+$, $^{56}\text{Fe}^{2+}$, and $^{24}\text{Mg}^{2+}$ across the grain boundary were conducted to analyze the ^4He -concentrated region. As shown in Fig. 8c, a $^4\text{He}^+$ peak appears at the grain boundary with a width of 150 nm ($\pm 1\sigma$) and concentration of 2.5×10^{21} atoms cm^{-3} (Fig. 8c). However, ^4He is not detected within the grain or the surrounding fine-grained groundmass. The ^4He concentrations of the troilite grains in Areas 2, 4, and 5 are 2.9×10^{21} , 10.9×10^{21} , and 2.7×10^{21} atoms cm^{-3} , respectively. These grains are attached to ferrihydrite grains. Because the sizes of these grains were smaller than the spatial resolution of the analysis, the spatial distributions of

^4He within them could not be determined.

Fine-grained groundmass

All areas imaged in this study contained a fine-grained groundmass surrounding the mineral grains. In Areas 4 and 5, $^4\text{He}^+$ signals were detected from the groundmass (Figs. 6 and 7), with a typical concentration of approximately 0.6×10^{21} atoms cm^{-3} . ^4He -rich spots with concentrations of $1.8\text{--}8.4 \times 10^{21}$ atoms cm^{-3} were also observed, often corresponding to S and Fe enrichments (Figs. 6 and 7), suggesting that they represent fine-grained iron sulfides. The areas with $\sim 0.6 \times 10^{21}$ atoms cm^{-3} (blue areas in Fig. 6b, 6f, and 7b) appeared to be composed mainly of fine-grained silicates, having a concentration is similar to the peak concentrations of the olivine and pyroxene grains (Figs. 6d and 7h). By contrast, $^4\text{He}^+$ signals were not detected from the groundmass in Areas 1–3 and 6 (Figs. 3–5, 8). Therefore, I conclude that ^4He content varies significantly within the groundmass.

2.4. DISCUSSION

2.4.1. Evidence of SW Implantation

The line profiles of the mineral grains showed that the $^4\text{He}^+$ signals characteristically originated from the grain boundaries (Figs. 3–8). These distributions suggest that the ^4He is trapped either in the crystal edges or in the fine-grained groundmass attached to the crystals. In the former case, the trapped ^4He would be located at shallower depths than the spatial resolution of the image (~ 100 nm) from the crystal surface. Otherwise, the fine-grained groundmass, smaller than 100 nm, would contain ^4He .

The $^4\text{He}^+$ signals on the diagonally polished edge of the Fe-Ni metal grain demonstrated that the peak position is within the metal (Fig. 4d), providing evidence that ^4He is trapped within the metal itself. I first observe abundant ^4He at the grain surface due to the SW implantation into the grain. The ^4He images shown in Figs. 6 and 7 also validate that the ^4He peaks are within the minerals rather than the groundmass. By contrast, no observations showed groundmass containing ^4He concentrations comparable to the ^4He peak at the grain boundaries. The ^4He concentration in the groundmass was one to two orders of magnitude lower than that in the ^4He peaks at the grain boundaries. Therefore, it is plausible that ^4He is trapped at shallow depths from the surfaces of the mineral grains. The spatial resolution of the isotope images is not sufficient to observe the shallow SW implantation depth of approximately 20–40 nm from the surface of the minerals, which is expected by the TRIM (the Transport of Ions in Matter) code (Ziegler et al., 2012).

The ^4He -enriched layer beneath the grain surface, less than 100 nm thick, can be produced through ion implantation with kinetic energies of $\sim\text{keV/nucleon}$ (Futagami et al., 1990; Kuhlman et al., 2015; Wiens et al., 2004; Grimberg et al., 2008; Bajo et al. 2015). In interplanetary space, such low-energy ions are delivered by the SW. The ^4He enrichment observed within 100 nm for the Fe-Ni metal grains (Fig. 3c) is consistent with the typical SW range of 20–40 nm from the surface. Therefore, I conclude that the ^4He line profiles in Figs. 3–8 provide direct evidence of SW implantation into the mineral grains. These grains were exposed to SW on the surface of the parent body and mechanically mixed with other meteorite precursor materials (Hashiguchi et al., 2013) to form the current structure of NWA 801 by subsequent compaction in the parent body (Charles et al., 2018).

Additionally, abundant SW- ^4He was observed at the edges of the ferrihydrite

grains (Figs. 6 and 7). Ferrihydrite is a typical terrestrial weathering product (Noguchi, 1994), and trapped light noble gases are considered to be released during terrestrial weathering (Scherer et al., 1994; Stelzner et al., 1999; Bland et al., 2006). The presence of SW- ^4He in ferrihydrite suggests that the implanted ^4He remained in the ferrihydrite even though the original phase, likely metal, weathered to ferrihydrite on Earth. Another interpretation is that the ferrihydrite formed during aqueous alteration on the parent body, similar to that suggested for CI chondrites by Tomeoka and Buseck (1988). In later scenario, the altered ferrihydrite grains were exposed to SW on the parent body after formation. The results of this study do not allow us to determine which process is more plausible for the formation of ferrihydrite grains. However, in either case, the ^4He distributions in the ferrihydrite grains were produced by SW irradiation.

Bajo et al. (2024) using micrometer ^4He analysis firstly showed that ^4He is highly concentrated and sparsely scattered in the matrix in a gas-rich meteorite. This study advanced their findings through sub-micrometer ^4He analysis and is the first to identify the ^4He concentrated on the surfaces of individual mineral grains. Thus, it provides direct evidence for SW implantation into the constituent minerals of the meteorite. Furthermore, the observed heterogeneity of SW- ^4He implantation may be related to the regolith formation dynamics on the parent body.

2.4.2. SW- ^4He Fluence of the Individual Mineral Grains

Because the ^4He was implanted by SW irradiation (with a typical energy of ~ 1 keV/nucleon), the actual peak width should be several tens of nanometers. The observed line profiles (Figs. 3–8) were likely broader than the true profiles and exhibited lower peak concentrations because of the ~ 100 nm spatial resolution of the images.

However, the integral of the ^4He concentrations in each line profile represents the total amount of ^4He in the sputtered volume. Therefore, the SW- ^4He fluence, Φ_{sample} , can be calculated from the line profile as follows:

$$\Phi_{\text{sample}} = \sum_{k=1}^n (C_{\text{sample}} \times L)_k \quad (3)$$

where C_{sample} is the ^4He concentration (atoms cm^{-3}) and L is the pixel width of ^4He (cm). This method enables direct quantification of the SW- ^4He fluence to the grain surface.

The SW- ^4He fluences of the mineral grains are summarized in Table 3. The SW- ^4He fluences for the ferrihydrite grains in Areas 4 and 5 are $(58.4 \pm 2.2) \times 10^{16}$ and $(3.0 \pm 0.2) \times 10^{16}$ atoms cm^{-2} , respectively. Two ferrihydrite grains in Areas 4 and 5 were analyzed in the same analytical session, with one exhibiting the highest $^4\text{He}^+$ intensity and the other a relatively low $^4\text{He}^+$ intensity. Similar $^{56}\text{Fe}^{2+}$ signal emissions, a major element of ferrihydrite, were measured from the two grains (Figs 6, 7). Therefore, the differences of $^4\text{He}^+$ fluences are not artifact and the calculated ^4He concentration in Fig. 6c is 26 times higher than that in Fig. 7g. These results suggest that the ferrihydrite in Area 4 has the highest SW- ^4He fluence. The SW- ^4He fluences for the Fe-Ni metal grains range from (3.7 ± 0.8) to $(11.5 \pm 1.7) \times 10^{16}$ atoms cm^{-2} . The troilite grain in Area 6 has a fluence of $(5.6 \pm 0.9) \times 10^{16}$ atoms cm^{-2} . The pyroxene and olivine have fluences of (2.7 ± 0.2) and $(2.9 \pm 0.5) \times 10^{16}$ atoms cm^{-2} , respectively. These values range from 2.7×10^{16} to 58.4×10^{16} atoms cm^{-2} , representing an approximately 20-fold difference in the SW- ^4He fluence. Because SW- ^4He in minerals may not be quantitatively retained in regolith samples (e.g., Signer et al. 1977), it should be noted that the ^4He fluences obtained in this study represent lower limits. Fe-Ni metals in gas-rich meteorites have the $^4\text{He}/^{20}\text{Ne}$ ratio of ~ 630 and $^4\text{He}/^{36}\text{Ar}$ ratio of $\sim 30,000$ (Murer et al. 1997), nearly identical

to the SW ratios of 648 and 27,300, respectively (Heber et al. 2009). The bulk $^4\text{He}/^{20}\text{Ne}$ ratio in NWA 801 is 458 (Obase et al. 2021), which indicates at least $\sim 30\%$ of ^4He escaped from NWA 801. Thus, the ^4He loss of the individual grains, especially Fe-Ni metal grains, is relatively small so we do not consider ^4He loss in this discussion.

Based on these large variations in SW- ^4He fluence, I can estimate the SW irradiation duration for each mineral grain. Assuming $1/\pi$ SW exposure owing to the rotation of the parent body, the SW irradiation duration, T (yr), is calculated as follows:

$$T = \Phi_{\text{sample}} / (\varphi_{\text{pb}} \times \frac{1}{\pi}) \quad (4)$$

The SW flux φ_r (atoms $\text{cm}^{-2} \text{yr}^{-1}$) decreases with the square of the distance, r (astronomical unit: au), from the Sun (Adhikari et al., 2014). The SW flux at the parent body φ_{pb} (atoms $\text{cm}^{-2} \text{yr}^{-1}$) is calculated as

$$\varphi_{\text{pb}} = \varphi_{1\text{au}} \times \frac{1}{r^2} \quad (5)$$

Obase and Nakashima (2023) showed that the SW flux recorded in solar-gas-rich meteorites is comparable to the current levels. The present SW- ^4He flux at 1 au ($\varphi_{1\text{au}}$) is 3.57×10^{14} atoms $\text{cm}^{-2} \text{yr}^{-1}$ (Heber et al., 2009). The heliocentric distance of the CR chondrite parent body during SW irradiation might have been between the current main asteroid belt (2.2–3.3 au; DeMeo and Carry, 2014) and the presumed formation region of the CR chondrite parent body (4 au; Desch et al., 2018). The calculated SW irradiation durations range from 1.1 to 25 kyr at 2.2 au, 2.6 to 56 kyr at 3.3 au, and 3.8 to 82 kyr at 4 au (Fig. 9, Table 3).

The SW irradiation durations represent the residence time of the individual mineral grains on the top surface of the parent body, reflecting the timescale of local mixing on a millimeter-scale in its regolith. If irradiation occurred at 4 au, the residence

times of the seven out of the eight individual mineral grains are 3.8–16 kyr, which is consistent with the ranges observed for Ryugu samples (≤ 10 kyr; Okazaki et al., 2023) and lunar regolith (3.8–13 kyr; Signer et al., 1977; Li et al., 2023). These estimated SW irradiation durations suggest that the typical timescale of millimeter-scale regolith mixing on the parent body was about ten thousand years. The high ^4He fluence for the ferrihydrite grain in Area 4 (Fig. 6) suggests a SW irradiation duration of 82 kyr, which is more than ten times longer than that of the other grains. This exceptionally high ^4He fluence suggests that the local mixing process lasted longer than that for the other grains. Additionally, the long residence time may indicate that this is an exotic regolith grain incorporated from a location where the mixing process is less frequent. If this is indeed the case, this implies that there is a horizontal transportation mechanism for regolith (e.g., electrostatic levitation; Lee, 1996). Such large variations in regolith mixing frequencies, over several tens of times, have been observed on the surface of the asteroid Vesta (Denevi et al., 2016).

2.5. CONCLUSIONS

I presented a novel technique for observing past SW-implanted ^4He from individual mineral grains in meteorites and a method for quantifying SW- ^4He fluence within them. *In situ* ^4He isotope imaging with LIMAS, applied to the CR2 chondrite NWA 801, provided the first direct evidence of SW implantation in individual grains of a solar-gas-rich meteorite. The isotope images of Fe-Ni metal, ferrihydrite, olivine, pyroxene, and troilite grains showed that ^4He is detected along grain interfaces, indicating its presence at depths shallower than 100 nm from the grain surfaces. These results demonstrated that SW implantation occurred in the individual grains. Additionally, the existence of SW- ^4He in the ferrihydrite suggested two possibilities: either the implanted

^4He has survived in the ferrihydrite formed by terrestrial weathering of metal, or that formed through aqueous alteration on the parent body before SW exposure. The SW- ^4He fluence of the mineral grains ranged from $(2.7 \text{ to } 58) \times 10^{16} \text{ atoms cm}^{-2}$. Assuming irradiation occurred at 4 au, this corresponds to SW irradiation durations ranging from 3.8 to 82 kyr, representing the residence time of the individual mineral grains on the surface of the parent body. The residence times of most mineral grains suggest that the local mixing process on the parent body typically occurred with a frequency of about ten thousand years.

FIGURES

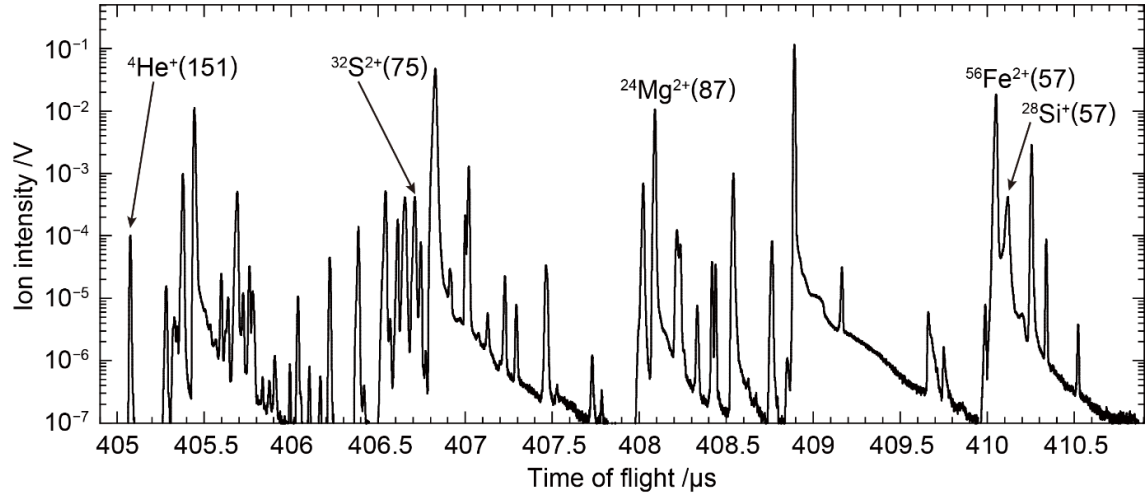


Figure 1. TOF mass spectrum of Area 4 of the NWA 801 sample shown in Fig. 6b. The arrows indicate the measured ion species, where the numbers in parentheses indicate the numbers of laps for each ion in MULTUM-II.

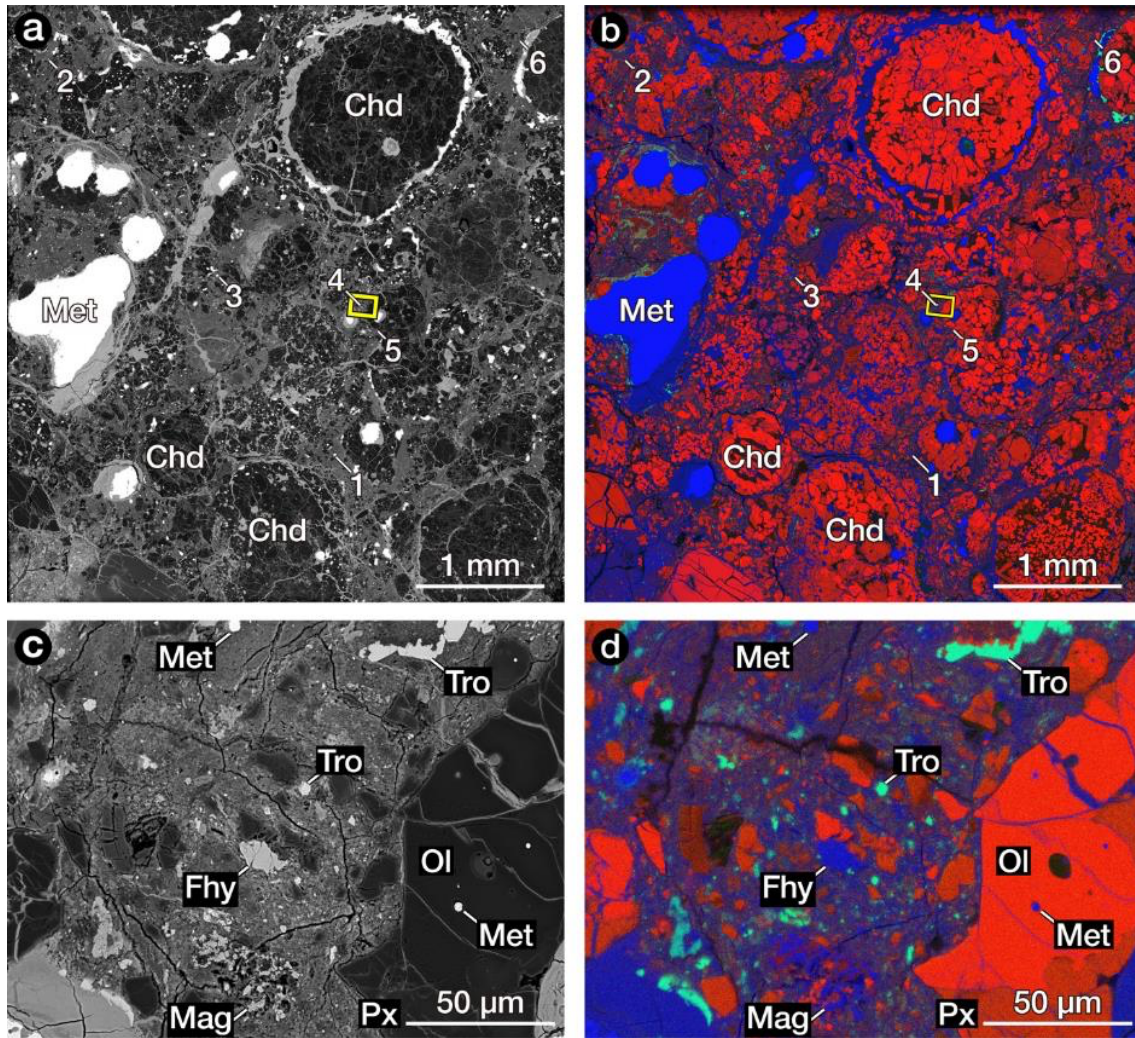


Figure 2. (a) BSE image of a section of NWA 801. (b) Composite X-ray elemental map of the area shown in (a), with Mg (red), Si (green), and Fe (blue). (c) Magnified BSE image of the area indicated by yellow boxes in (a) and (b). (d) Composite X-ray elemental map of the area shown in (c), with Mg (red), Si (green), and Fe (blue). Numbers 1–6 correspond to locations shown in Figs. 3–8, respectively. Abbreviations: Chd, chondrule; Fhy, ferrihydrite; Mag, magnetite; Met, Fe-Ni metal; Ol, olivine; Px, pyroxene; Tro, troilite.

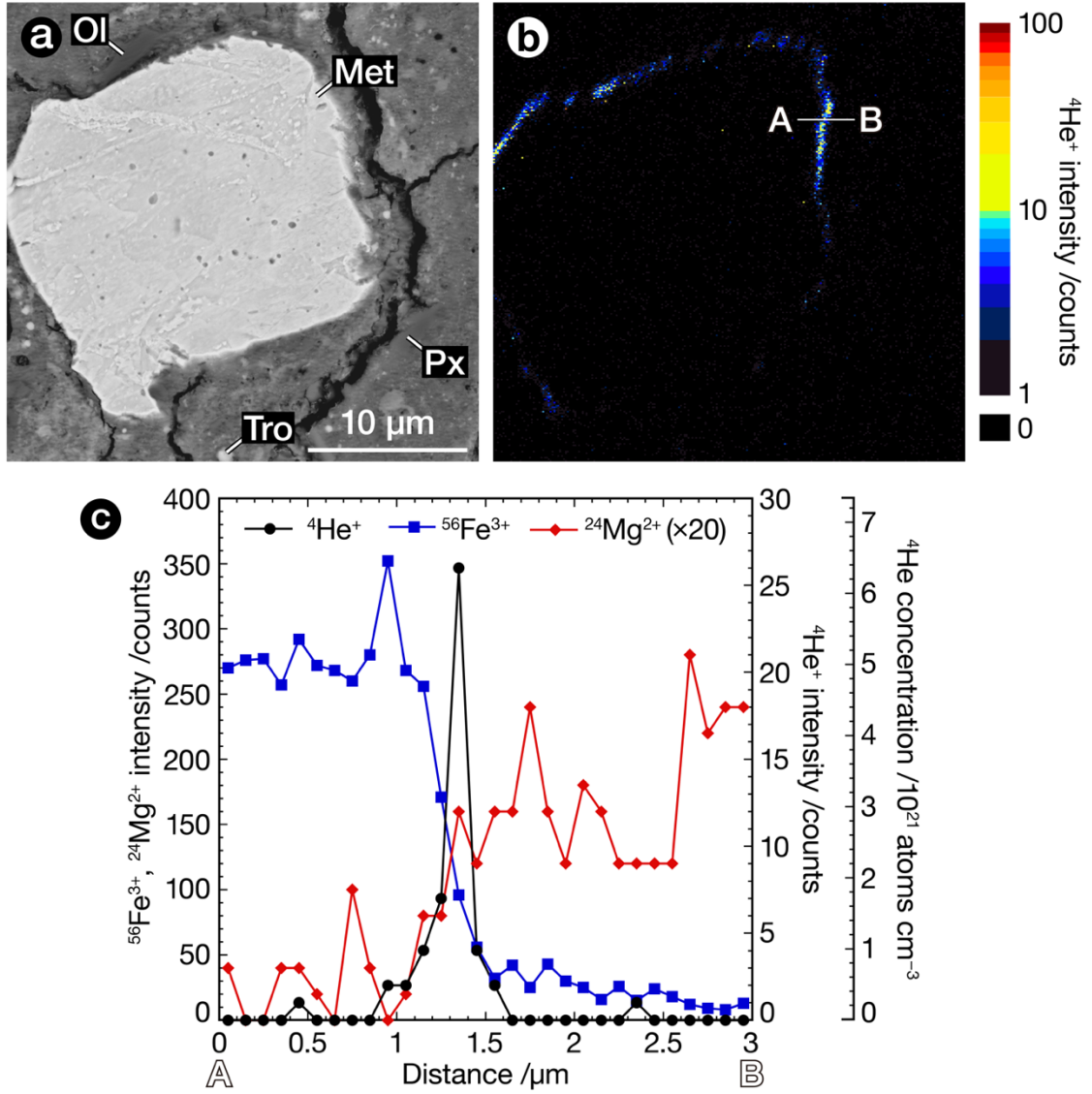


Figure 3. (a) BSE image of Area 1. (b) $^4\text{He}^+$ image of the area delineated by the yellow box in (a). (c) Line profiles for $^4\text{He}^+$, $^{56}\text{Fe}^{3+}$, and $^{24}\text{Mg}^{2+}$ along traverse A–B across the metal interface. Abbreviations: Met, Fe–Ni metal; Ol, olivine; Px, pyroxene; Tro, troilite.

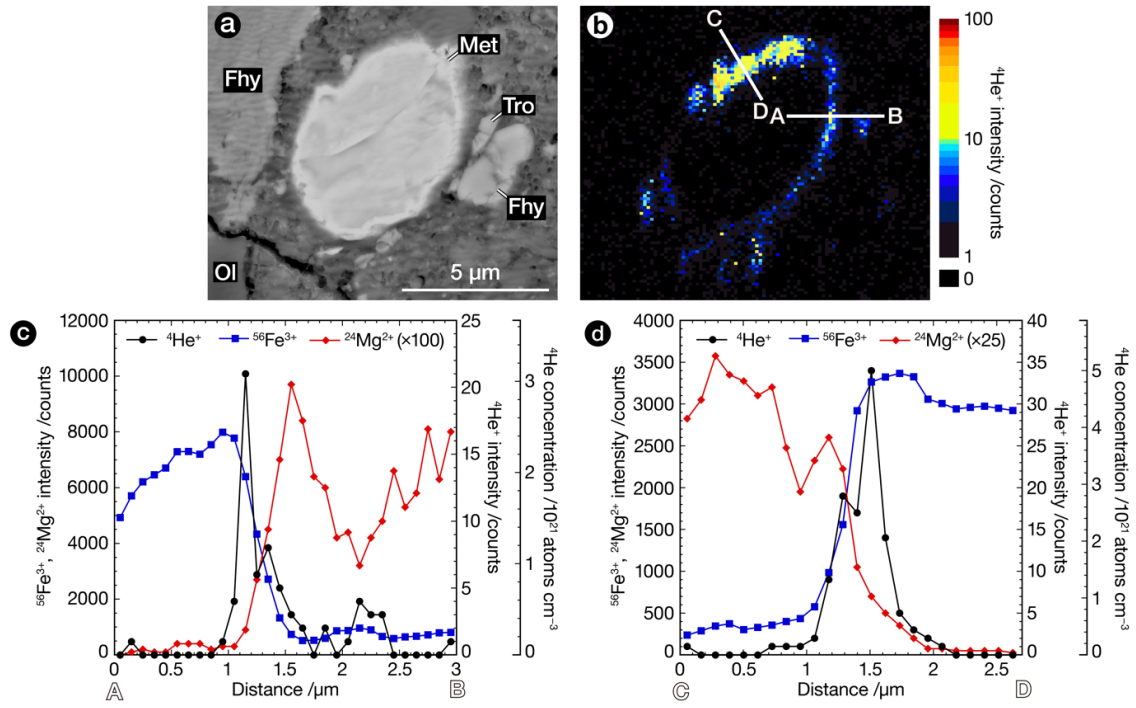


Figure 4. (a) BSE image of Area 2. (b) $^4\text{He}^+$ image of the area delineated by the yellow box in (a). (c) Line profiles for $^4\text{He}^+$, $^{56}\text{Fe}^{2+}$, and $^{24}\text{Mg}^{2+}$ along traverse A–B across the metal interface in (b). (d) Line profiles for $^4\text{He}^+$, $^{56}\text{Fe}^{2+}$, and $^{24}\text{Mg}^{2+}$ along traverse C–D across the metal interface in (b). Abbreviations: Met, Fe–Ni metal; Ol, olivine; Tro, troilite.

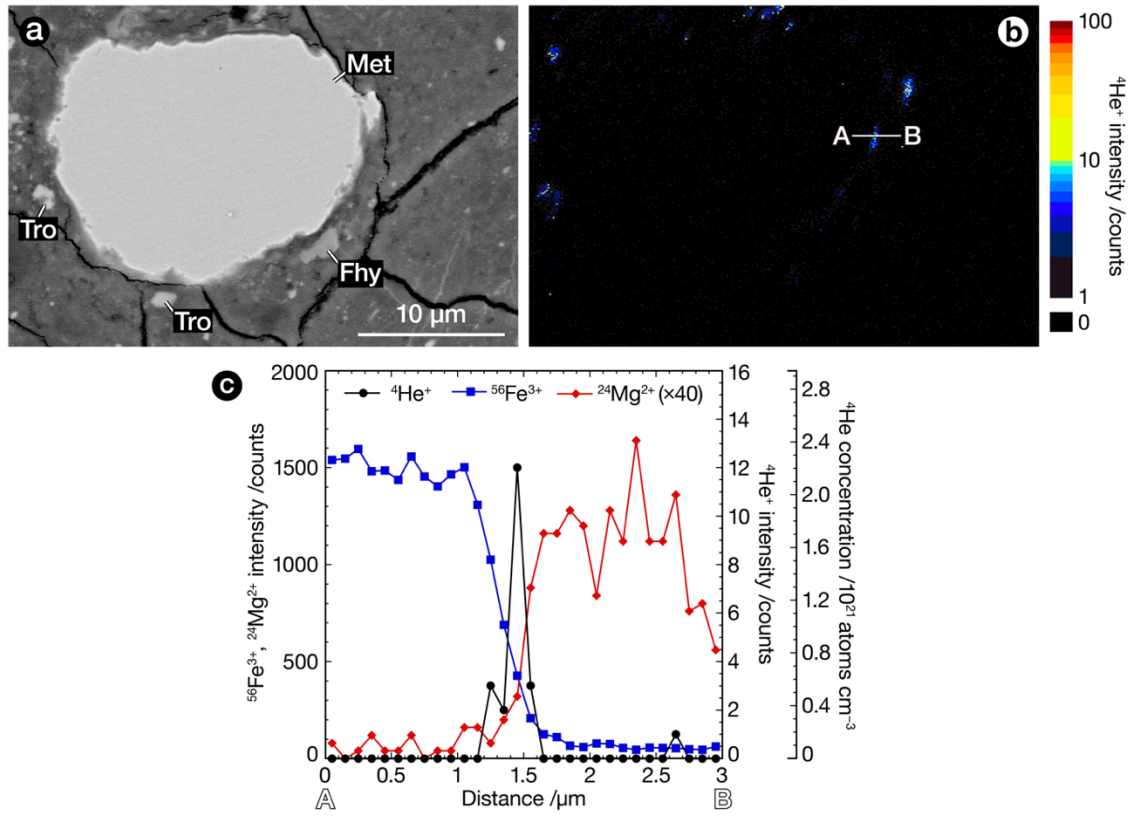


Figure 5. (a) BSE image of Area 3. (b) $^4\text{He}^+$ image of the area delineated by the yellow box in (a). (c) Line profiles for $^4\text{He}^+$, $^{56}\text{Fe}^{3+}$, and $^{24}\text{Mg}^{2+}$ along traverse A–B across the metal interface in (b). Abbreviations: Met, Fe–Ni metal; Tro, troilite.

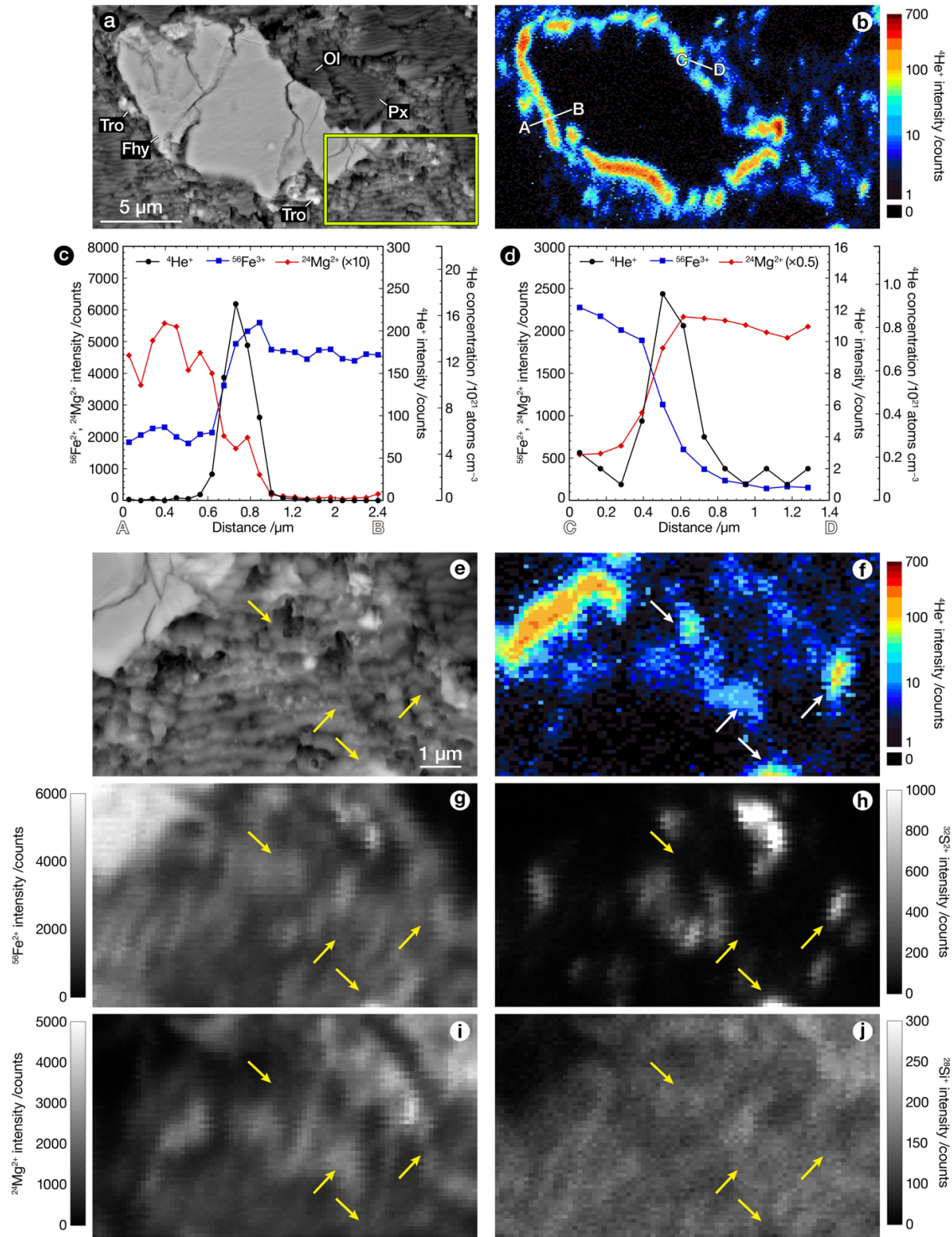


Figure 6. (a) BSE image of Area 4. (b) $^4\text{He}^+$ image of the area delineated by the yellow box in (a). (c) Line profiles for $^4\text{He}^+$, $^{56}\text{Fe}^{2+}$, and $^{24}\text{Mg}^{2+}$ along traverse A–B across the ferrihydrite interface in (b). (d) Line profiles for $^4\text{He}^+$, $^{56}\text{Fe}^{2+}$, and $^{24}\text{Mg}^{2+}$ ion intensities along traverse C–D across the pyroxene interface in (b). (e) Magnified BSE image of Area 4 delineated by the small yellow box in (a). (f) $^4\text{He}^+$ image of Fig. 6e. (g) $^{56}\text{Fe}^{2+}$

image of Fig. 6e. (h) $^{32}\text{S}^{2+}$ image of Fig. 6e. (i) $^{24}\text{Mg}^{2+}$ image of Fig. 6e. (j) $^{28}\text{Si}^{2+}$ image of Fig. 6e. The yellow arrows in (e) and (g)–(j) and white arrows in (f) indicate the locations of $^4\text{He}^+$ enriched spots in the groundmass. Abbreviations: Fhy, ferrihydrite; Ol, olivine; Px, pyroxene; Tro, troilite.

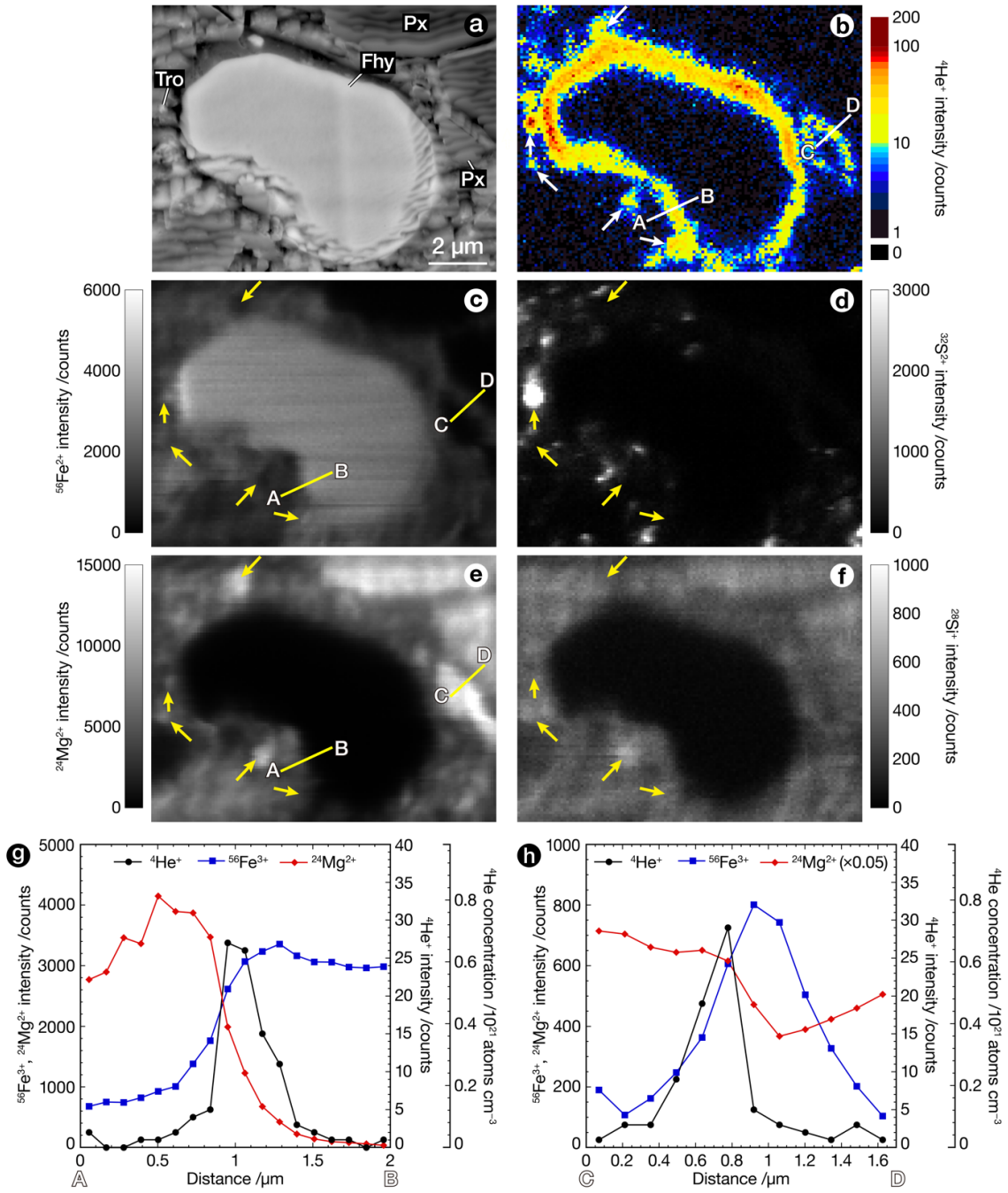


Figure 7. (a) BSE image of Area 5. (b) $^4\text{He}^+$ image of the area delineated by the yellow box in (a). (c) $^{56}\text{Fe}^{2+}$ image of Area 5. (d) $^{32}\text{S}^{2+}$ image of Fig. 7b. (e) $^{24}\text{Mg}^{2+}$ image of Fig. 7b. (f) $^{28}\text{Si}^{2+}$ image of Fig. 7b. (g) Line profiles for $^4\text{He}^+$, $^{56}\text{Fe}^{2+}$, and $^{24}\text{Mg}^{2+}$ along traverse A–B in (b). (h) Line profiles for $^4\text{He}^+$, $^{56}\text{Fe}^{2+}$, and $^{24}\text{Mg}^{2+}$ ion intensity along traverse C–D in (b). The yellow arrows in (a)–(f) indicate the locations of $^4\text{He}^+$ enriched spots in the groundmass. Abbreviations: Fhy, ferrihydrite; Px, pyroxene; Tro, troilite.

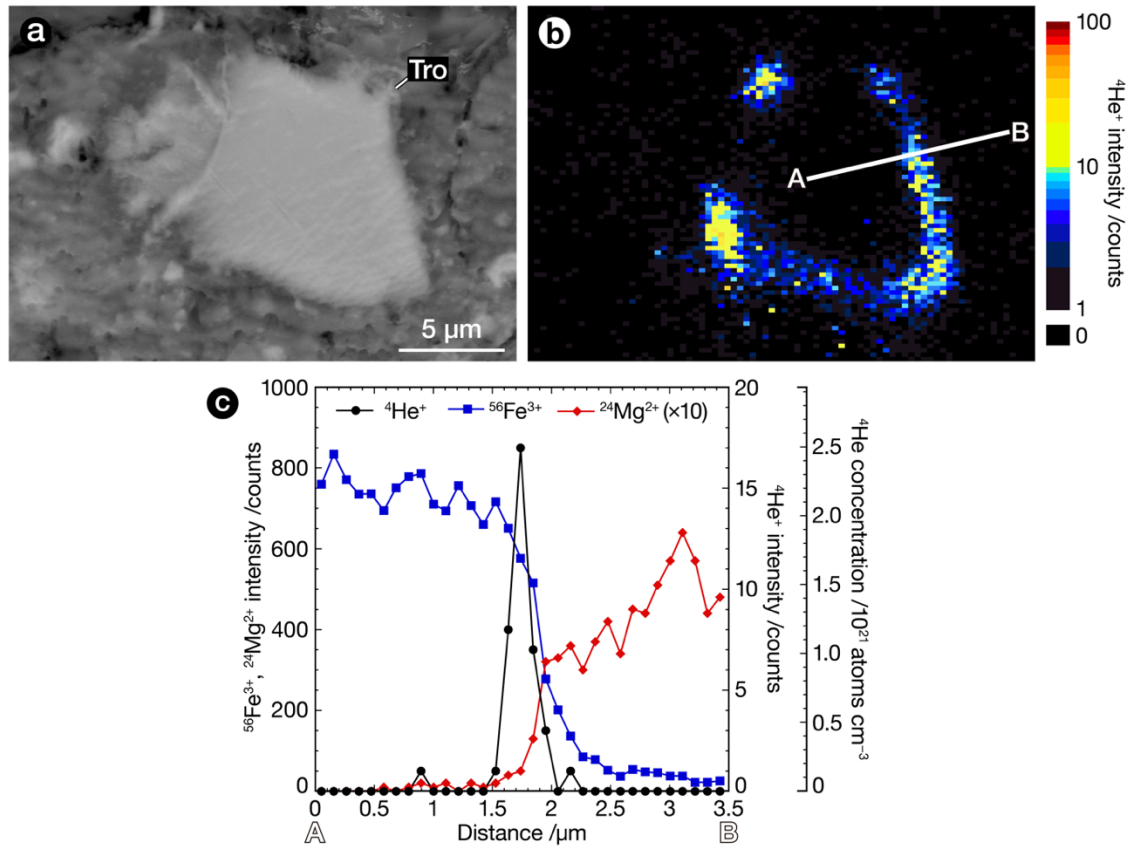


Figure 8. (a) BSE image of Area 6. (b) $^4\text{He}^+$ image of the area delineated by the yellow box in (a). (c) Line profiles for $^4\text{He}^+$, $^{56}\text{Fe}^{3+}$, and $^{24}\text{Mg}^{2+}$ along traverse A–B in (b). Abbreviations: Ol, olivine; Tro, troilite.

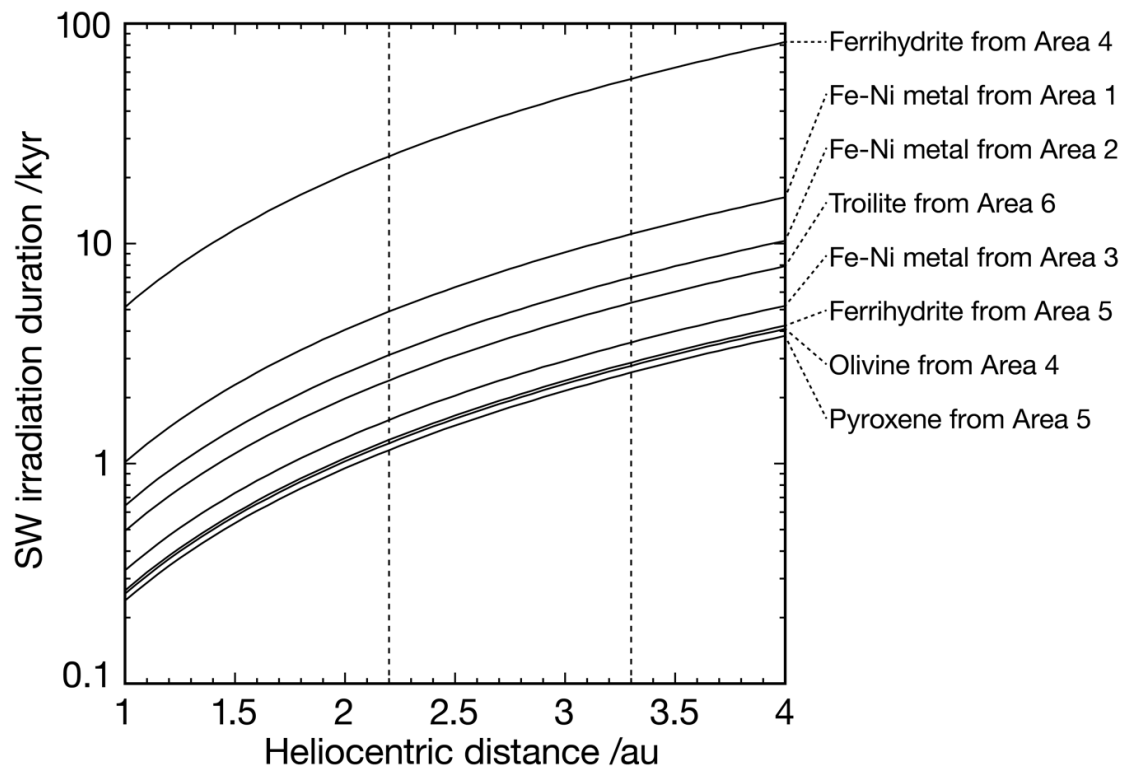


Figure 9. SW irradiation duration for individual mineral grains.

TABLES

Table 1. Analytical conditions for isotope imaging.

Area	Image size / μm^2	Pixels	Num. of mass scan per pixel
1	25×30	250×300	600
2	10×10	100×100	1000
3	30×25	300×250	800
4	10×12	100×120	2000
5	15×20	150×200	6000
6	8×8	80×80	1000

Table 2. Spatial resolutions of isotope images.

Area	Mineral	$^4\text{He}^+$ * /nm	$^{56}\text{Fe}^{2+, 3+}$ ** /nm	$^{24}\text{Mg}^{2+}$ ** /nm
1	Fe-Ni metal	100	260	240
2	Fe-Ni metal	100	260	300
3	Fe-Ni metal	100	350	220
4	Ferrihydrite	250	160	440
4	Olivine	230	270	210
5	Ferrihydrite	240	310	330
5	Pyroxene	240	300	170
6	Troilite	150	450	400

*Peak width of $^4\text{He}^+$, **16–84% width from the line scan (Figs. S1–S6)

Table 3. SW-⁴He fluences and SW irradiation durations for mineral grains.

Area	Mineral	Line profile	⁴ He fluence	SW irradiation duration /kyr		
			/10 ¹⁶ atoms cm ⁻²	2.2 au	3.3 au	4.0 au
1	Fe-Ni metal	A-B	11.5 ± 1.7	4.9 ± 0.7	11.0 ± 1.6	16.2 ± 2.4
2	Fe-Ni metal	A-B	7.3 ± 1.0	3.1 ± 0.4	7.0 ± 1.0	10.3 ± 1.4
3	Fe-Ni metal	A-B	3.7 ± 0.8	1.6 ± 0.3	3.6 ± 0.8	5.2 ± 1.1
4	Ferrihydrite	A-B	58.4 ± 2.2	24.9 ± 0.9	56.1 ± 2.1	82.4 ± 3.1
4	Olivine	C-D	2.9 ± 0.5	1.2 ± 0.2	2.8 ± 0.5	4.1 ± 0.7
5	Ferrihydrite	A-B	3.0 ± 0.2	1.3 ± 0.1	2.9 ± 0.2	4.2 ± 0.3
5	Pyroxene	C-D	2.7 ± 0.2	1.1 ± 0.1	2.6 ± 0.2	3.8 ± 0.3
6	Troilite	A-B	5.6 ± 0.9	2.4 ± 0.4	5.4 ± 0.9	7.9 ± 1.3

Errors in 1 σ

3. VISUALIZATION OF SEDIMENTARY STRUCTURE OF AN ASTEROIDAL REGOLITH IN A CARBONACEOUS CHONDRITE

3.1. INTRODUCTION

Origin of SW noble gas in gas-rich meteorites is the past SW into the regolith material on the surface of their parent body (Wieler et al., 1989). Before gas-rich meteorites were finally lithified as meteorites, the surface regolith materials were mechanically mixed with subsurface materials through impact gardening, leading to gas-rich meteorites commonly exhibiting a brecciated appearance with a mixture of solar-gas-poor portion so called “light clast” and solar-gas-rich portion so called “dark matrix” (Suess et al., 1964). Although the chemical compositions of minerals in the light clast and dark matrix are identical, there are petrographically differences. In non-carbonaceous chondrite, the light clast with various shapes (as angular, round, and oval), are generally scattered in the dark matrix (Wlotzka, 1963; Fredriksson and Keil, 1963). Since carbonaceous chondrites are inherently dark, the light-dark structure associated with SW noble gas concentrations have not been observed, except for rare cases such as the CM chondrite Nogoya (Heymann and Mazor, 1967).

Solar-gas-rich portions in gas-rich meteorites have heterogeneity in SW noble gas concentrations (Wieler et al., 1989; Pun et al., 1998; Nakamura et al., 1999). The presence of this heterogeneity indicates that the solar-gas-rich portion is a mixture of materials with various irradiation histories, and that the heterogeneous mechanical mixing

of the surface regolith of their parent body has been preserved in gas-rich meteorites. Therefore, clarifying the relationship between the heterogeneity of SW noble gas abundance in materials derived from the surface regolith of gas-rich meteorites and their petrologic characteristics can provide new insights into the mixing and sedimentation processes of the surface regolith on their parent body.

In situ analysis can clarify the location and concentration of SW noble gases, and their distribution can be compared with petrographic observations. The conventional *in situ* noble gas analysis approach using laser ablation with sub-millimeter spatial resolution revealed the heterogeneous SW noble gas concentrations within the clastic matrix of CM chondrites (Nakamura et al., 1999). *In situ* ^4He isotope imaging using laser ionization mass nanoscope (LIMAS) with sub-micrometer spatial resolution showed that SW- ^4He was implanted on the surface of individual mineral grains distributed in the matrix of the gas-rich CR chondrite NWA 801, demonstrating that there is variation in SW- ^4He fluence among individual mineral grains (Wada et al., 2025). However, the relationship between the petrologic characteristics and the distribution of SW- ^4He -bearing individual mineral grains in meteorites, as well as the relationship between the concentrations of SW- ^4He and the grain size distribution and mineral phase of individual mineral grains, remain unknown.

In this study, I improved the isotope imaging method, which can measure the distribution of ^4He with a spatial resolution (micrometer spatial resolution) that can identify the distribution of mineral grains in meteorites, and expanded the imaging area, which is about 10 times larger than the previous analysis conditions. By using the improved method to conduct ^4He isotope imaging of the NWA 801, I clarified the relationship between the heterogeneity of SW- ^4He and petrologic characteristics.

Furthermore, the implications for the history of regolith formation on the parent body of carbonaceous chondrites are discussed.

3.2. INSTRUMENTAL MODIFICATIONS

I used LIMAS for He isotope analysis (Wada et al., 2025). Figure 1 illustrates the geometry of sample, laser beam, and primary ion beam in the sample chamber. The post-ionization area is limited by the focused diameter of laser beam in the direction perpendicular to the center of laser beam. In general, the beam diameter of a laser beam is defined as a width of 13.5% ($1/e^2$) of the peak intensity. The focused diameter (ϕ) of the laser beam is expressed as follows:

$$\phi = \frac{4\lambda f M^2}{\pi D} \quad (6)$$

where λ , f , M^2 and D denote the wavelength of the laser beam (794 nm), the focal length of the lens (30 cm), the degree of deviation from an ideal Gaussian beam (1.25), and the incident beam diameter, respectively. The incident beam diameter D was measured using the variable aperture method (Taira, 1998). The method involves measuring the laser power while adjusting the aperture diameter, to determine the diameter at which 86.5% of the total irradiance is reached. The incident beam diameter D was ~ 18 mm. The focused diameter calculated using equation (6) was ~ 21 μm . Therefore, the maximum range in the direction perpendicular to the center of laser beam in previous isotope imaging using LIMAS was limited up to 40 μm (Nagata et al., 2021; Bajo et al., 2024).

During isotope imaging, if the focal point of the laser beam is adjusted to consistently align with the irradiation position of the primary ion beam, the post-

ionization area can be expanded in the perpendicular direction to the focal point of the laser beam. To achieve this expansion in the post-ionization area for LIMAS, a large-area ion imaging system was developed. In this study, “large-area” is defined as the maximum area that can be analyzed using a scanning primary ion beam without moving the sample stage (i.e., the field of view of the mass spectrometer). This system comprises an objective lens (Sigma Koki), electronic micrometers (OptMike; Sigma Koki), a controller for the electronic micrometers (OMEC; Sigma Koki), and control software programmed using LabVIEW (National Instruments). Figure 1b illustrates a schematic view of the large-area imaging system. Electronic micrometers were installed on the three axes (X, Y, and Z) of the objective lens used to focus the laser beam. The sample plane defined as the XY plane. The micrometers were connected to a PXI-controller (NI PXIe-8881, National Instruments) via a controller through serial communication (RS-232C). By synchronizing the movement of Y-axis of objective lens with the scanning of the primary ion beam, the focal point of the laser beam was moved in the Y direction above the sample plane, ensuring alignment with the primary ion beam.

3.3. MATERIALS AND METHODS

3.3.1. Isotope Imaging of ^4He -implanted Si Wafer

To evaluate the performance of the large-area ion imaging system, isotope imaging was conducted using a ^4He -implanted Si wafer (hereafter He-Si). The total fluence of the ^4He was 1×10^{16} atoms cm^{-2} , and the implantation energy was 15 keV. A primary ion beam of $^{69}\text{Ga}^+$ (30 keV and 32 nA) was focused onto the sample surface and pulsed at 400 ns.

Ions with m/z 4 ($^4\text{He}^+$) and 7 ($^{28}\text{Si}^{4+}$) were measured using MULTUM II, with an acceleration voltage of -4 kV. The flight time was adjusted to achieve a mass resolving power of $\sim 10,000$ (calculated using $T/2\Delta T$) for m/z 4, corresponding to 52 laps to separate from interference ions for $^4\text{He}^+$. A typical TOF spectrum of MULTUM II from the He-Si is shown in Fig. 2. To obtain a 2D isotope image, the primary beam was scanned horizontally and vertically at $2.5\text{ }\mu\text{m}$ intervals. Each point in the image represents a pixel, and 40 TOF mass spectra were collected for each pixel. The Y-axis of objective lens was moved 60 times every $5\text{ }\mu\text{m}$ to adjust the position of laser beam. Ion images are $300 \times 300\text{ }\mu\text{m}^2$ consisting of 120×120 pixel.

3.3.2. Sample preparation and electron microscopy for NWA 801

The sample used in this study is the same polished section of the NWA 801 meteorite as that used in Chapter 2. The details of the sample preparation are described in Section 2.2.1.

Backscattered electron (BSE) images of the sample were obtained using FE-SEM (JSM-7000F, JEOL) to characterize the petrography prior to isotope-imaging analysis. Quantitative elemental analysis and X-ray elemental mapping were performed using the FE-SEM-EDS system. A 15 keV electron beam probe was used with a current of 4 nA .

3.3.3. Large-area Isotope Imaging of NWA 801

To investigate the SW- ^4He distribution in gas-rich meteorites and clarified the relationship between the SW- ^4He distribution and petrologic characteristics, large-area isotope imaging for NWA 801 was conducted using the developed system. The sample used in this study is the same as that in Section 2. A primary ion pulses at 200 ns

of $^{69}\text{Ga}^+$ (30 keV and ~ 32 nA) was focused onto the sample surface.

Ions with m/z 4 ($^4\text{He}^+$), 12 ($^{24}\text{Mg}^{2+}$), 16 ($^{32}\text{S}^{2+}$), and 28 ($^{56}\text{Fe}^{2+}$, $^{28}\text{Si}^+$) were measured using MULTUM II, with an acceleration voltage of -5 kV. The flight time was adjusted to achieve a mass resolving power of $\sim 20,000$ (calculated using $T/2\Delta T$) for m/z 4, corresponding to 151 laps. Interference ions, such as $^{12}\text{C}^{3+}$ and $^{16}\text{O}^{4+}$ for $^4\text{He}^+$, were removed using ion gates in the ion trajectory. A typical TOF spectrum from the sample is shown in Fig. 3. To obtain a 2D isotope image, the primary beam was scanned horizontally and vertically at $1\text{ }\mu\text{m}$ intervals. Each point in the image represents a pixel, and 200 TOF spectra were collected for each pixel. The Y-axis of objective lens was moved 150 times every $1\text{ }\mu\text{m}$ to adjust the position of laser beam. Ion images are $100 \times 150\text{ }\mu\text{m}^2$ consisting of 100×150 pixel.

The blank ^4He as the residual ^4He gas above the sample surface was measured without the primary beam pulse after each isotope imaging. The $(3-7.5) \times 10^5$ TOF spectra were collected to determine the blank ^4He intensity. The blank ^4He was $0.3-7 \times 10^{-4}$ cpms.

3.3.4. Spatial Resolution of Isotope Imaging Analysis

The spatial resolution for ion images was determined using line scan profiles of $^{56}\text{Fe}^{2+}$ and $^{24}\text{Mg}^{2+}$ (Figs. S7–S21) across the mineral–matrix interface, defined by the 16–84% criterion (Nagata et al., 2019). This is equivalent to the $\pm 1\sigma$ criterion for a Gaussian beam. The spatial resolutions for the analyses are listed in Table 1. The spatial resolution of the ion image in this study is defined as the average width of the $^{56}\text{Fe}^{2+}$ and $^{24}\text{Mg}^{2+}$, determined to be $2.4\text{ }\mu\text{m}$.

3.3.5. Identification of ^4He -bearing Individual Mineral Grain

In this study, an individual mineral grain is defined as a single mineral phase, larger than 1 μm in size, and embedded in the matrix in isolation. I identified individual mineral grain on the BSE images. ^4He -bearing individual mineral grain was identified by position matching of BSE image and ion images of $^4\text{He}^+$, $^{24}\text{Mg}^{2+}$, $^{32}\text{S}^{2+}$, and $^{56}\text{Fe}^{2+}$. The criterion for determining that a ^4He -bearing individual mineral grain contains $^4\text{He}^+$ signal from a pixel overlapping the location of the grain identified in the BSE image, with at least two counts per pixel because one count is indistinguishable from ^4He blank.

3.3.6. Quantification of ^4He Concentration

To determine the ^4He concentration of each ^4He -bearing individual mineral grain, the average $^4\text{He}^+$ intensity was calculated by accumulating the pixels with two or more counts that overlapped with the position of the individual mineral grains identified in the BSE image. Based on equation (1) in Section 2.2.4, the ^4He concentration was determined by multiplying the average $^4\text{He}^+$ intensity and SF^{He} . Since the sputtering rate differs for each mineral phase, the SF^{He} differs for each mineral phase. In this study, the SF^{He} for opaque minerals (Fe-Ni metal, ferrihydrite, magnetite, troilite) was determined using ^4He -implanted Fe-Ni metal as a standard. For silicate phases, the SF^{He} determined from ^4He -implanted olivine (Bajo et al., 2024) was adopted. The total fluence of the ^4He was $3 \times 10^{15} \text{ atoms cm}^{-2}$, and the implantation energy was 20 keV.

3.4. RESULTS

3.4.1. Evaluation of the Large-area Ion Imaging System

Figure 5 shows $^4\text{He}^+$ and $^{28}\text{Si}^{4+}$ ion images obtained in the experiment. The horizontal and vertical axes correspond to the X and Y directions of the sample and lens,

respectively. The ion intensities were normalized to the centroid intensity of 100% and smoothed by a simple moving average of nine pixels. Fig. 4a and 4b show $^4\text{He}^+$ and $^{28}\text{Si}^{4+}$ ion images using the conventional imaging system. In both the $^4\text{He}^+$ and $^{28}\text{Si}^{4+}$ ion images, the intensity decreases more steeply in the direction perpendicular to the center of laser beam (Y direction) than in the direction parallel to the center of laser beam (X direction). This observation indicates that the maximum range of ion imaging was limited by the focused diameter of laser beam. Fig. 4c and 4d show $^4\text{He}^+$ and $^{28}\text{Si}^{4+}$ ion images after the implementation of the large-area ion imaging system. In both the $^4\text{He}^+$ and $^{28}\text{Si}^{4+}$ ion images, the intensity decreases more gradually in the Y direction than the ion images without the large-area ion imaging system.

To investigate the $^4\text{He}^+$ and $^{28}\text{Si}^{4+}$ intensity distribution in the Y direction, I obtained line profiles of the row crossing the centroid of each image (Fig. 5). In $^4\text{He}^+$ and $^{28}\text{Si}^{4+}$ ion images without the large-area ion imaging system, the ranges above 90% of the centroid intensity were 40 μm and 45 μm , respectively (Fig. 5a and 5b). In $^4\text{He}^+$ and $^{28}\text{Si}^{4+}$ ion images after the implementation of large-area ion imaging system, the ranges above 90% of the centroid intensity were 150 μm and 190 μm , respectively (Fig. 5c and 5d). The results of experiments using a He-Si have confirmed that LIMAS can image up to 150 μm in the Y direction by implementing the large-area ion imaging system.

3.4.2. Petrography of measurement areas

BSE and X-ray elemental images of the polished section of NWA 801 are shown in Fig. 6. SEM analysis reveals that NWA 801 contains millimeter-sized chondrules, fragments of chondrules, and an interstitial matrix. Magnified BSE and X-ray elemental images (Fig. 6c, 6d, 6e, 6f) shows that the interstitial matrix contains individual mineral grains of Fe-Ni metal, ferrihydrite, magnetite, olivine, pyroxene, and

troilite, ranging from approximately 1 to 50 μm . These individual mineral grains are embedded in a fine-grained groundmass typically less than 0.1 μm in size. In this study, I examined the interstitial matrix in Areas 1–15 (Fig. 6).

3.4.3. Isotope Maps

Enlarged BSE and $^4\text{He}^+$, $^{56}\text{Fe}^{2+}$, $^{24}\text{Mg}^{2+}$, $^{32}\text{S}^{2+}$, $^{28}\text{Si}^+$ images of Areas 1–15 are shown in Figs. 7–21. The images show ^4He distribution in the interstitial matrix between Chondrule 1 and Chondrule 2 (Areas 1–7, Figs. 7–13), around Chondrule 3 (Areas 8–11, Figs. 14–17), and in Areas containing individual Fe-Ni metal grains (Areas 12–15, Figs. 18–21). I identified 1,595 individual mineral grains in these fifteen areas.

3.4.4. ^4He Distribution in the Interstitial Matrix

The Interstitial Matrix between Chondrule 1 and Chondrule 2

I obtained seven isotope maps (Area 1–7, Figs. 7–13) from the interstitial matrix between Chondrule 1 and Chondrule 2.

Area 1 contains the matrix and rim of Chondrule 1 (Fig. 7). The rim of Chondrule 1 is located on the right side of this area. $^4\text{He}^+$ signals were not detected in the rim, whereas $^4\text{He}^+$ signals above two counts were detected from individual mineral grains in the matrix. The highest ^4He in Area 1 was observed from an individual troilite grain. ^4He -bearing grains were predominantly distributed in the coarse grain-rich matrix on the upper portion of Area 1. ^4He signals were detected along the edge of olivine grains approximately 100 μm in size, indicative of SW-implanted ^4He , consistent with the distribution observed in Chapter 2. Additionally, $^4\text{He}^+$ signals were identified within the fine-grained groundmass of the coarse grain-rich matrix, as corroborated by the high-resolution ^4He imaging presented in Chapter 2. In the lower right of Area 1, there is also

a coarse-grain-rich matrix, but no grains containing ^4He . Furthermore, a fine grain-rich matrix composed of mainly fine-grained groundmass adjacent to Chondrule 1 in the lower left of Area 1, where $^4\text{He}^+$ signals were not detected.

Area 2 contains the matrix and rim of Chondrule 1 (Fig. 8). $^4\text{He}^+$ signals were not detected in the rim, whereas $^4\text{He}^+$ signals above two counts were detected from individual mineral grains in the matrix. The highest ^4He in Area 2 was observed from an individual troilite grain. ^4He -bearing grains were distributed in the coarse-grain-rich matrix. $^4\text{He}^+$ signals were identified within the fine-grained groundmass of the coarse grain-rich matrix, as corroborated by the high-resolution ^4He imaging presented in Chapter 2.

Area 3 contains the matrix and rim of Chondrule 1 (Fig. 9). $^4\text{He}^+$ signals were not detected in the rim, whereas $^4\text{He}^+$ signals above two counts were detected from individual mineral grains in the matrix. The highest ^4He in Area 3 was observed from an individual troilite grain. ^4He -bearing grains were predominantly distributed in the coarse grain-rich matrix. $^4\text{He}^+$ signals were detected along the edges of troilite grains ($\sim 10\text{ }\mu\text{m}$, $15\text{ }\mu\text{m}$, $25\text{ }\mu\text{m}$), indicative of SW-implanted ^4He . To the left of the coarse grain-rich matrix, and the portion between Chondrule 1 and the coarse grain-rich matrix, a fine grain-rich matrix containing a small number of $1\text{--}5\text{ }\mu\text{m}$ grains and fine-grained groundmass, where $^4\text{He}^+$ signals were not detected.

Area 4 contains the matrix, rim of Chondrule 1, and chondrule fragments (Fig. 10). $^4\text{He}^+$ signals were not detected in the rim and chondrule fragments, whereas $^4\text{He}^+$ signals above two counts were detected from individual mineral grains in the matrix. The highest ^4He in Area 4 was observed from an individual troilite grain. ^4He -bearing grains were predominantly distributed in the coarse grain-rich matrix. $^4\text{He}^+$ signals were

detected along the edge of Fe-Ni metal grain approximately 10 μm in size, indicative of SW-implanted ^4He . Between Chondrule 1 and the coarse grain-rich matrix, a fine grain-rich matrix containing a small number of 1–5 μm grains and fine-grained groundmass, where $^4\text{He}^+$ signals were not detected.

Area 5 contains the matrix and chondrule fragments (Fig. 11). $^4\text{He}^+$ signals were not detected in chondrule fragments, whereas $^4\text{He}^+$ signals above two counts were detected from individual mineral grains in the matrix. The highest ^4He in Area 5 was observed from an individual ferrihydrite grain. ^4He -bearing grains were predominantly distributed in the coarse grain-rich matrix. $^4\text{He}^+$ signals were detected along the edge of olivine grain approximately 15 μm in size, indicative of SW-implanted ^4He . $^4\text{He}^+$ signals were not detected in a fine grain-rich matrix containing a small number of 1–5 μm grains and fine-grained groundmass.

Area 6 contains the matrix and chondrule fragments (Fig. 12). $^4\text{He}^+$ signals were not detected in chondrule fragments, whereas ^4He above two counts were detected from individual mineral grains in the matrix. The highest $^4\text{He}^+$ signals in Area 6 was observed from an individual troilite grain. ^4He -bearing grains were distributed around relatively large (~25–30 μm) olivine and pyroxene. $^4\text{He}^+$ signals were not detected in a fine grain-rich matrix which contains only approximately 1 μm grains and fine-grained groundmass.

Area 7 contains the matrix, rim of Chondrule 2, and chondrule fragments (Fig. 13). $^4\text{He}^+$ signals were not detected in rim of Chondrule 2 and chondrule fragments, whereas $^4\text{He}^+$ signals above two counts were detected from individual mineral grains in the matrix. The highest ^4He in Area 7 was observed from an individual troilite grain. ^4He -bearing grains were distributed in the coarse grain-rich matrix in the upper portion of

Area 7. Additionally, $^4\text{He}^+$ signals were identified within the fine-grained groundmass of the coarse grain-rich matrix. In the right side of Area 7, a fine grain-rich matrix containing only 1–5 μm grains and fine-grained groundmass, where $^4\text{He}^+$ signals were not detected.

The Interstitial Matrix around Chondrule 3

I obtained four isotope maps (Areas 8–11, Figs. 14–17) from the interstitial matrix around Chondrule 3.

Area 8 contains the matrix and Chondrule 3 (Fig. 14). $^4\text{He}^+$ signals were not detected in Chondrule 3, whereas $^4\text{He}^+$ signals above two counts were detected from individual mineral grains in the matrix. The highest ^4He in Area 8 was observed from an individual troilite grain. ^4He -bearing grains were predominantly distributed in the coarse grain-rich matrix. Additionally, $^4\text{He}^+$ signals were identified within the fine-grained groundmass of the coarse grain-rich matrix. In the upper left of Area 8, a fine grain-rich matrix containing only 1–5 μm grains and fine-grained groundmass, where $^4\text{He}^+$ signals were not detected.

Area 9 contains the matrix, compound of olivine and various size troilites, and Chondrule 3 (Fig. 15). $^4\text{He}^+$ signals were not detected in the interior of Chondrule 3 and the compound, whereas $^4\text{He}^+$ signals above two counts were detected from the edge of Chondrule 3 and individual mineral grains in the matrix. The highest ^4He in Area 9 was observed from an individual troilite grain. ^4He -bearing grains were predominantly distributed in the coarse grain-rich matrix. Additionally, $^4\text{He}^+$ signals were identified within the fine-grained groundmass of the coarse grain-rich matrix.

Area 10 contains the matrix and Chondrule 3 (Fig. 16). $^4\text{He}^+$ signals were not detected in the interior of Chondrule 3, whereas $^4\text{He}^+$ signals above two counts were detected from the edge of Chondrule 3 and individual mineral grains in the matrix. The

highest ^4He in Area 10 was observed from an individual ferrihydrite grain. ^4He -bearing grains were distributed throughout the entire matrix of Area 10, which is scattered with various-sized coarse grains. ^4He signals were detected along the edge of ferrihydrite grain approximately 15 μm in size, indicative of SW-implanted ^4He . Additionally, $^4\text{He}^+$ signals were identified within the fine-grained groundmass of the coarse grain-rich matrix. Furthermore, the matrix contains 30–40 μm lumps composed of a small number of 1–5 μm grains and fine-grained groundmass, where $^4\text{He}^+$ signals were not detected.

Area 11 contains the matrix and Chondrule 3 (Fig. 17). $^4\text{He}^+$ signals were not detected in Chondrule 3, whereas $^4\text{He}^+$ signals above two counts were detected from the edge of Chondrule 3 and individual mineral grains in the matrix. The highest ^4He in Area 11 was observed from an individual troilite grain. ^4He -bearing grains were distributed throughout the entire matrix of Area 11, which is scattered with various-sized coarse grains. $^4\text{He}^+$ signals were detected along the edge of ferrihydrite grain approximately 8 μm in size, indicative of SW-implanted ^4He . Additionally, $^4\text{He}^+$ signals were identified within the fine-grained groundmass of the coarse grain-rich matrix. Furthermore, the matrix contains ~40 μm lumps composed of a small number of 1–5 μm grains and fine-grained groundmass, where $^4\text{He}^+$ signals were not detected.

The Interstitial Matrix containing Individual Fe-Ni metal Grains

I obtained seven isotope maps (Area 12–15, Figs. 18–21) from the interstitial matrix containing individual Fe-Ni metal grains. These areas are not adjacent to each other, unlike Areas 1–7 and 8–11.

Area 12 contains the matrix and olivine + Fe-Ni metal + ferrihydrite compounds thought to be fragments of chondrule (Fig. 18). $^4\text{He}^+$ signals were not detected in compounds, whereas $^4\text{He}^+$ signals above two counts were detected from individual

mineral grains in the matrix. The highest ^4He in Area 12 was observed from an individual troilite grain. ^4He -bearing grains were distributed throughout the center of matrix of Area 12, which is scattered with various-sized coarse grains. $^4\text{He}^+$ signals were detected along the edge of Fe-Ni metal grain approximately 7 μm in size, indicative of SW-implanted ^4He . Additionally, $^4\text{He}^+$ signals were identified within the fine-grained groundmass of the coarse grain-rich matrix. In the upper left of Area 12, a fine grain-rich matrix containing only 1–5 μm grains and fine-grained groundmass, where $^4\text{He}^+$ signals were not detected.

Area 13 contains the matrix and part of a chondrule (Fig. 19). $^4\text{He}^+$ signals were not detected in the chondrule, whereas $^4\text{He}^+$ signals above two counts were detected from individual mineral grains in the matrix. The highest ^4He in Area 13 was observed from an individual troilite grain. ^4He -bearing grains were distributed throughout the matrix of Area 13, which is scattered with various-sized coarse grains. Additionally, $^4\text{He}^+$ signals were identified within the fine-grained groundmass of the coarse grain-rich matrix. Furthermore, a fine grain-rich matrix containing only 1–5 μm grains and fine-grained groundmass, where $^4\text{He}^+$ signals were not detected.

Area 14 contains the matrix and part of a chondrule (Fig. 20). $^4\text{He}^+$ signals were not detected in the chondrule, whereas $^4\text{He}^+$ signals above two counts were detected from individual mineral grains in the matrix. The highest ^4He in Area 14 was observed from an individual ferrihydrite grain. ^4He -bearing grains were distributed throughout the matrix of Area 14, which is scattered with various-sized coarse grains. ^4He signals were detected along the edge of Fe-Ni metal ($\sim 25 \mu\text{m}$) and olivine grain ($> 30 \mu\text{m}$), indicative of SW-implanted ^4He . Additionally, $^4\text{He}^+$ signals were identified within the fine-grained groundmass of the coarse grain-rich matrix. Furthermore, a fine grain-rich matrix adjacent to the $>30 \mu\text{m}$ olivine grain containing only 1–5 μm grains and fine-grained

groundmass, where $^4\text{He}^+$ signals were not detected.

Area 15 contains the matrix and part of a chondrule (Fig. 21). $^4\text{He}^+$ signals were not detected in the chondrule, whereas $^4\text{He}^+$ signals above two counts were detected from individual mineral grains in the matrix. The highest ^4He in Area 15 was observed from an individual troilite grain. ^4He -bearing grains were distributed throughout the matrix of Area 15, which is scattered with various-sized coarse grains. $^4\text{He}^+$ signals were detected along the edge of Fe-Ni metal ($\sim 20\ \mu\text{m}$), indicative of SW-implanted ^4He . Additionally, a fine grain-rich matrix containing only 1–5 μm grains and fine-grained groundmass, where $^4\text{He}^+$ signals were not detected.

3.4.5. Grain Size Distribution of Individual Mineral Grains

The proportion of individual mineral grains of each grain size was 83.8% for 1–5 μm ($n = 1,337$), 9.7% for 5–10 μm ($n = 154$), 3.1% for 10–15 μm ($n = 49$), 1.7% for 15–20 μm ($n = 28$), 0.9% for 20–25 μm ($n = 15$), 0.4% for 25–30 μm ($n = 6$), and 0.4% for grains larger than 30 μm in size ($n = 6$). Smaller individual mineral grains (1–5 μm) are more abundant, and the size distribution appears to follow a power law (Fig. 22). The proportion of individual mineral grains of each mineral phase was 38.7% for troilite ($n = 618$), 28.1% for ferrihydrite ($n = 448$), 3.5% for Fe-Ni metal ($n = 56$), 0.3% for magnetite ($n = 4$), and 29.4% for silicate ($n = 469$). Grain size distribution of each mineral phase is summarized in Table 2. The grain size distributions differ while 94% of opaque minerals fall within the 1–5 μm range (Fig. 23a), silicate grains exhibit a continuous distribution ranging from 1–5 μm to over 30 μm (Fig. 23b).

3.4.6. SW- ^4He Concentration of Mineral Grains and Fine Grain-rich Matrix

The number of ^4He -bearing grains ($n = 330$) of each grain size is summarized

in Table 3. The proportion of ^4He -bearing grains of each grain size was 20% for 1–5 μm ($n = 268$), 20% for 5–10 μm ($n = 30$), 20% for 10–15 μm ($n = 10$), 32% for 15–20 μm ($n = 9$), 47% for 20–25 μm ($n = 7$), 50% for 25–30 μm ($n = 3$), and 50% for grains larger than 30 μm ($n = 3$). The proportion of ^4He -bearing grains increase with increasing grain size (Fig. 24).

The number of ^4He -bearing grains ($n = 330$) of each mineral phase is summarized in Table 4. The proportion of ^4He -bearing grains of each mineral phase was 27% for troilite ($n = 168$), 13% for ferrihydrite ($n = 55$), 19% for Fe-Ni metal ($n = 11$), 75% for magnetite ($n = 3$), and 20% for silicate ($n = 93$). The proportion of SW- ^4He -bearing grains of the opaque mineral (troilite + ferrihydrite + Fe-Ni metal + magnetite) was 21%, which was consistent with that of the silicate (Fig. 25).

The $^4\text{He}^+$ intensities and ^4He concentrations of individual mineral are summarized in Table S1. The range of $^4\text{He}^+$ intensity of each mineral phase was 2–142 counts pixel^{-1} for troilite, 2–91 counts pixel^{-1} for ferrihydrite, 3–17 counts pixel^{-1} for Fe-Ni metal, 9–28 counts pixel^{-1} for magnetite, 2–42 counts pixel^{-1} for silicate. Fig. 26 shows the histogram of $^4\text{He}^+$ ion intensity of opaque mineral and silicate. The histogram of $^4\text{He}^+$ intensity of the opaque mineral shows the continuous distribution ranging from 2–10 counts pixel^{-1} to 50–60 counts pixel^{-1} (Fig. 26a), whereas 89% of silicate fall within the 2–10 counts pixel^{-1} (Fig. 26b).

For opaque minerals in $^4\text{He}^+$ ion images 1 count pixel^{-1} is estimated to be 2.3×10^{19} atoms cm^{-3} using ^4He -implanted Fe-Ni metal standard. For silicates in $^4\text{He}^+$ ion images, 1 count pixel^{-1} is estimated to be 5.2×10^{19} atoms cm^{-3} using ^4He -implanted olivine standard. Therefore, the ^4He concentrations of each mineral phase was 0.5–32.5 $\times 10^{20}$ atoms cm^{-3} for troilite, 0.5–20.8 $\times 10^{20}$ atoms cm^{-3} for ferrihydrite, 0.6–3.9 $\times 10^{20}$

atoms cm^{-3} for Fe-Ni metal, $2.1\text{--}6.5 \times 10^{20}$ atoms cm^{-3} for magnetite, $0.9\text{--}21.4 \times 10^{20}$ atoms cm^{-3} for silicate. Fig. 27 shows the histogram of ^4He concentrations of opaque mineral and silicate. The histogram of ^4He concentrations of opaque mineral shows the continuous distribution ranging from 0.5×10^{20} atoms cm^{-3} to 32.5×10^{20} atoms cm^{-3} (Fig. 27a), whereas 77% of silicate fall within the $1\text{--}3 \times 10^{20}$ atoms cm^{-3} (Fig. 27b).

The average $^4\text{He}^+$ intensity of the fine grain-rich matrix was 0.04 counts pixel^{-1} corresponding to a ^4He concentration of 2.1×10^{18} atoms cm^{-3} . This concentration is comparable to the blank ^4He , ranging from 3.1×10^{17} to 1.3×10^{18} atoms cm^{-3} .

3.5. DISCUSSION

3.5.1. Formation Process of the Heterogeneous Distribution of SW- ^4He

The distribution of ^4He in NWA 801 exhibits three characteristics: 1) ^4He is mainly contained within individual mineral grains. 2) ^4He -bearing mineral grains are predominantly distributed within the coarse grain-rich matrix (e.g., Fig. 11). 3) The fine grain-rich matrix, composed of $1\text{--}5 \mu\text{m}$ grains and fine-grained groundmass, occurs as accessory rims around chondrules (e.g., Fig. 10) and an isolated olivine grain (Fig. 20) and also appears as scattered lumps ranging from several tens to hundreds of micrometers in size (e.g., Figs. 11 and 16). No detectable ^4He was observed in the fine grain-rich matrix.

In various chondrite groups, the fine-grained material adjacent to the chondrules is called fine-grained rims (FGRs), which are distinguished from the surrounding matrix (intercomponent matrix; ICM) by their texture (grain size, compaction, and porosity) (e.g., MacPherson et al., 1985; Zanetta et al., 2021). In the case of CR chondrites, approximately 12% of chondrules are entirely surrounded by FGRs

(Pinto et al., 2022). FGRs are also observed surrounding isolated olivine grains (Hanna and Ketcham, 2018) and scattering as fragments within the ICM (Pinto et al., 2022). Based on the similarities in occurrence, the fine grain-rich matrix observed in this study is thought to be the same material as FGRs. The origin of FGRs remains poorly constrained. One hypothesis is that FGRs formed in a nebular environment through the accretion of dust around free-floating chondrules before their incorporation into planetesimals (e.g., Metzler et al., 1992). Alternatively, FGRs may have formed during parent body evolution through secondary processes such as fluid and/or thermal alteration (e.g., Tomeoka and Ohnishi, 2010) or compaction prior to aqueous alteration (Trigo-Rodriguez et al., 2006). Although no consensus has been reached regarding the origin of FGRs, all hypotheses agree that their formation predates the onset of aqueous alteration on the parent body. The age of aqueous alteration events in CR chondrites has been estimated at 4–5 Myr after CAI formation (Jilly-Rehak et al., 2017), which is older than the epoch of disk gas dispersal (<10 Myr after CAI formation; Pascucci and Tachibana, 2010). Thus, it is inferred that the fine grain-rich matrix existed in a consolidated material prior to the SW irradiation on the NWA 801 parent body. No detectable $^4\text{He}^+$ signal in the fine grain-rich matrix indicates not merely that the ^4He concentration is below the detection limit ($\sim 10^{18}$ atoms cm^{-3}), but rather that SW- ^4He was not implanted into its interior, likely due to solidification prior to the regolith processes.

Based on the above discussion, I estimated the formation process of the heterogeneous distribution of SW- ^4He in NWA 801. Fig. 28 shows a schematic cartoon of the formation process. Firstly, fragmentation of CR chondrules is thought to have occurred in the nebular (Schrader et al., 2008), suggesting that a certain proportion of individual mineral grains were already present when the constituent materials accreted

onto the parent body, where aqueous alteration later took place. Accordingly, it is reasonable to assume that the parent body was already composed of a heterogeneous mixture of chondrules with FGRs, coarse mineral grains, and FGR fragments by the time nebular gas dissipation occurred and SW irradiation began (Fig. 28a).

The partial preservation of FGRs, which possess the lowest mechanical strength among all meteorite components, along with the intact state of many chondrules, suggests that the impacts associated with regolith gardening primarily caused the disaggregation of loosely bound components, without sufficient energy to fracture chondrules (Fig. 28b). SW- ^4He accumulates on the surface of unconsolidated material (Fig. 28c). Repeated low-energy impacts form an unconsolidated layer containing SW- ^4He (Fig. 28d). Once the surface layer has been excavated to a certain depth, further excavation at the same scale impacts is no longer possible (Melosh, 2011). As a result, subsequent impacts merely stir the unconsolidated layer (Fig. 28e). This stirring exposes material not previously exposed to the SW, leading to the accumulation of new SW- ^4He and promoting the formation of a SW- ^4He -enriched unconsolidated layer at the surface of parent body over time (Fig. 28f, g).

The accumulation of SW- ^4He in unconsolidated layers is terminated by a higher-energy impact capable of excavating material to a depth of several millimeters underground (Fig. 28h). The typical period during which the unconsolidated layer is buried by the higher-energy impact is considered to be approximately 10,000 years, corresponding to the SW irradiation duration for coarse mineral grains (Wada et al., 2025). Over the several tens of millions of years that the NWA 801 meteorite resided at depths of several tens of centimeters to ~1 meter below the surface (Nakashima et al., 2009), the processes described in steps Fig. 28b to Fig. 28i repeated, ultimately leading to the

heterogeneous distribution of SW- ^4He observed in NWA 801. The heterogeneous distribution of SW- ^4He in the interstitial matrix is originated from the low-energy impacts that induce the semi-conservative fragmentation of FGRs, which were the main process of regolith gardening on the parent body of NWA 801.

3.5.2. Concentrations and Proportions of SW- ^4He -bearing Mineral Grains

The ^4He concentrations of opaque minerals (troilite, ferrihydrite, Fe-Ni metal, and magnetite) and silicate have almost the same range but different frequency distributions (Fig. 27). The number of grains with higher ^4He concentrations is significantly higher than silicate for opaque minerals. The strong mineral dependence of ^4He retentivities has been previously reported (Baur et al., 1972; Jull and Pillinger, 1977). Silicates have lower ^4He retentivity compared to Fe-Ni metals and troilite. Consequently, SW- ^4He in silicates may have been lost through degassing, leading to lower ^4He signals in silicates compared to opaque minerals.

On the other hand, the proportions of ^4He -bearing grains in opaque minerals and silicates are nearly identical (~20%; Fig. 25). This similarity suggests that SW irradiation occurred irrespective of mineral type, affecting both opaque minerals and silicates. Assuming that the regolith preserved in the meteorite reflects the global surface characteristics of the NWA 801 parent body, the proportion of ^4He -bearing grains implies that approximately 20% of the unconsolidated surface layer was exposed to SW for a sufficiently long duration to accumulate detectable SW- ^4He , while the remaining 80% consisted of “immature regolith” that experienced minimal SW exposure (likely less than a few hundred years). The presence of immature regolith as the dominant surface component is consistent with asteroid surfaces today (Denevi et al., 2016), suggesting

that the NWA 801 parent body may have resided in a similar environment to present-day asteroids (Pieters et al., 2012).

3.6. CONCLUSIONS

I presented a new technique that enables the observation of the distribution of past SW-bearing minerals in meteorites. By implementing a “large-area ion imaging system” into LIMAS, in which the focal point of the laser beam is adjusted to consistently align with the irradiation position of the primary ion beam to expand the post-ionization area, I achieved ^4He imaging over a $100 \times 150 \mu\text{m}$ area. Applying this improved *in situ* ^4He imaging technique to the NWA 801 CR2 chondrite allowed us to elucidate the relationship between the heterogeneity of SW- ^4He and petrological characteristics. The distribution of SW- ^4He in NWA 801 exhibits the following characteristics: 1) ^4He is mainly contained within individual mineral grains. 2) ^4He -bearing mineral grains are predominantly distributed within the coarse grain-rich matrix. 3) The fine grain-rich matrix, composed of 1–5 μm grains and fine-grained groundmass, occurs as accessory rims around chondrules and an isolated olivine grain and also appears as scattered lumps ranging from several tens to hundreds of micrometers in size. No detectable ^4He was observed in the fine grain-rich matrix. Based on the similarities in occurrence, the fine grain-rich matrix observed in this study is thought to be the same material as FGRs. The partial preservation of FGRs along with the intact state of many chondrules, suggests that the daily impacts associated with gardening process primarily caused the disaggregation of loosely bound components, without sufficient energy to fracture chondrules. The heterogeneous distribution of SW- ^4He in the interstitial matrix is originated from the low-energy impacts that induce the semi-conservative fragmentation of FGRs, which were the

main process of regolith gardening on the parent body of NWA 801. Furthermore, the proportions of ^4He -bearing grains in opaque minerals and silicates are nearly identical ($\sim 20\%$), suggesting that SW irradiation occurred irrespective of mineral type, affecting both opaque minerals and silicates. If the regolith preserved in the meteorite reflects the global surface characteristics of the NWA 801 parent body, the proportion of ^4He -bearing grains implies that approximately 20% of the unconsolidated surface layer was exposed to SW for a sufficiently long duration to accumulate detectable SW- ^4He , while the remaining 80% consisted of “immature regolith” that experienced minimal SW exposure (likely less than a few hundred years). The surface of the parent body of NWA 801 was likely composed largely of immature regolith, similar to present-day asteroids.

FIGURES

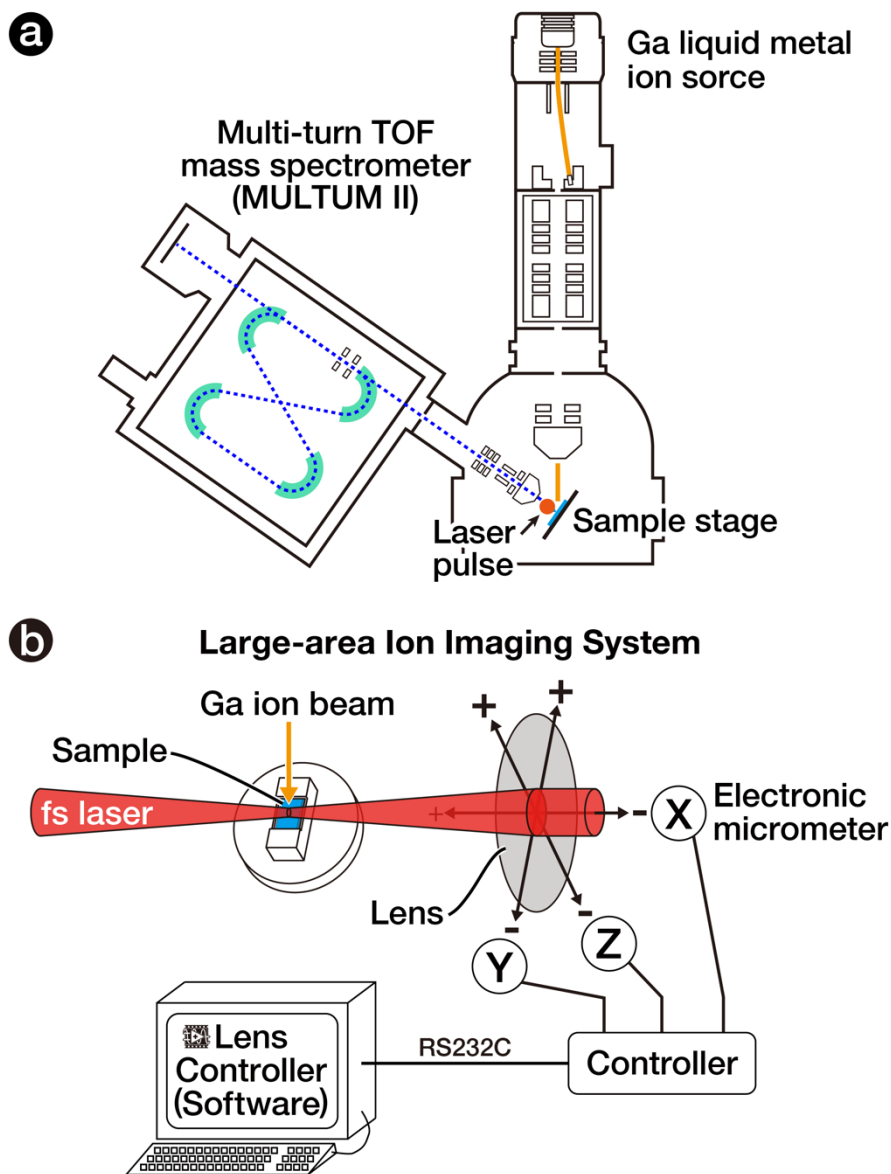


Figure 1. (a) Schematic view of LIMAS. Orange lines show primary ion beam. Dotted blue line shows the flight path of ions. Red circle shows the irradiated laser pulse. (b) Schematic view of the large-area ion imaging system.

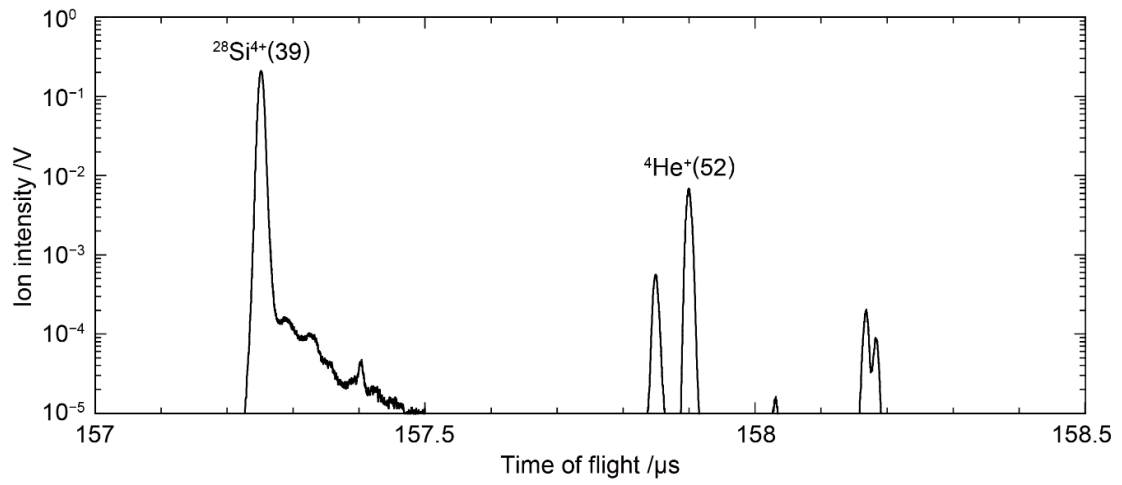


Figure 2. TOF spectrum of the He-Si shown in Fig. 5. The arrows indicate the measured ion species, where the numbers in parentheses indicate the numbers of laps for each ion in MULTUM II.

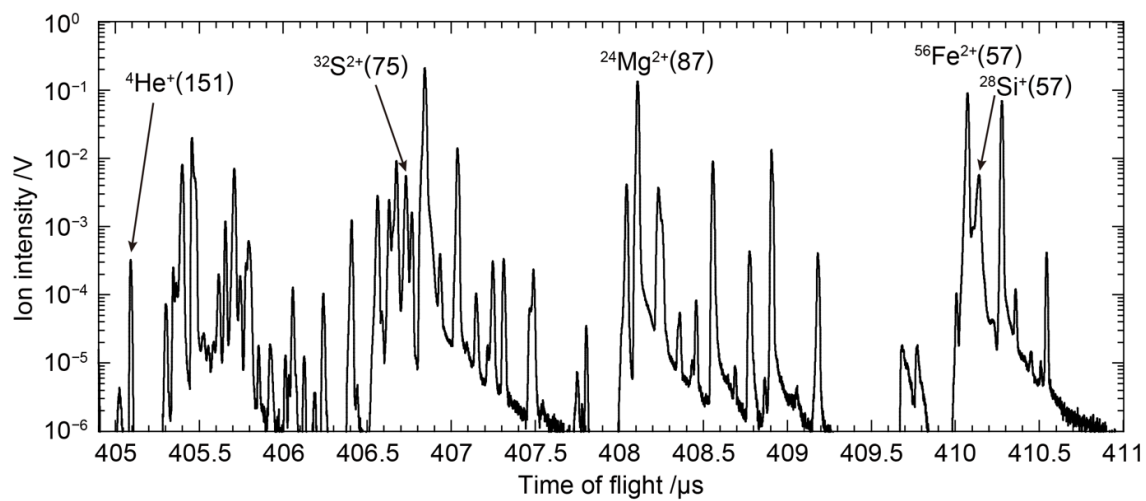


Figure 3. TOF spectrum of Area 10 of the NWA 801 sample shown in Fig. 17. The arrows indicate the measured ion species, where the numbers in parentheses indicate the numbers of laps for each ion in MULTUM II.

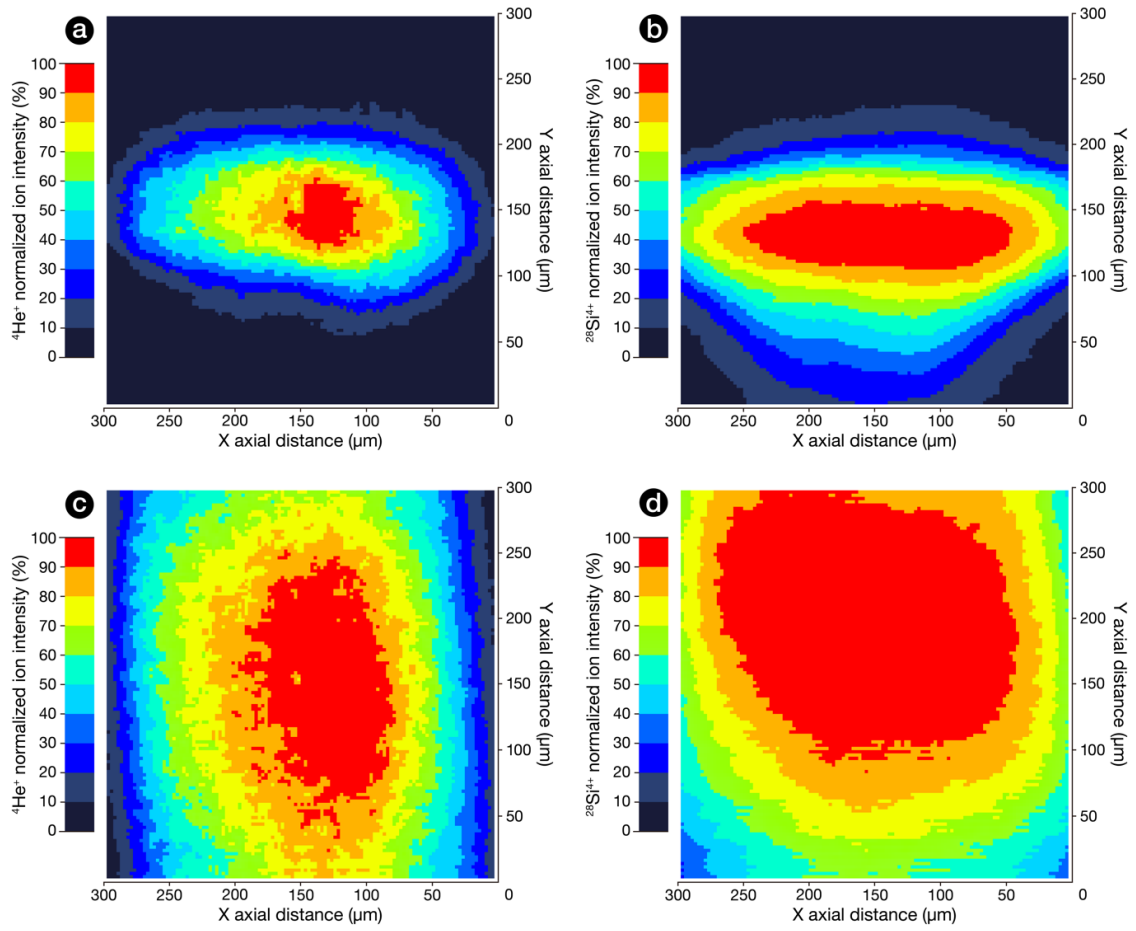


Figure 4. (a) $^4\text{He}^+$ ion image of He-Si wafer without the large-area ion imaging system. (b) $^{28}\text{Si}^{4+}$ ion image of He-Si wafer without the large-area ion imaging system. (c) $^4\text{He}^+$ ion image of He-Si wafer with the large-area ion imaging system. (d) $^{28}\text{Si}^{4+}$ ion image of He-Si wafer with the large-area ion imaging system.

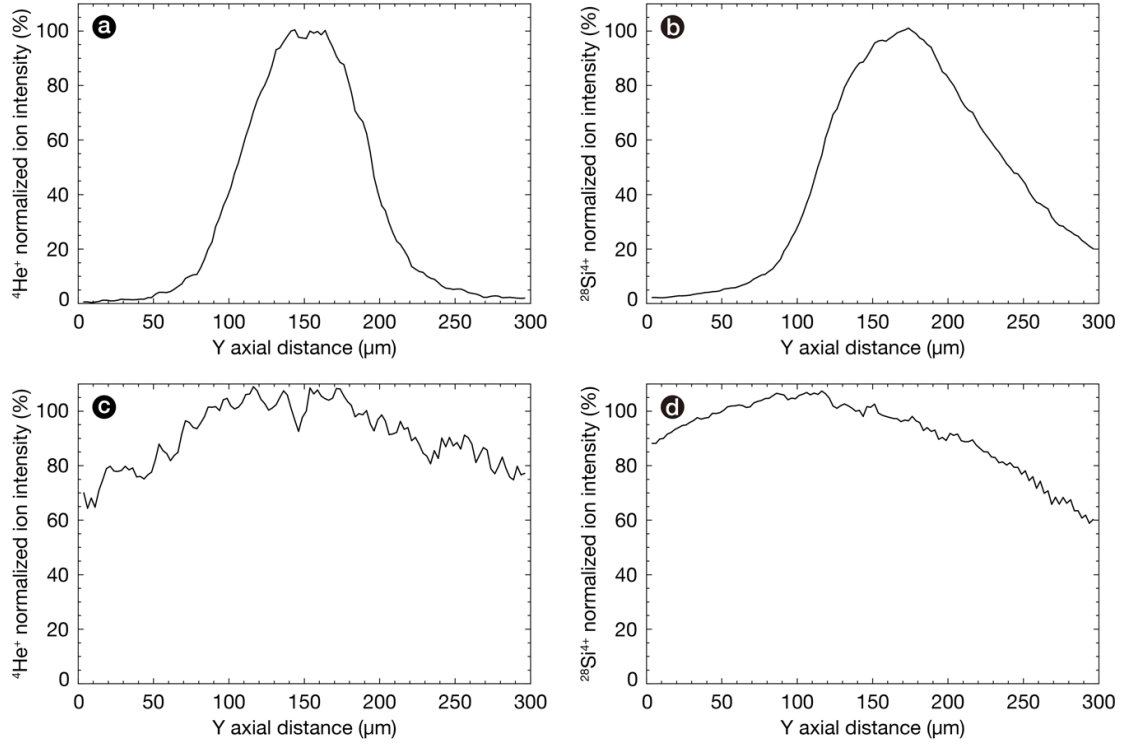


Figure 5. (a) $^4\text{He}^+$ ion intensity line profile in the Y direction without the large-area ion imaging system. The ion intensities were normalized to the centroid intensity of 100% and smoothed by a simple moving average of nine pixels. (b) $^{28}\text{Si}^{4+}$ ion intensity line profile in the Y direction without the large-area ion imaging system. (c) $^4\text{He}^+$ ion intensity line profile in the Y direction after the implementation of large-area ion imaging system. (d) $^{28}\text{Si}^{4+}$ ion intensity line profile in the Y direction after the implementation of large-area ion imaging system.

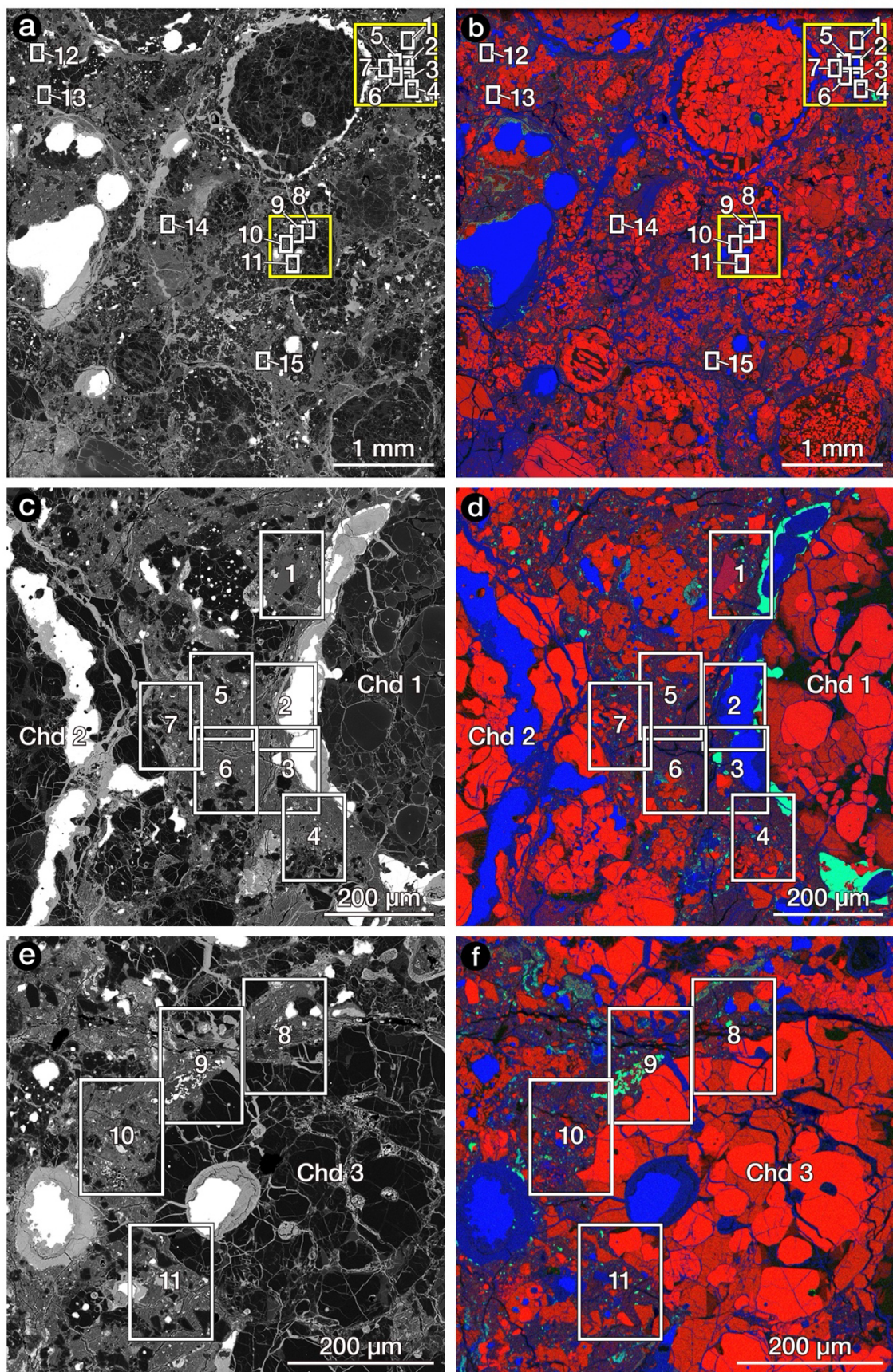


Figure 6. (a) BSE image of NWA 801 meteorite. (b) Composite X-ray elemental map of the same area shown in (a), with Mg (red), Si (green), and Fe (blue). White boxes numbered 1 to 15 correspond to the areas 1 to 15 shown in Figs. 9 to 23. (c) Magnified BSE image shown in the upper right yellow box in (a). White boxes numbered 1 to 7 correspond to the areas 1 to 7 shown in Figs. 9 to 15. (d) Magnified composite X-ray elemental map of the same area shown in (c). White boxes numbered 1 to 7 correspond to the areas 1 to 7 shown in Figs. 9 to 15. (e) Magnified BSE image shown in the center yellow box in (a). White boxes numbered 8 to 11 correspond to the areas 8 to 11 shown in Figs. 16 to 19. (d) Magnified composite X-ray elemental map of the same area shown in (e). White boxes numbered 1 to 7 correspond to the areas 8 to 11 correspond to the areas 8 to 11 shown in Figs. 16 to 19.

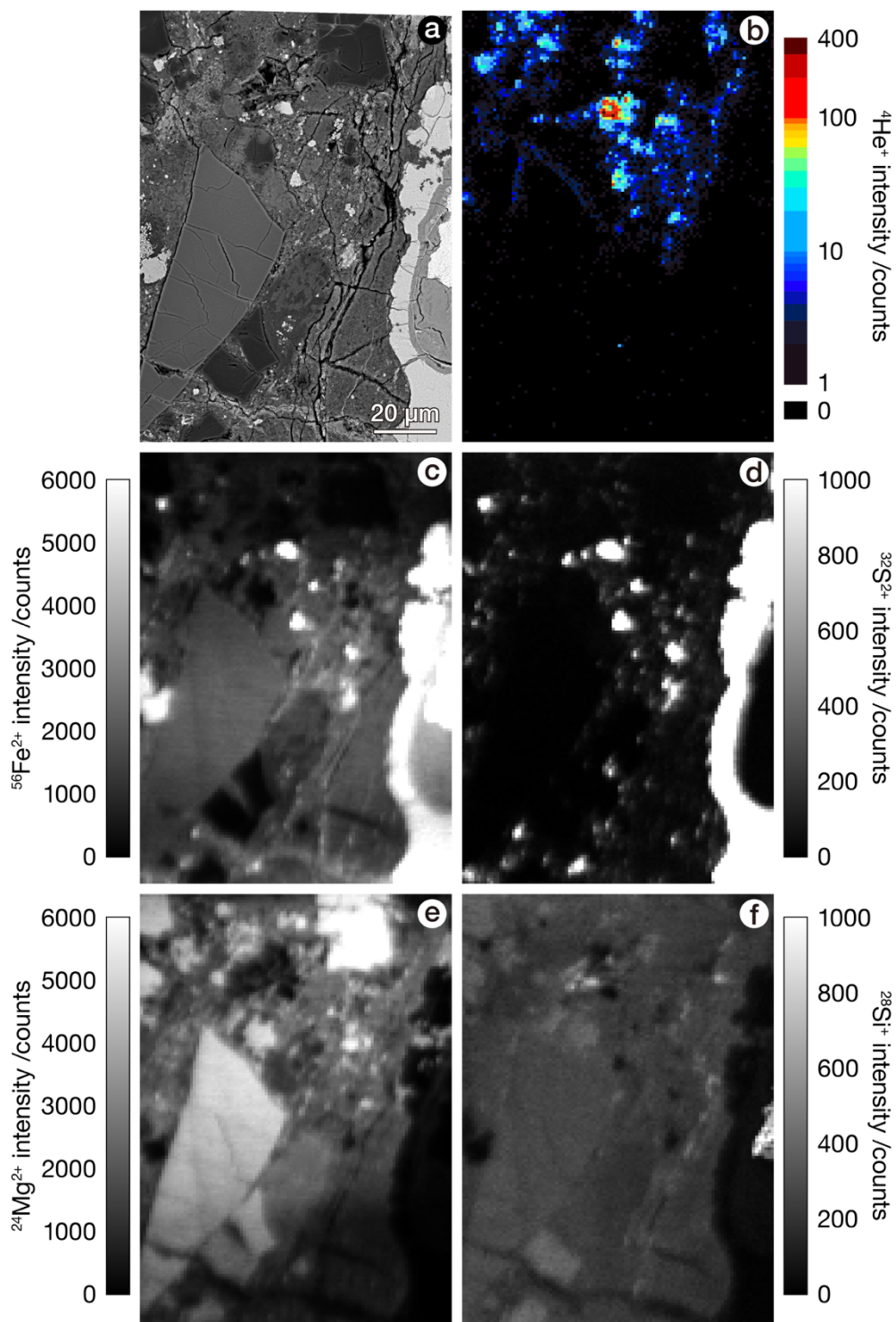


Figure 7. (a) BSE image of Area 1. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

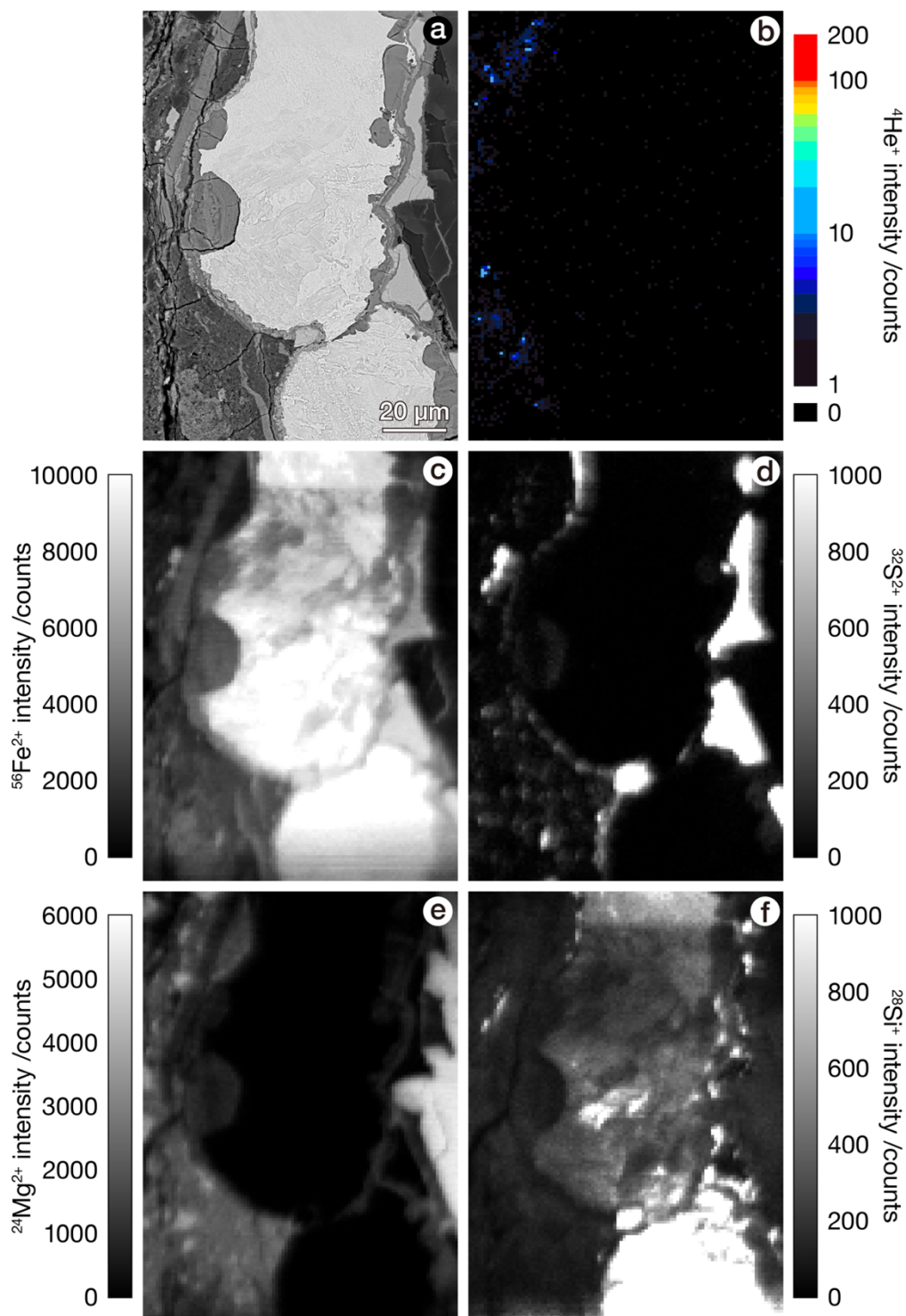


Figure 8. (a) BSE image of Area 2. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

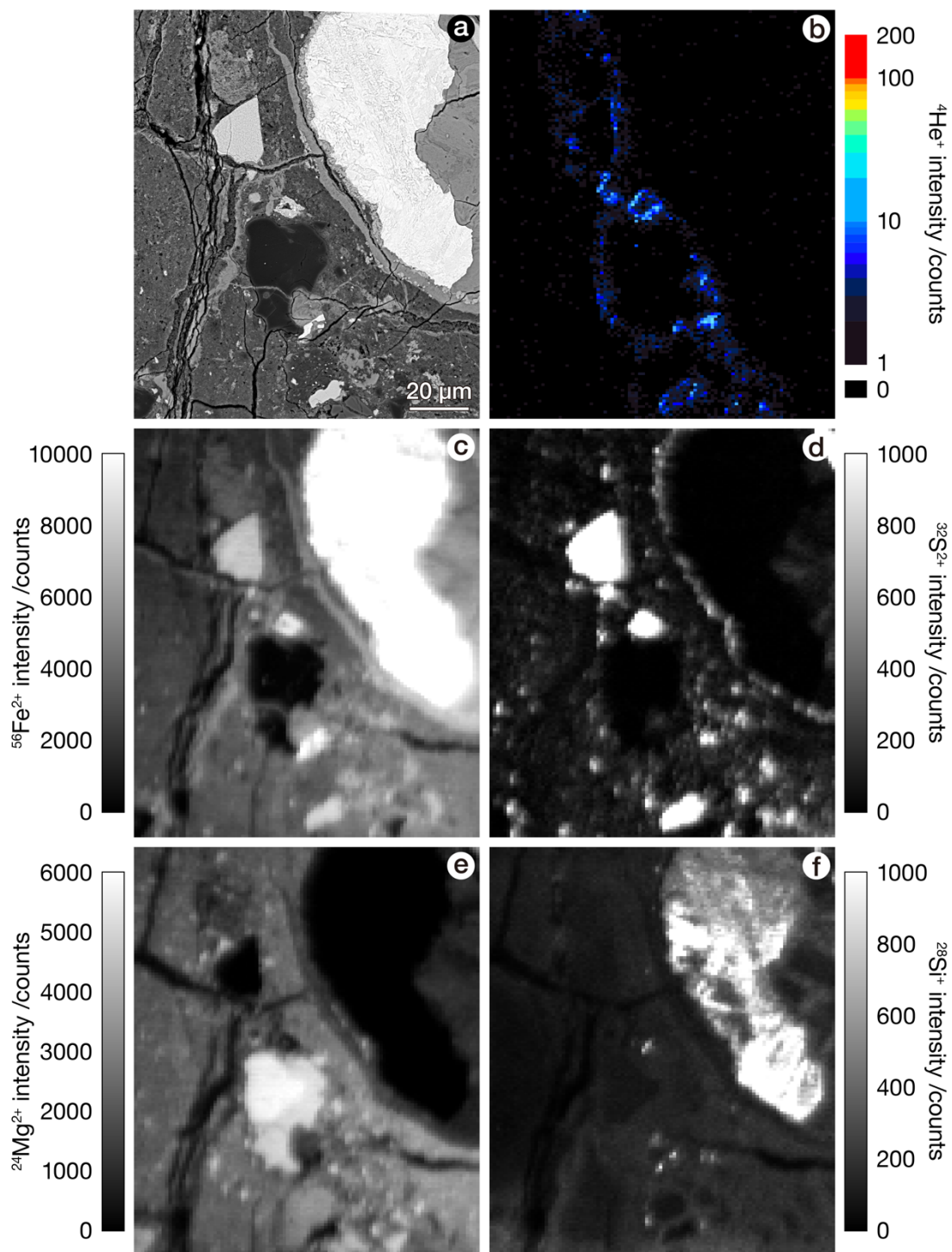


Figure 9. (a) BSE image of Area 3. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

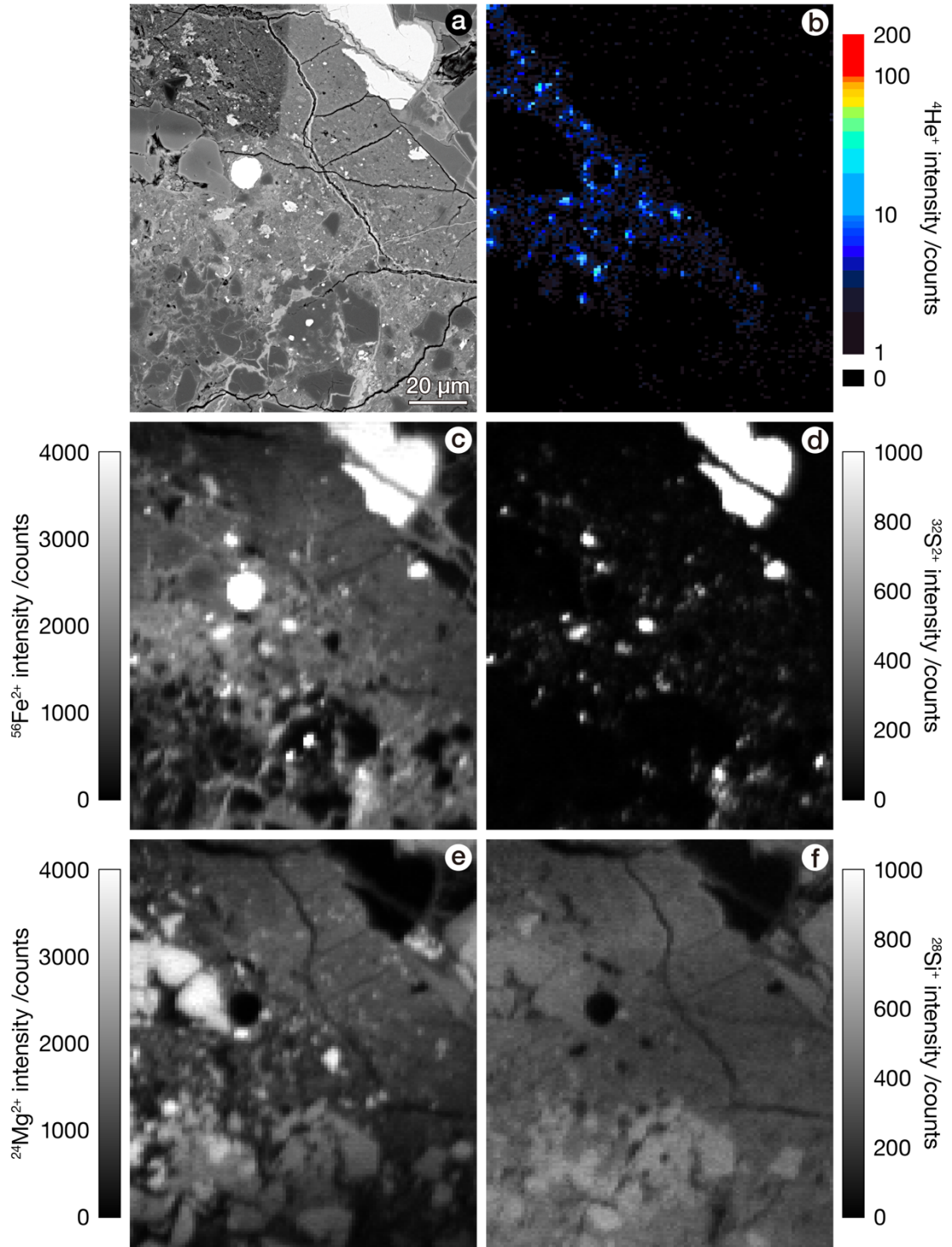


Figure 10. (a) BSE image of Area 4. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

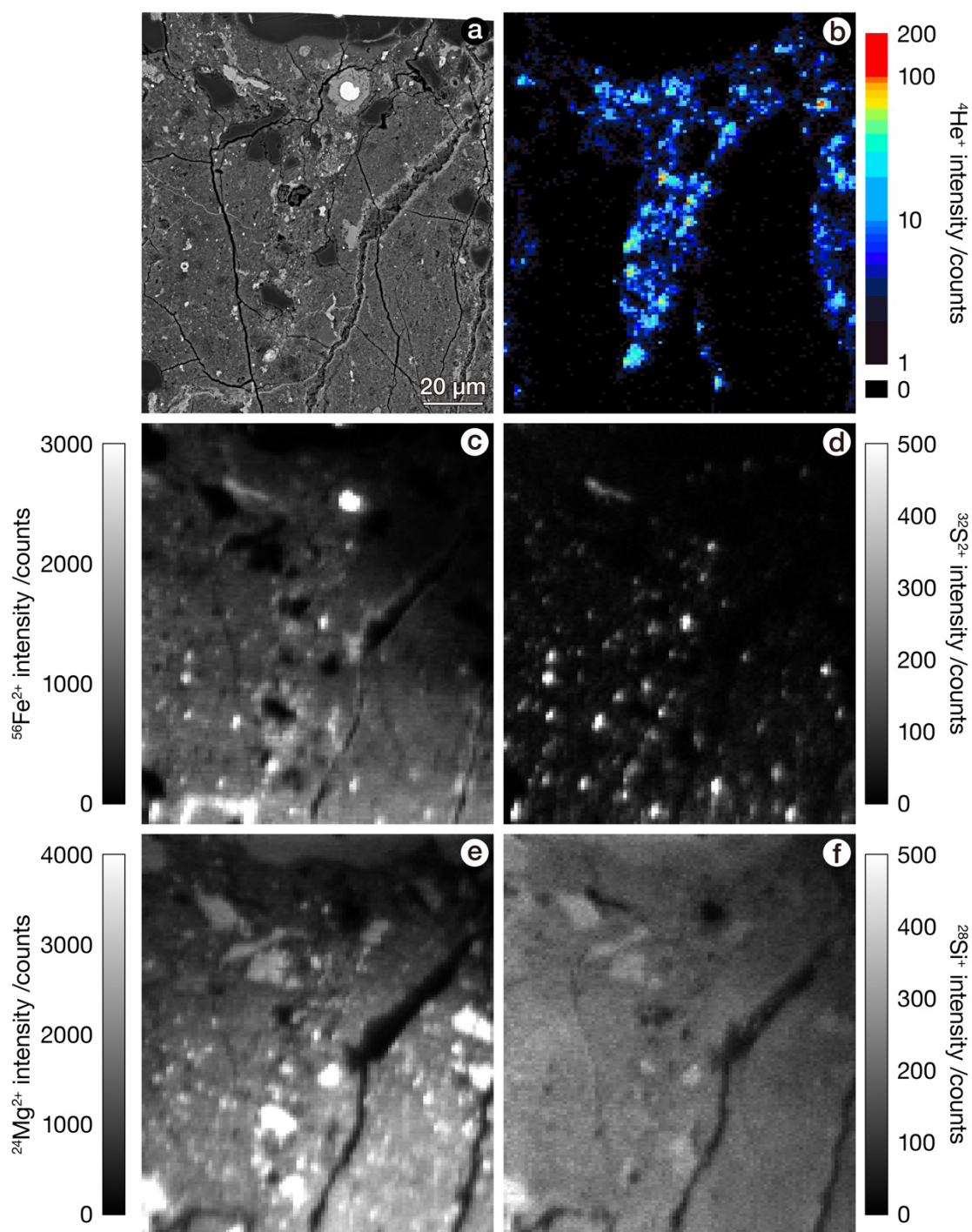


Figure 11. (a) BSE image of Area 5. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

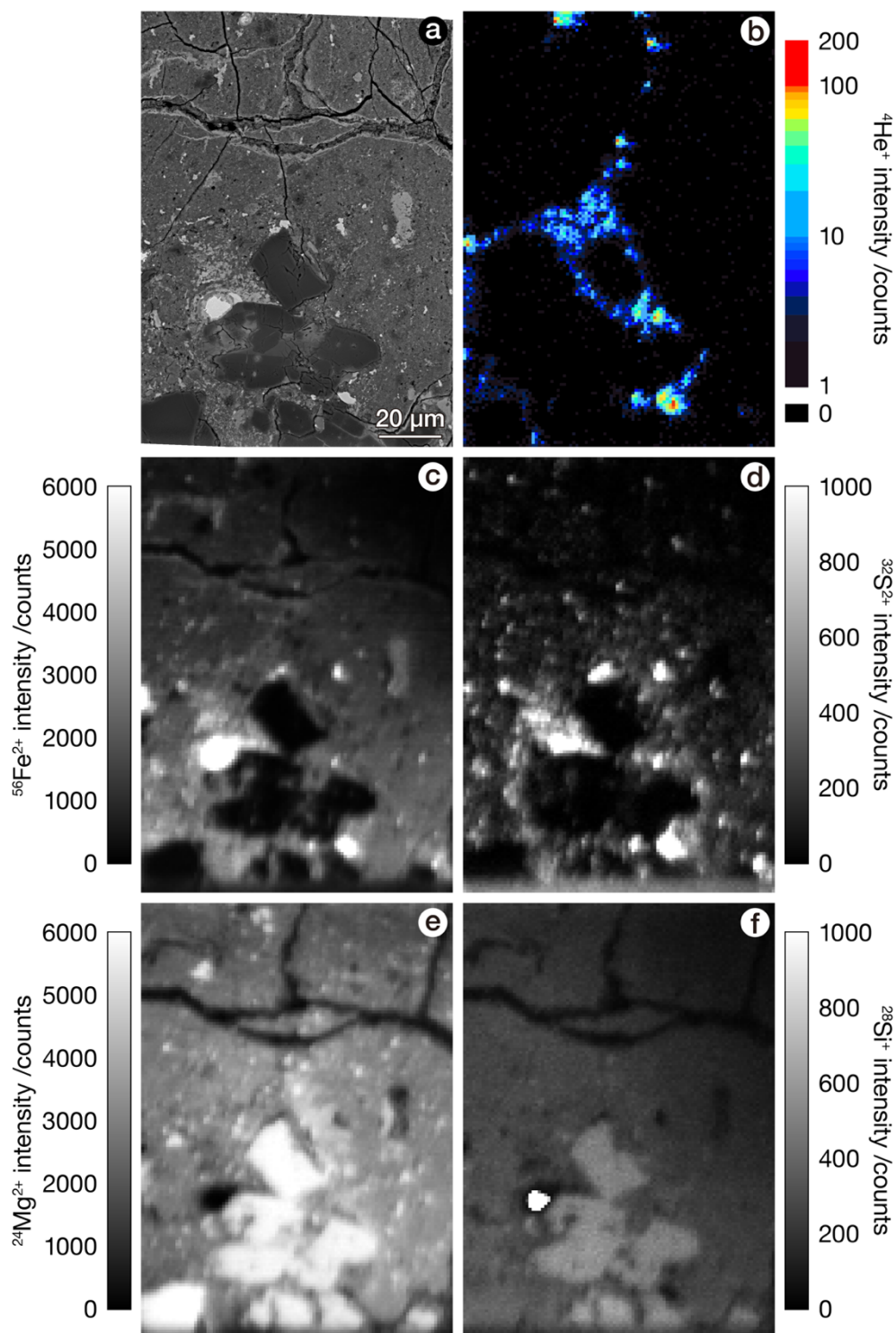


Figure 12. (a) BSE image of Area 6. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

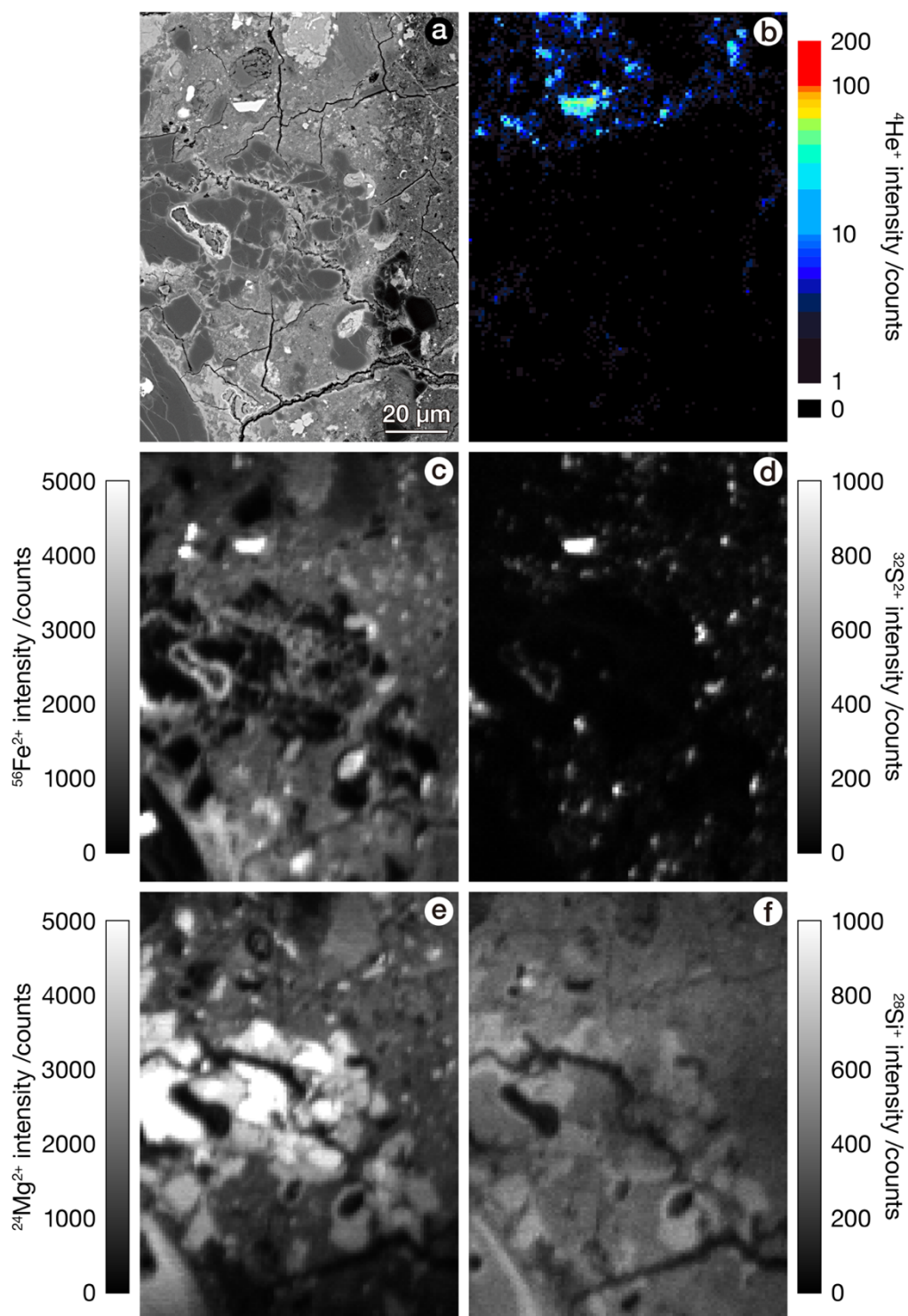


Figure 13. (a) BSE image of Area 7. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

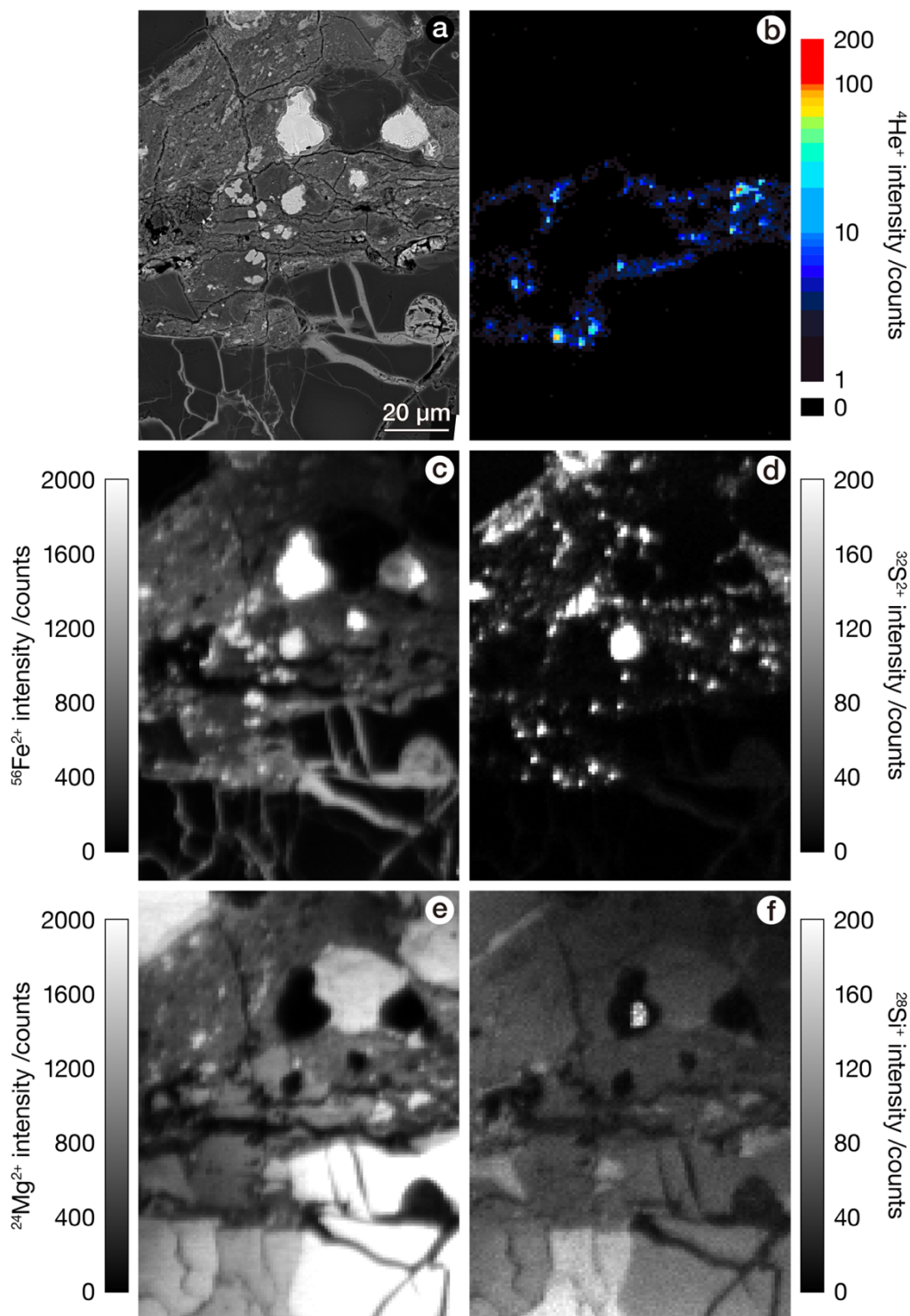


Figure 14. (a) BSE image of Area 8. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

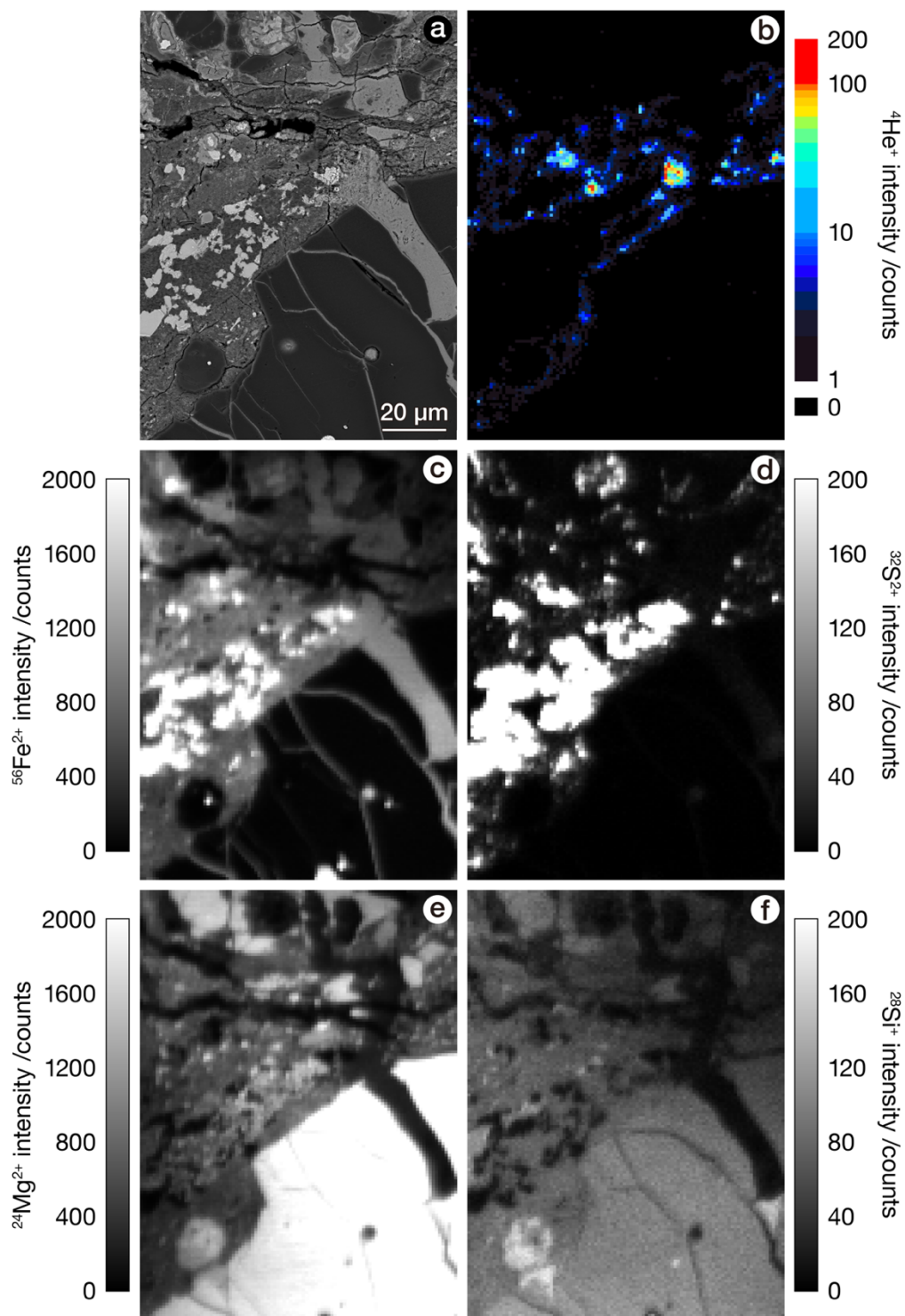


Figure 15. (a) BSE image of Area 9. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

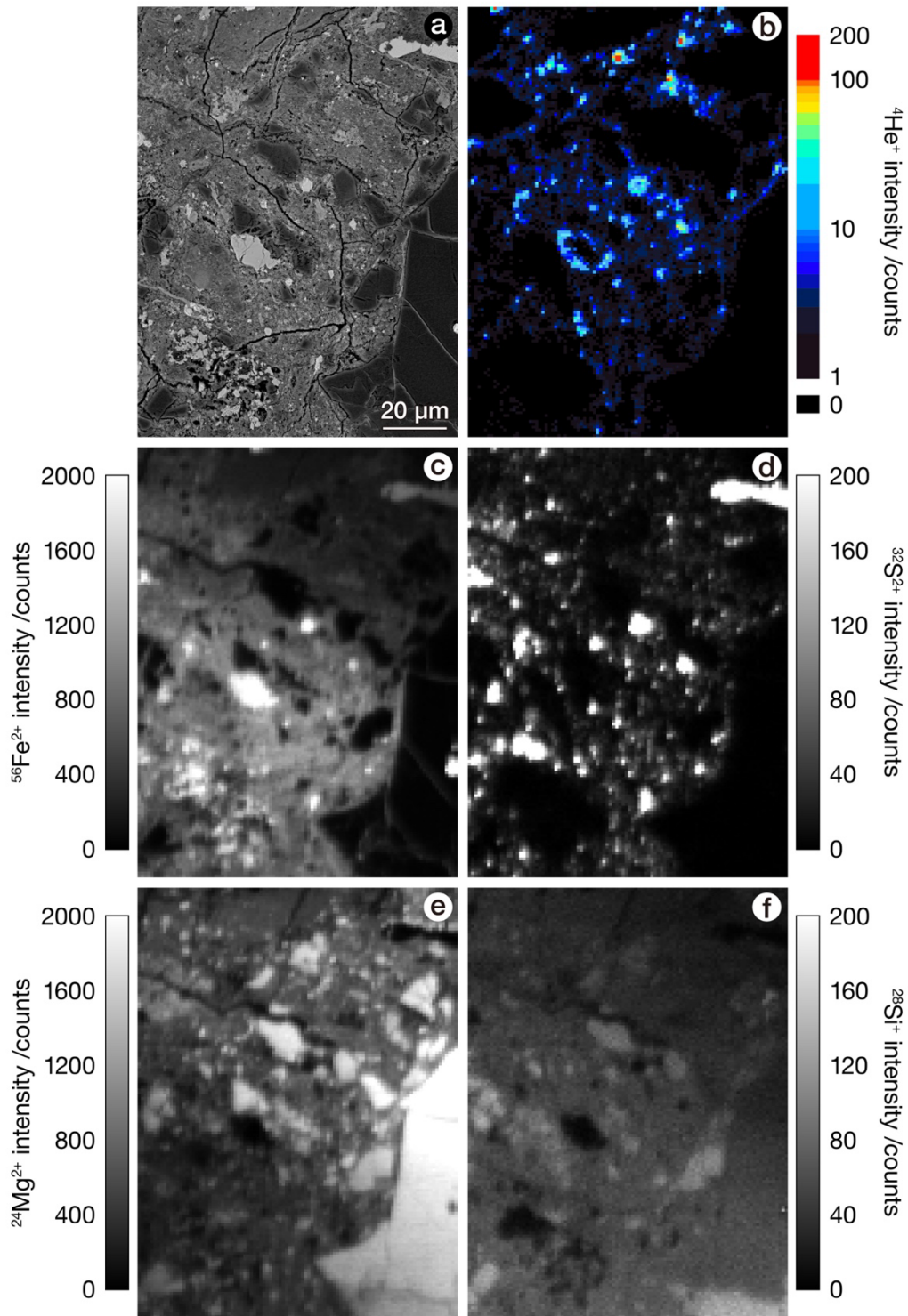


Figure 16. (a) BSE image of Area 10. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

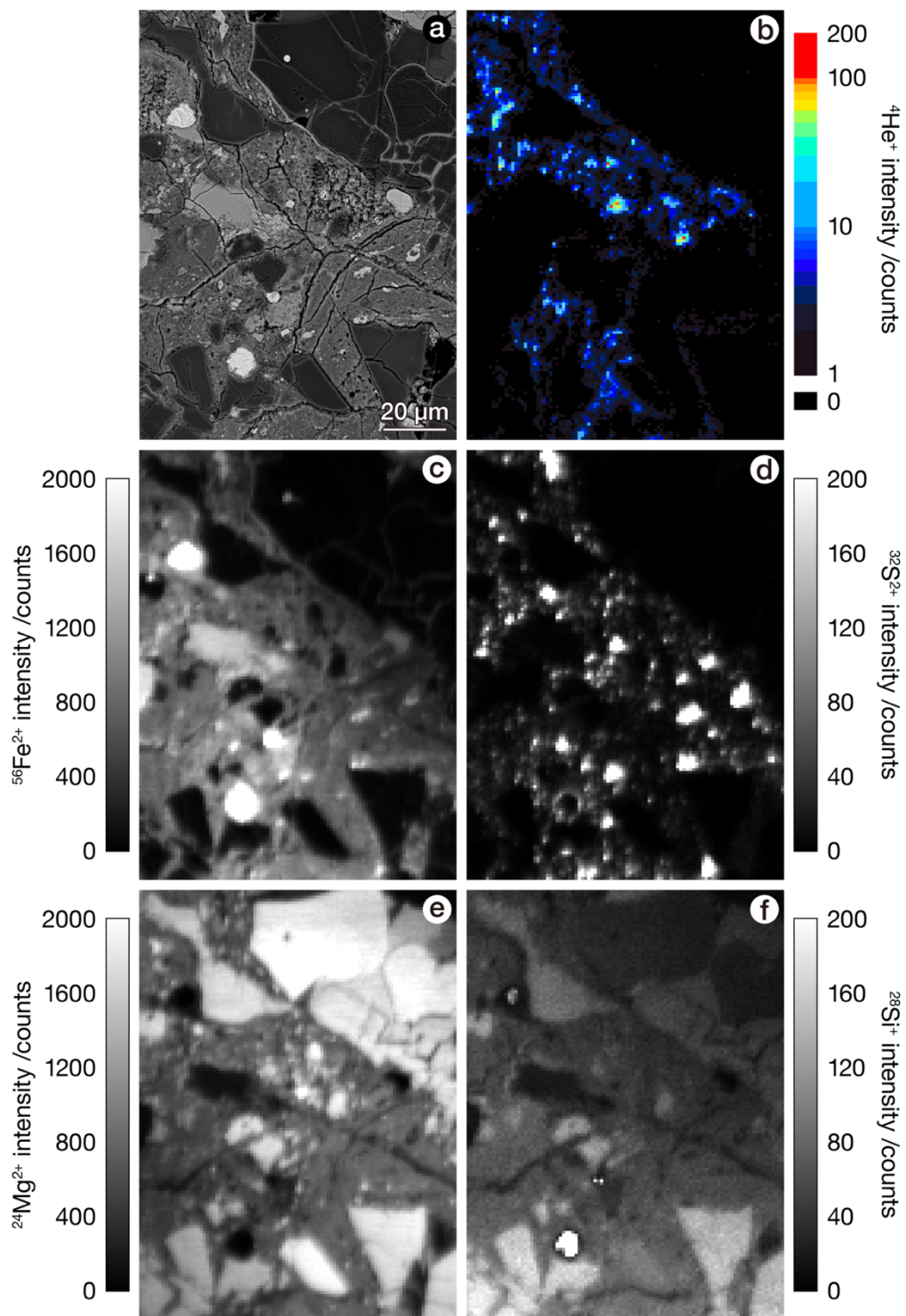


Figure 17. (a) BSE image of Area 11. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

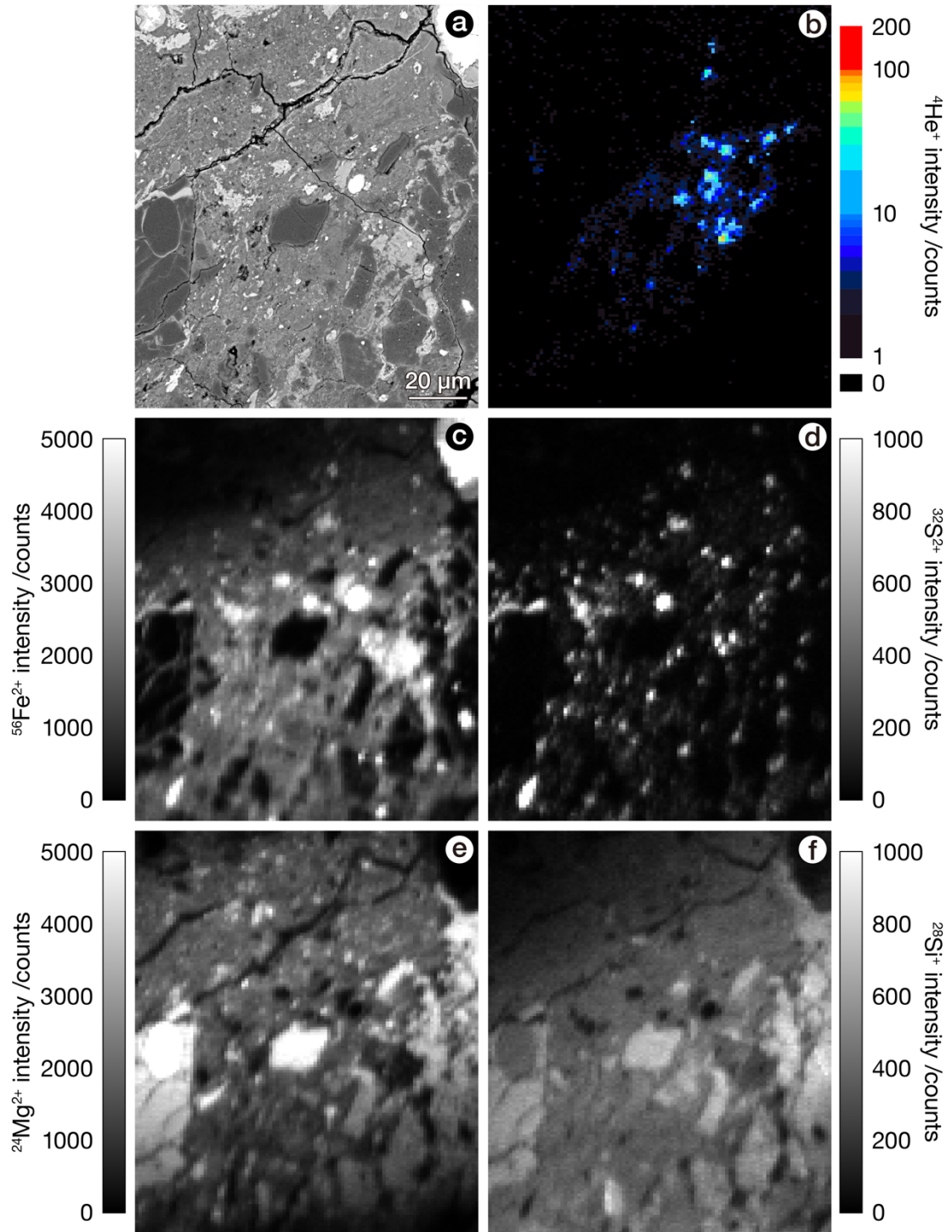


Figure 18. (a) BSE image of Area 12. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

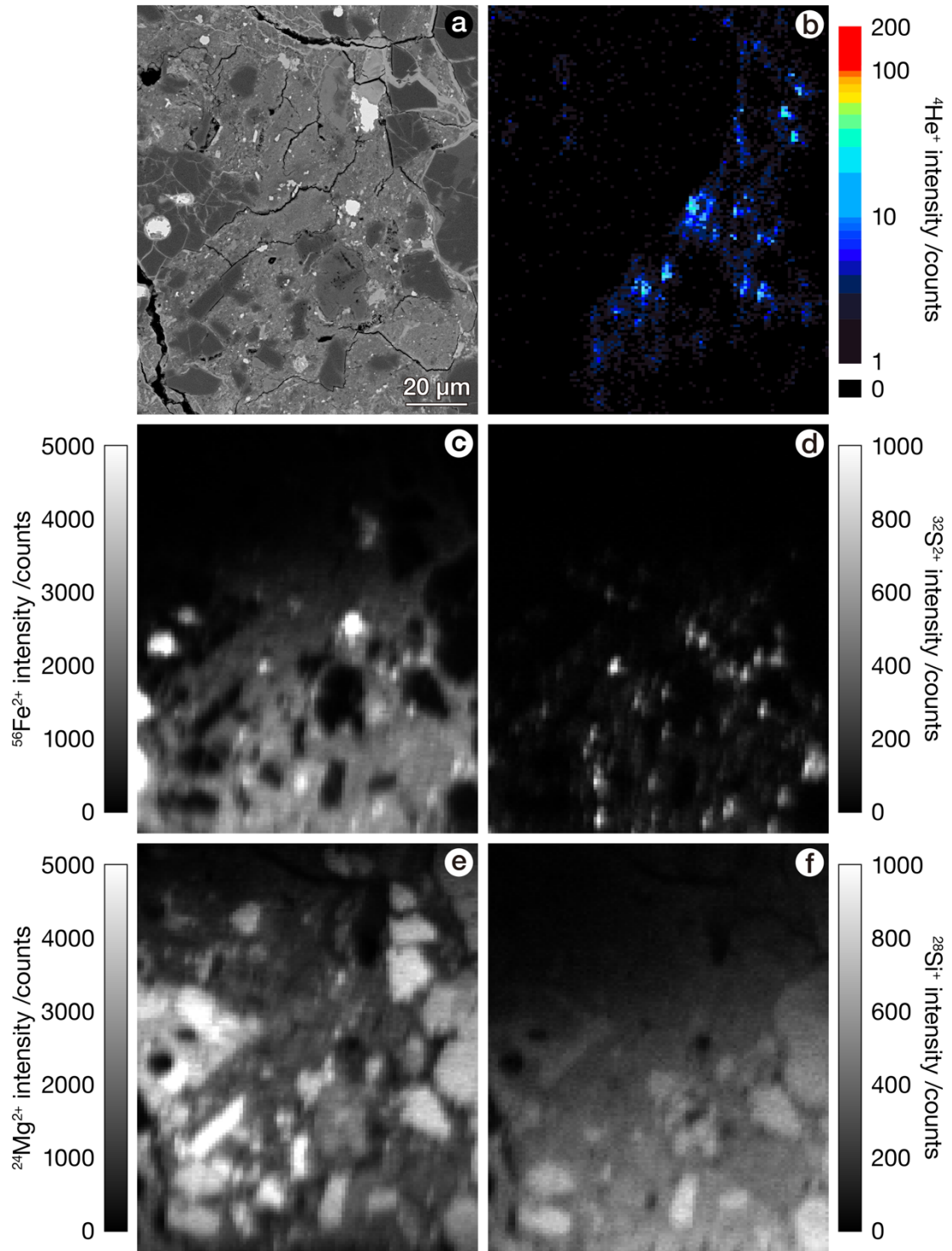


Figure 19. (a) BSE image of Area 13. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

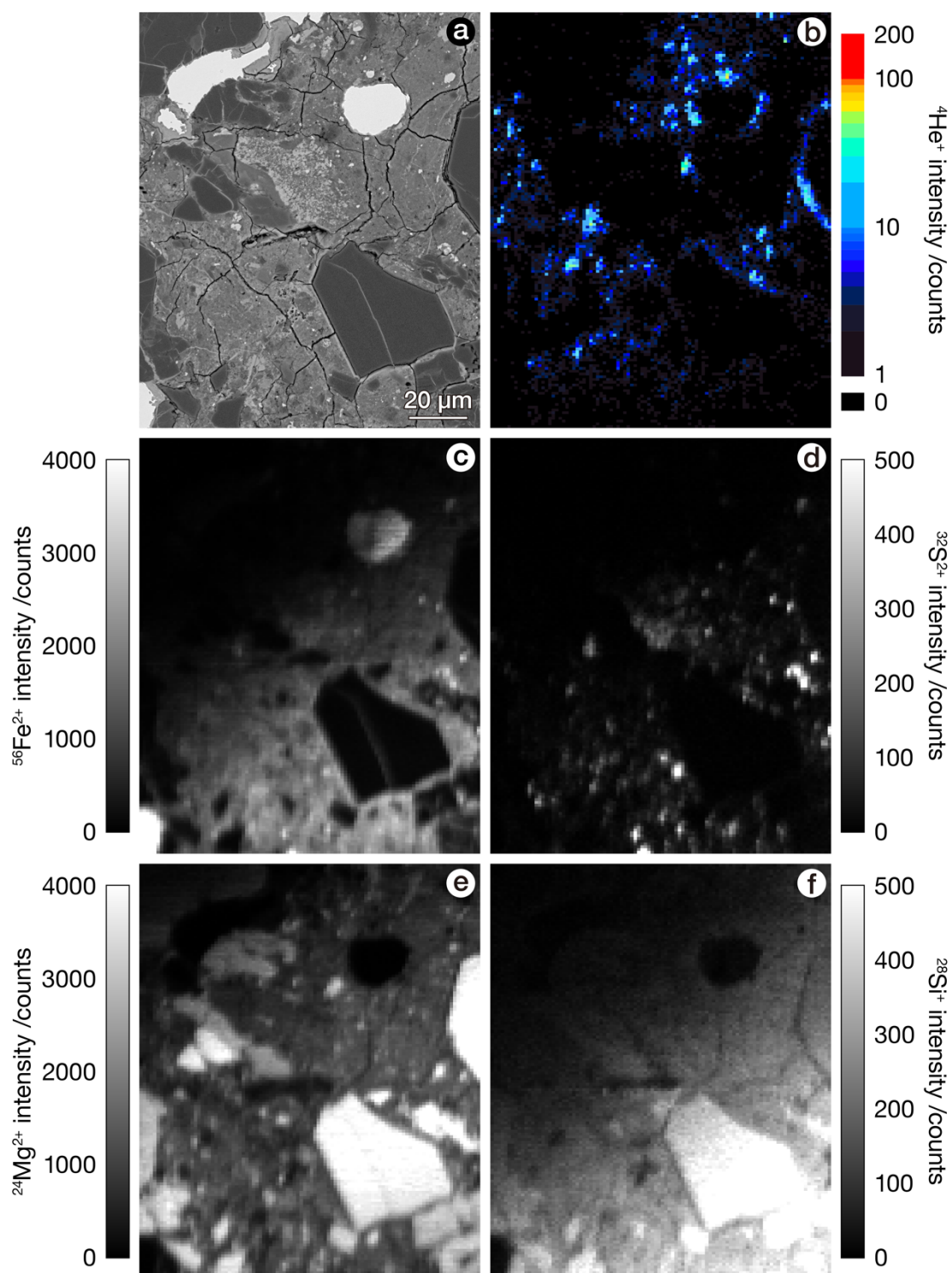


Figure 20. (a) BSE image of Area 14. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

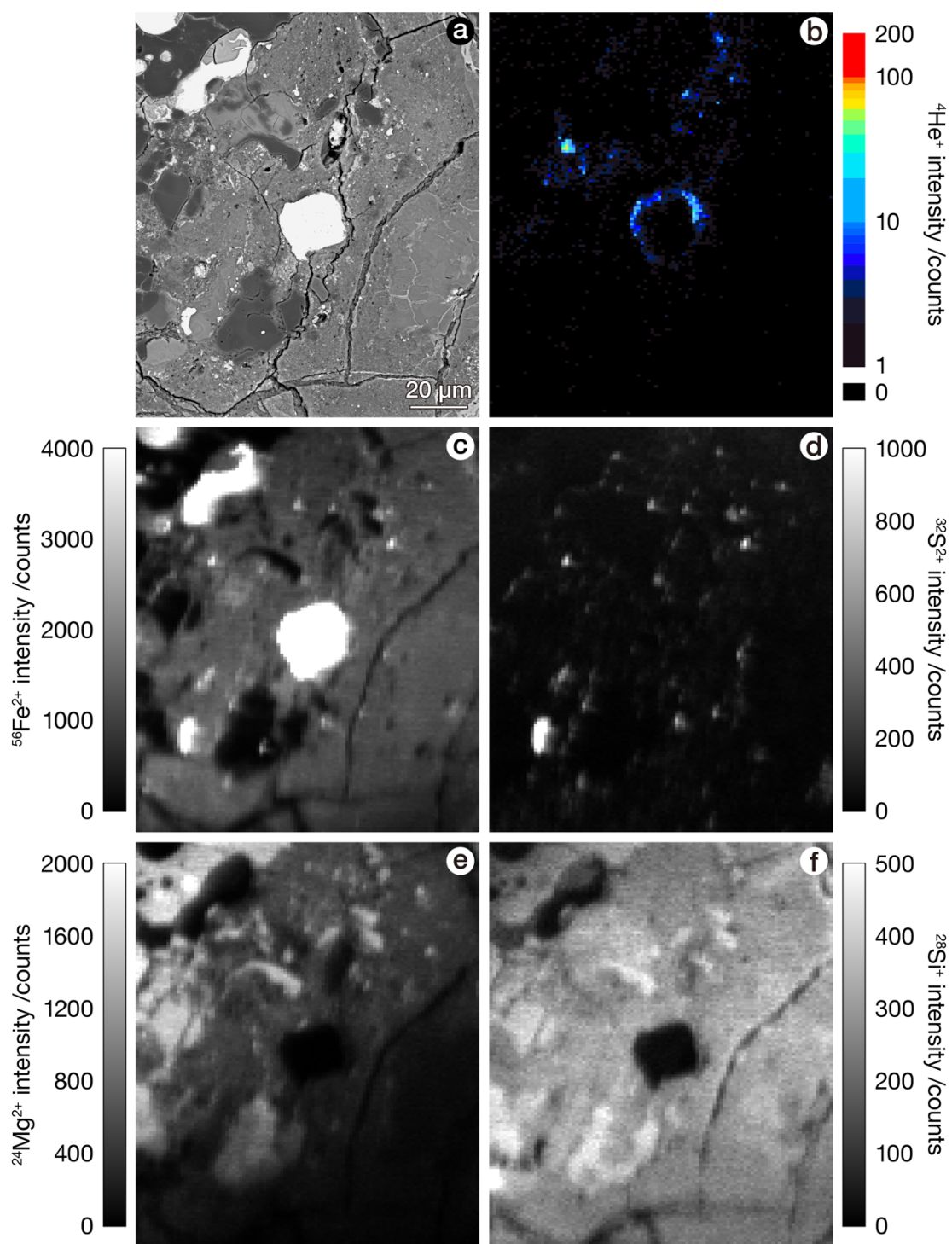


Figure 21. (a) BSE image of Area 15. (b) $^4\text{He}^+$ image of the same area shown in (a). (c) $^{56}\text{Fe}^{2+}$ image of the same area shown in (a). (d) $^{32}\text{S}^{2+}$ image of the same area shown in (a). (e) $^{24}\text{Mg}^{2+}$ image of the same area shown in (a). (f) $^{28}\text{Si}^+$ image of the same area shown in (a).

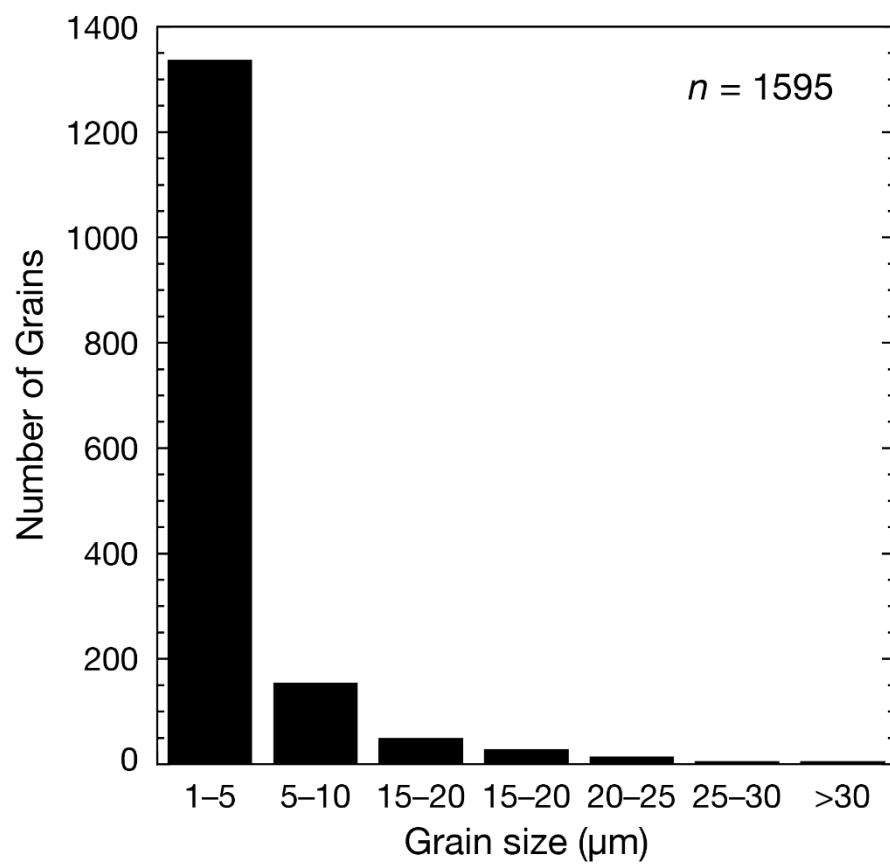


Figure 22. Histogram of grain size distribution for the individual grains.

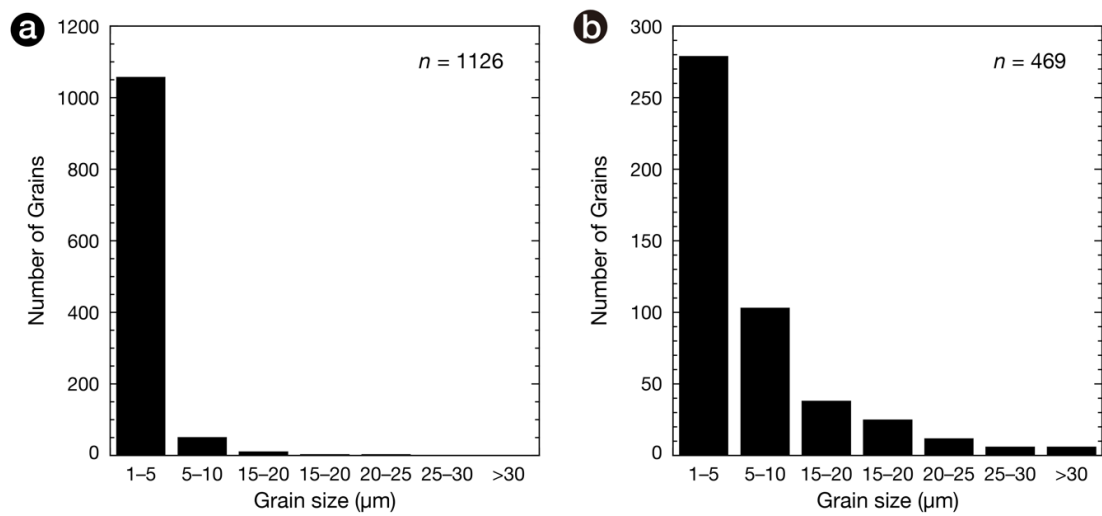


Figure 23. (a) Histogram of grain size distribution for the individual opaque mineral. (b) Histogram of grain size distribution for the individual silicate.

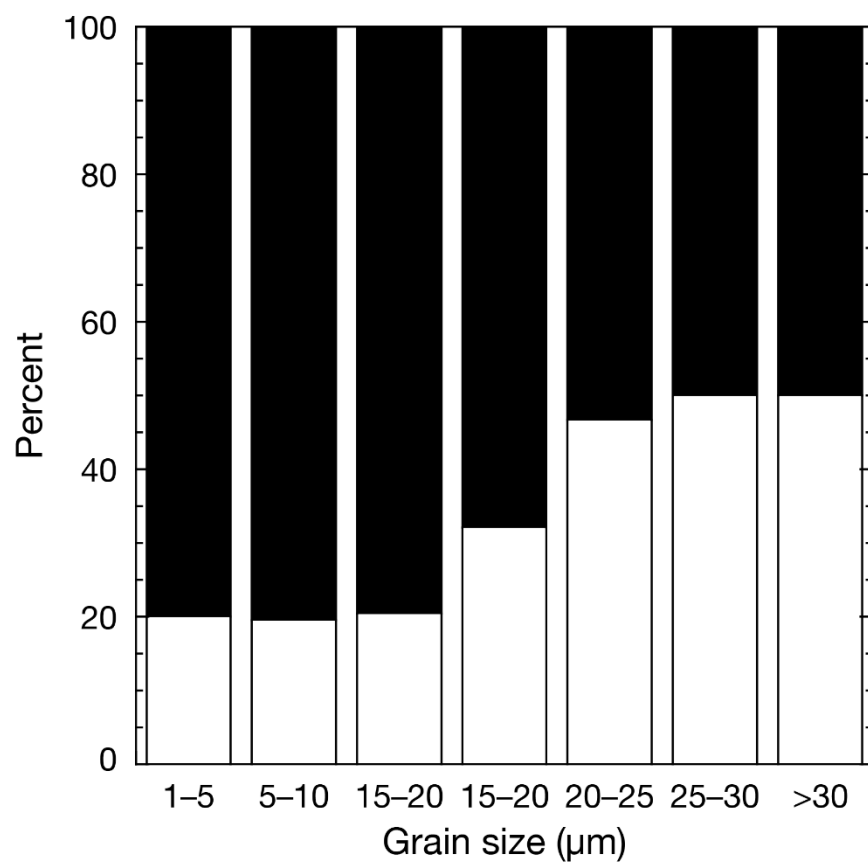


Figure 24. Proportions of ⁴He-bearing grain of each grain size for the individual grains. White bars indicate proportion of ⁴He-bearing grain.

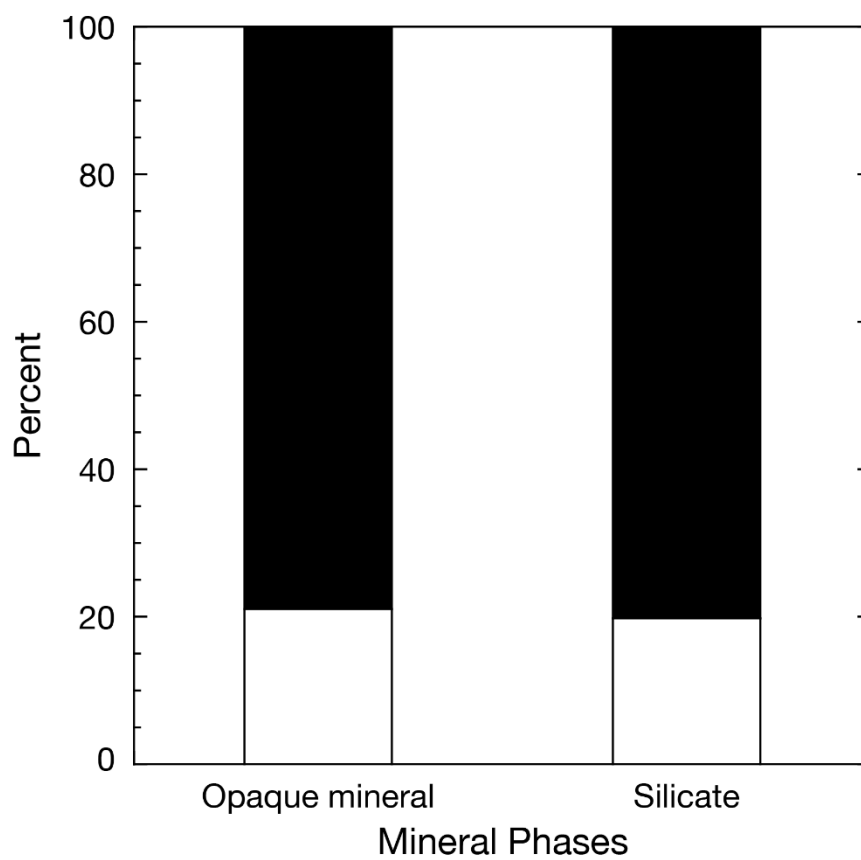


Figure 25. Proportions of ^4He -bearing grain for the individual opaque mineral and silicate. White bars indicate proportion of ^4He -bearing grain.

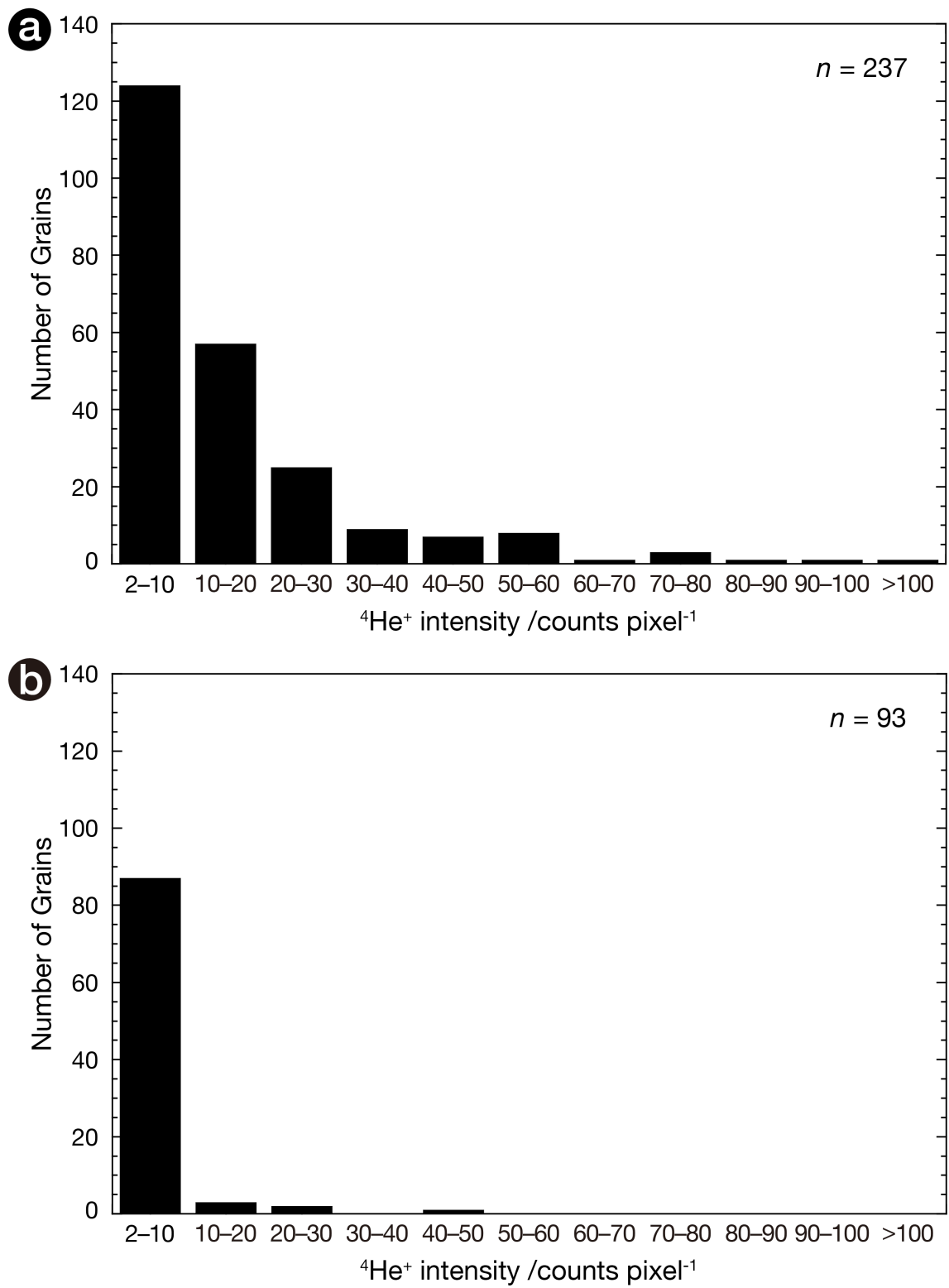


Figure 26. (a) Histogram of $^4\text{He}^+$ intensity for the individual opaque mineral. (b) Histogram of $^4\text{He}^+$ intensity for the individual silicate.

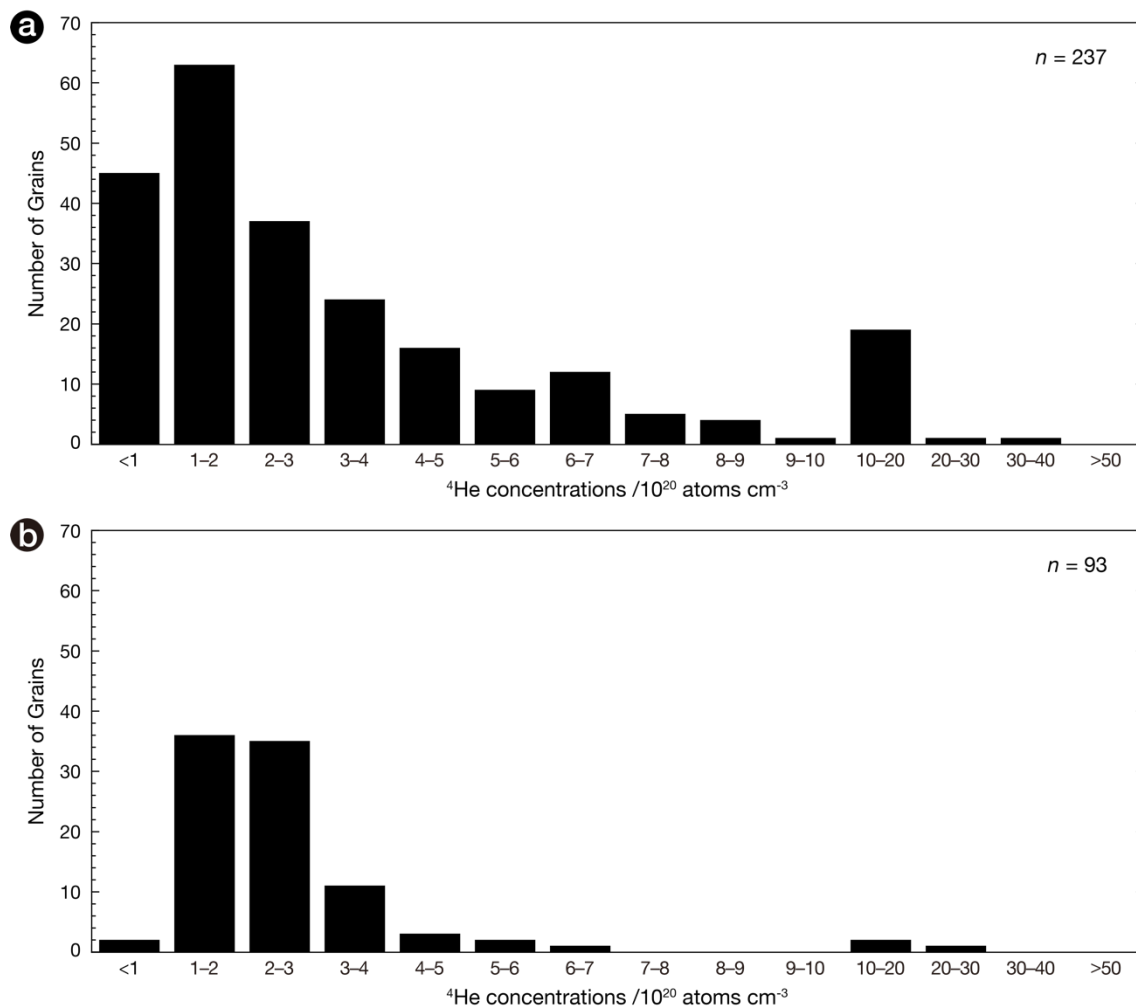


Figure 27. (a) Histogram of ^4He concentrations for the individual opaque mineral. (b) Histogram of ^4He concentrations for the individual silicate.

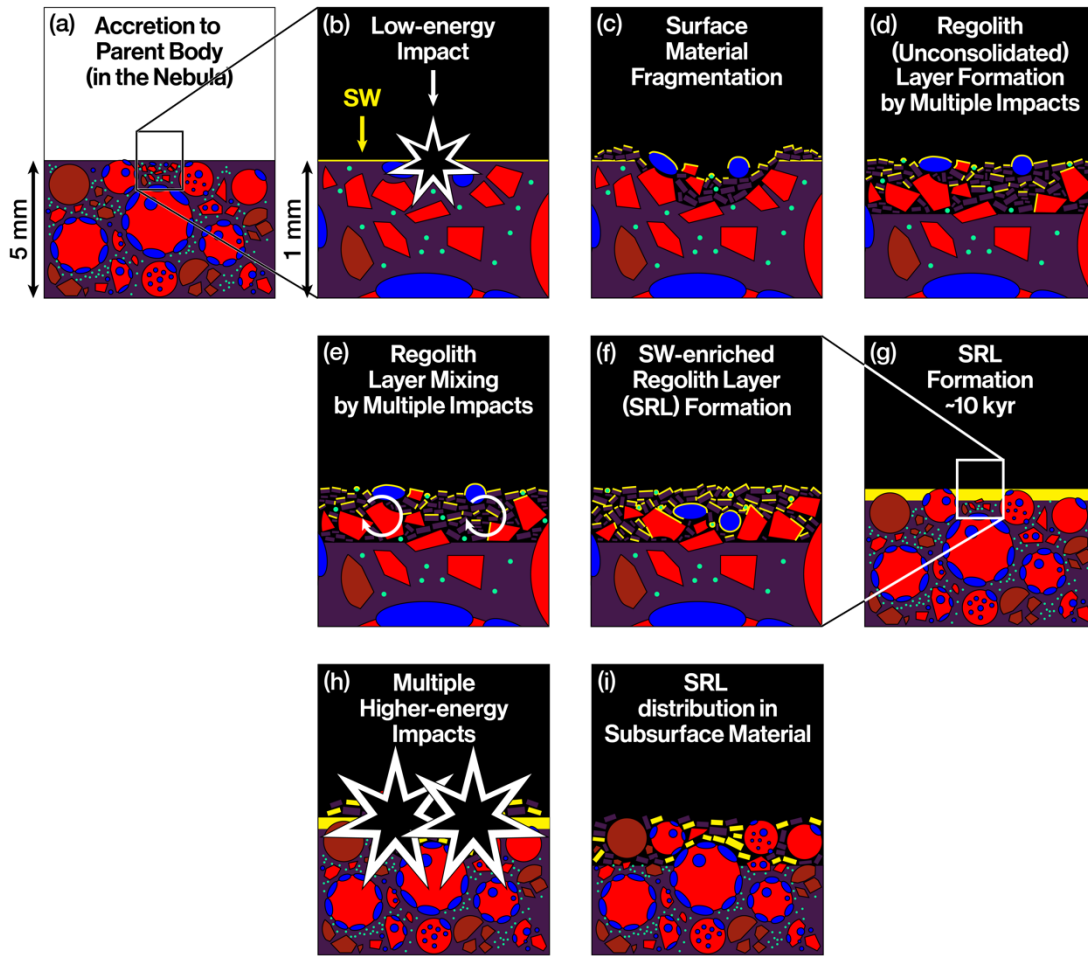


Figure 28. Schematic cartoon of the proposed scenario for the formation of SW- ^4He heterogeneity in NWA 801 meteorite. (a) Estimated initial state of the surface of the parent body. (b) Low-energy impact occurred. (c) Fragmentation of surface material by the low-energy impact. (d) Formation of unconsolidated layer by multiple low-energy impacts. (e) Mixing of the regolith layer by multiple low-energy impacts. (f) Formation of the SW-enriched regolith layer (SRL). (g) SRL covered the surface of the parent body in ~ 10 kyr. (h) Multiple higher-energy impacts occurred. (i) Formation of the SRL distribution in subsurface material.

TABLES

Table 1. Spatial resolutions of isotope images.

Area	$^{56}\text{Fe}^{2+} / \mu\text{m}$	$^{24}\text{Mg}^{2+} / \mu\text{m}$	Average $/ \mu\text{m}$
1	2.3	2.3	2.3
2	1.7	2.2	2.0
3	2.2	2.0	2.1
4	1.9	2.3	2.1
5	2.3	2.0	2.2
6	3.2	2.2	2.7
7	2.0	2.4	2.2
8	1.5	1.8	1.7
9	2.6	2.6	2.6
10	2.3	2.1	2.2
11	3.2	2.8	3.0
12	2.1	2.8	2.5
13	3.1	3.7	3.4
14	2.3	2.8	2.6
15	1.5	2.4	2.0
Average	2.3	2.4	2.4

Table 2. Grain size distribution for the individual opaque mineral and silicate.

Minral	Grain size ($n = 1595$)						
	1–5 μm	5–10 μm	10–15 μm	15–20 μm	20–25 μm	25–30 μm	>30 μm
Troilite	578	34	4	1	1	0	0
Ferrihydrite	429	12	5	2	0	0	0
Fe-Ni metal	47	5	2	0	2	0	0
Magnetite	4	0	0	0	0	0	0
Olivine	85	47	13	9	6	3	3
Pyroxene	194	56	25	16	6	3	3
Opaque*	1058	51	11	3	3	0	0
Silicate**	279	103	38	25	12	6	6

*Opaque: Troilite + Ferrihydrite + Fe-Ni metal + Magnetite, **Silicate: Olivine + Pyroxene

Table 3. The number of individual mineral grains of each grain size.

Area	⁴ He	Grain size (<i>n</i> = 1595)						
		1–5 µm	5–10 µm	10–15 µm	15–20 µm	20–25 µm	25–30 µm	>30 µm
1	detected	40	6	0	2	0	1	1
	n.d.	22	7	1	1	1	1	0
2	detected	12	5	0	0	0	0	0
	n.d.	7	4	0	0	0	0	0
3	detected	14	1	2	0	1	0	0
	n.d.	47	5	2	0	0	0	0
4	detected	14	4	2	0	0	0	0
	n.d.	29	8	3	4	0	0	0
5	detected	39	2	3	1	1	0	0
	n.d.	114	8	3	2	1	0	0
6	detected	13	1	0	0	0	2	0
	n.d.	63	4	0	1	0	2	0
7	detected	7	0	1	0	0	0	0
	n.d.	89	8	4	0	0	0	0
8	detected	12	1	0	0	0	0	0
	n.d.	92	7	0	0	0	0	0
9	detected	13	2	0	1	0	0	0
	n.d.	65	16	4	0	0	0	0
10	detected	30	2	1	3	0	0	0
	n.d.	91	11	4	1	0	0	0
11	detected	26	2	0	0	2	0	0
	n.d.	77	11	2	1	0	0	2
12	detected	18	1	0	0	0	0	0
	n.d.	145	8	5	2	2	0	0
13	detected	17	2	1	2	1	0	0
	n.d.	83	15	5	4	3	0	0
14	detected	10	1	0	0	1	0	2
	n.d.	62	7	4	1	0	0	0
15	detected	3	0	0	0	1	0	0
	n.d.	83	5	2	2	1	0	1
Total	detected	268	30	10	9	7	3	3
	n.d.	1069	124	39	19	8	3	3
	Percent (%)	20.0	19.5	20.4	32.1	46.7	50.0	50.0

Table 4. The number of individual mineral grains of each mineral phase.

Area	⁴ He	Mineral (<i>n</i> = 1595)							
		Troilite	Ferrihydrite	Fe-Ni metal	Magnetite	Olivine	Pyroxene	Opaque*	Silicate**
1	detected	26	6	1	2	12	3	35	15
	n.d.	19	0	0	0	13	1	19	14
2	detected	3	3	0	0	6	5	6	11
	n.d.	4	2	0	0	4	1	6	5
3	detected	10	3	1	0	0	4	14	4
	n.d.	11	14	1	0	17	11	26	28
4	detected	11	1	2	0	4	2	14	6
	n.d.	8	7	5	0	16	8	20	24
5	detected	16	9	0	0	4	17	25	21
	n.d.	58	22	0	0	17	33	80	50
6	detected	6	3	0	0	2	5	9	7
	n.d.	32	16	0	0	11	11	48	22
7	detected	4	3	0	1	0	0	8	0
	n.d.	24	36	3	1	4	33	64	37
8	detected	8	4	0	0	0	1	12	1
	n.d.	29	41	1	0	1	27	71	28
9	detected	10	4	0	0	1	1	14	2
	n.d.	69	12	0	0	0	4	81	4
10	detected	17	10	0	0	0	9	27	9
	n.d.	32	20	0	0	6	49	52	55
11	detected	17	3	1	0	3	6	21	9
	n.d.	22	20	12	0	10	29	54	39
12	detected	15	1	3	0	0	0	19	0
	n.d.	68	52	23	0	5	14	143	19
13	detected	15	2	1	0	1	4	18	5
	n.d.	34	50	0	0	14	12	84	26
14	detected	7	3	1	0	3	0	11	3
	n.d.	20	43	0	0	10	1	63	11
15	detected	3	0	1	0	0	0	4	0
	n.d.	22	58	0	0	2	12	80	14
Total	detected	168	55	11	3	36	57	237	93
	n.d.	452	393	45	1	130	246	891	376
	Percent (%)	27.1	12.3	19.6	75.0	21.7	18.8	21.0	19.8

*Opaque: Troilite + Ferrihydrite + Fe-Ni metal + Magnetite, **Silicate: Olivine + Pyroxene

4. SUMMARY OF THIS THESIS

Regolith materials on the airless bodies have been exposed to the SW. The SW irradiation timescale is a key indicator of the timescale of geological processes on an asteroid. Here I carried out SW- ^4He distributions in the NWA 801 CR2 chondrite, which contains a large amount of a solar wind noble gas, on the sub-micrometer to micrometer scale using He isotope imaging.

First, I conducted *in situ* ^4He isotope imaging to observe the direct evidence of SW implantation in individual mineral grains. The isotope images of individual Fe-Ni metal, ferrihydrite, olivine, pyroxene, and troilite grains showed that ^4He is detected along grain interfaces, indicating its presence at depths shallower than 100 nm from the grain surfaces. The SW- ^4He fluence of the individual mineral grains ranged from 2.7 to 58×10^{16} atoms cm^{-2} . The SW irradiation durations were calculated to range from 3.8 to 82 kyr, assuming the SW irradiation at 4 au. These durations correspond to representing the residence time of the individual mineral grains on the surface of the parent body. The residence times of most of the individual mineral grains suggest that the mixing process on the parent body typically occurred with a frequency of about ten thousand years.

Second, based on the study of the SW- ^4He sub-micrometer distribution for the individual mineral grains, I then visualized the sedimentary structure of the NWA 801 sample. To extend the imaging area, I implemented the “large-area ion imaging system” in LIMAS. ^4He imaging over an area of $100 \times 150 \mu\text{m}$ was achieved. Improved *in situ* ^4He imaging technique was used to elucidate the relationship between the heterogeneity of SW- ^4He and petrological characteristics in NWA 801. The SW- ^4He -bearing mineral grains are predominantly distributed within the coarse grain-rich matrix.

In contrast, the fine grain-rich matrix occurs interstitially between the coarse grain-rich matrix and chondrules, measuring several tens to hundreds of micrometers in size. No detectable ^4He was observed in the fine grain-rich matrix. Based on the similarities in occurrence, the fine grain-rich matrix observed in this study is thought to be the same material as FGRs. The partial preservation of FGRs suggests that the daily impacts associated with gardening process primarily caused the disaggregation of loosely bound components. The heterogeneous distribution of SW- ^4He in the interstitial matrix is originated from the low-energy impacts that induce the semi-conservative fragmentation of FGRs, which were the main process of regolith gardening on the parent body of NWA 801. Furthermore, the proportions of ^4He -bearing grains in opaque minerals and silicates are nearly identical ($\sim 20\%$), suggesting that SW irradiation occurred irrespective of mineral type, affecting both opaque minerals and silicates. If the regolith preserved in the meteorite reflects the global surface characteristics of the parent body, the proportion of ^4He -bearing grains implies that approximately 20% of the unconsolidated surface layer was exposed to SW for a sufficiently long duration to accumulate detectable SW- ^4He , while the remaining 80% consisted of “immature regolith” that experienced minimal SW exposure (likely less than a few hundred years). The surface of the parent body of NWA 801 was likely composed largely of immature regolith, similar to present-day asteroids.

The ^4He distribution in the NWA 801 meteorite, as revealed by ^4He isotope imaging, is direct evidence of the SW implantation on the uppermost surface of the parent body. Furthermore, the SW- ^4He fluence to individual mineral grains was successfully quantified from the ^4He intensity. High-resolution quantitative ^4He isotope imaging established in this study enable us to elucidate the past asteroidal regolith dynamics from various meteorites and returned samples.

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SUPPLEMENTARY MATERIALS

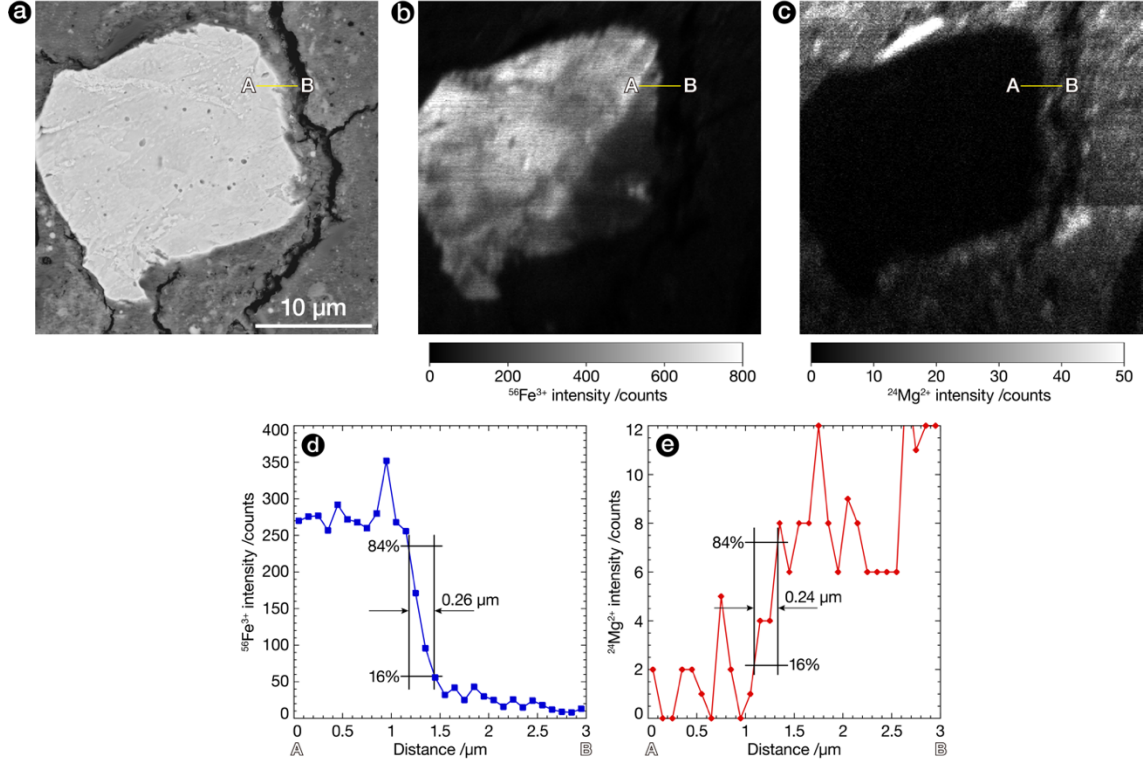


Figure S1. (a) BSE, (b) $^{56}\text{Fe}^{2+}$ ion, and (c) $^{24}\text{Mg}^{2+}$ ion images of Area 1. (d) Line profile for $^{56}\text{Fe}^{3+}$ along traverse A–B, representing the yellow line in (b). (e) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (c).

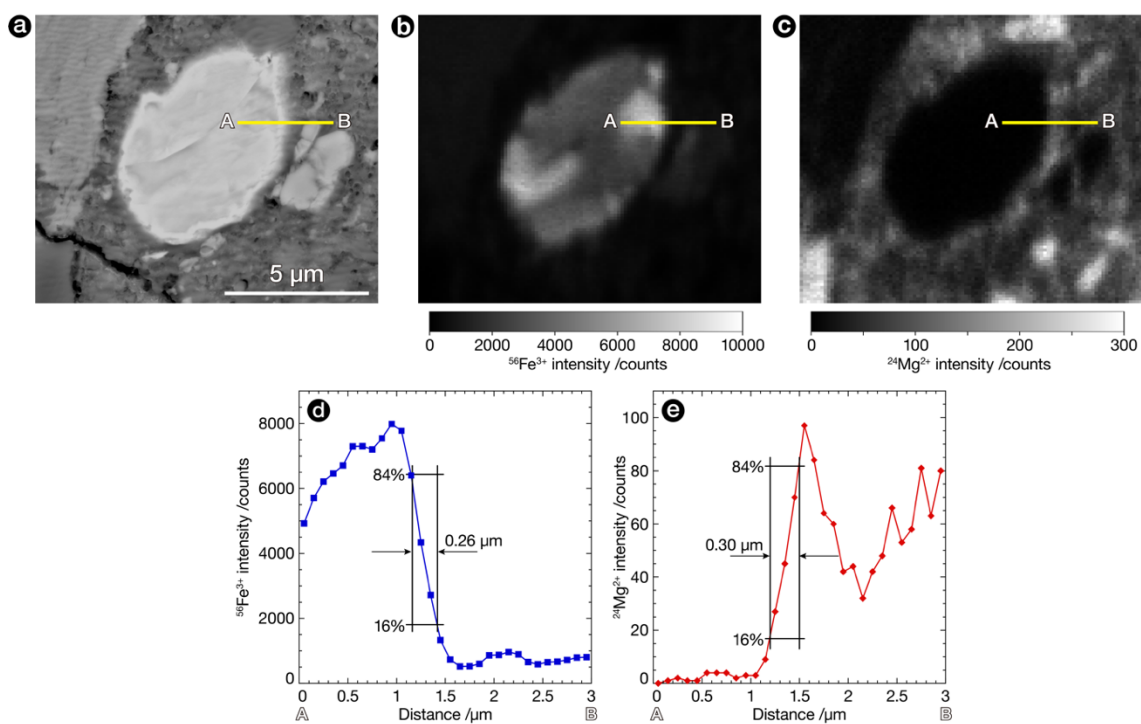


Figure S2. (a) BSE, (b) $^{56}\text{Fe}^{2+}$ ion, and (c) $^{24}\text{Mg}^{2+}$ ion images of Area 2. (d) Line profile for $^{56}\text{Fe}^{3+}$ along traverse A–B, representing the yellow line in (b). (e) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (c).

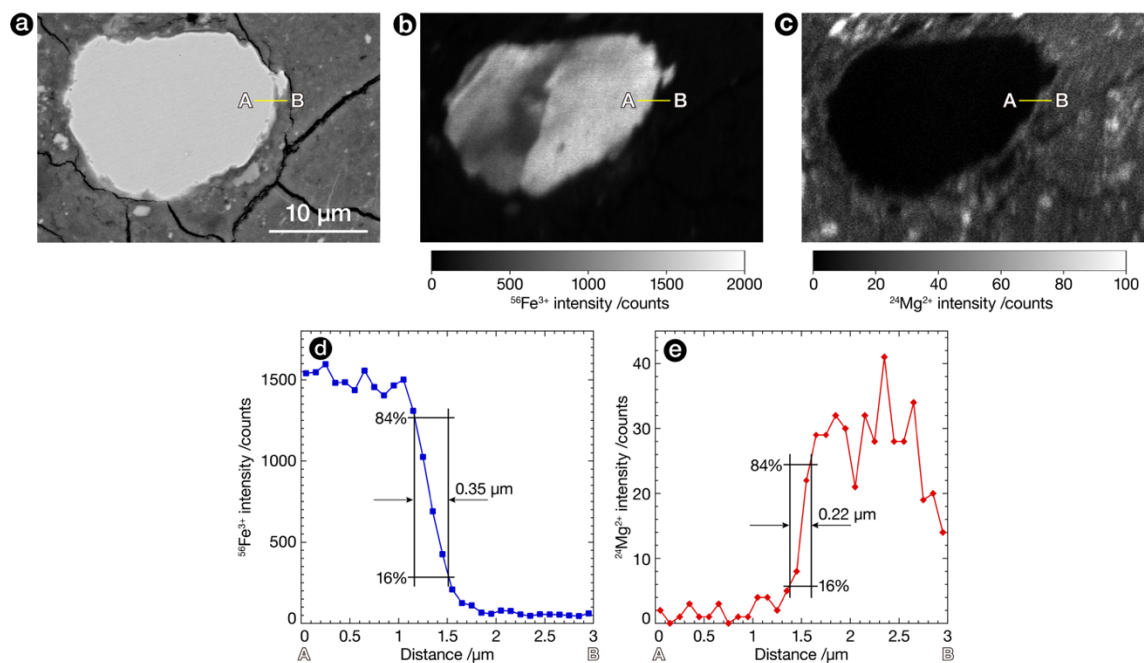


Figure S3. (a) BSE, (b) $^{56}\text{Fe}^{2+}$ ion, and (c) $^{24}\text{Mg}^{2+}$ ion images of Area 3. (d) Line profile for $^{56}\text{Fe}^{3+}$ along traverse A–B, representing the yellow line in (b). (e) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (c).

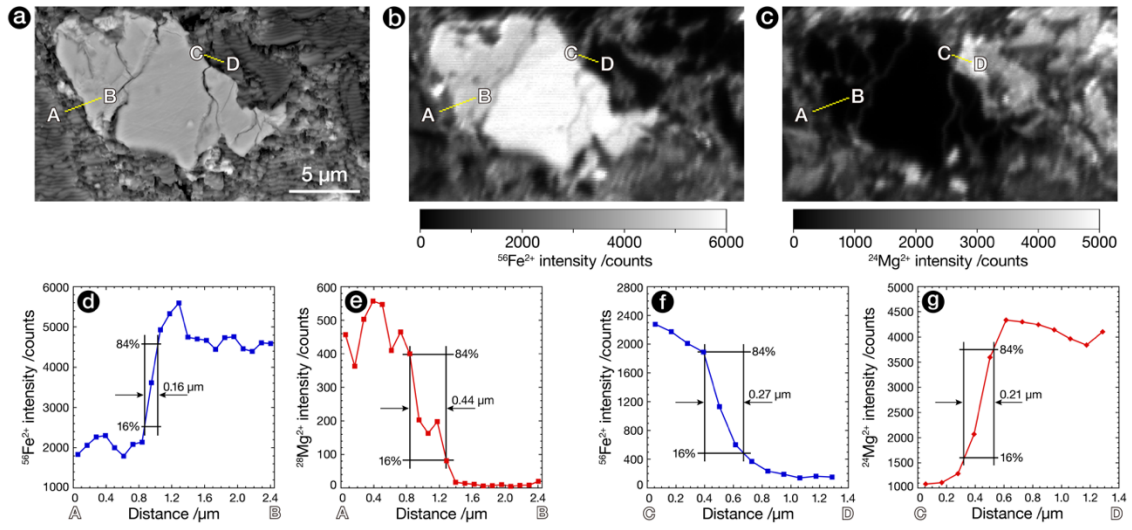


Figure S4. (a) BSE, (b) $^{56}\text{Fe}^{2+}$ ion, and (c) $^{24}\text{Mg}^{2+}$ ion images of Area 4. (d) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (b). (e) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (c). (f) Line profile for $^{56}\text{Fe}^{2+}$ along traverse C–D, representing the yellow line in (b). (g) Line profile for $^{24}\text{Mg}^{2+}$ along traverse C–D, representing the yellow line in (c).

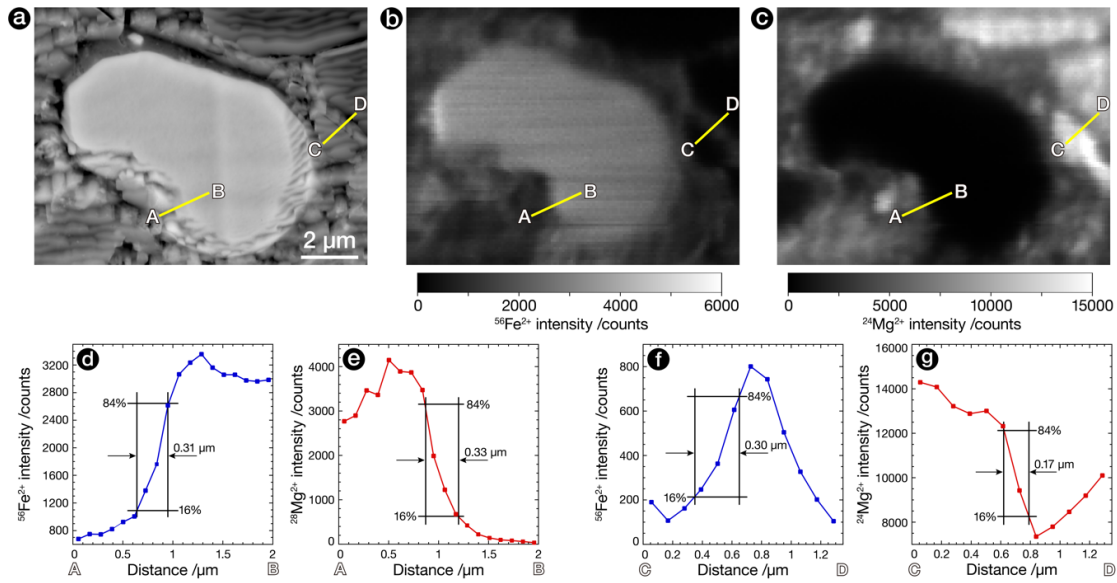


Figure S5. (a) BSE, (b) $^{56}\text{Fe}^{2+}$ ion, and (c) $^{24}\text{Mg}^{2+}$ ion images of Area 5. (d) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (b). (e) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (c). (f) Line profile for $^{56}\text{Fe}^{2+}$ along traverse C–D, representing the yellow line in (b). (g) Line profile for $^{24}\text{Mg}^{2+}$ along traverse C–D, representing the yellow line in (c).

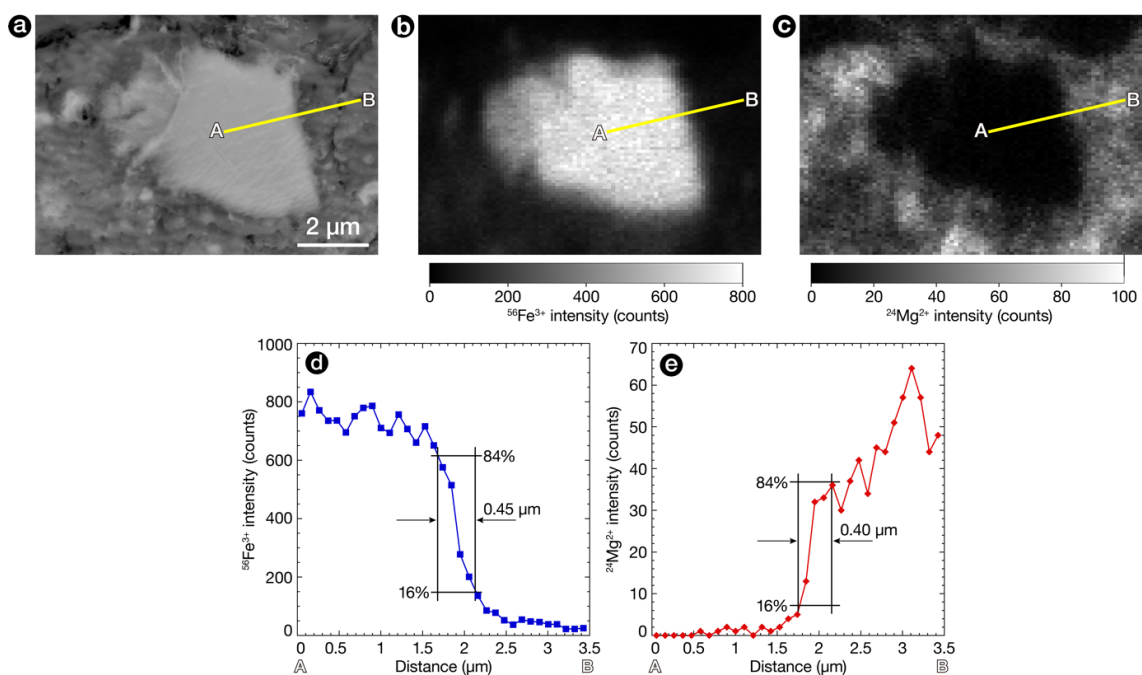


Figure S6. (a) BSE, (b) $^{56}\text{Fe}^{2+}$ ion, and (c) $^{24}\text{Mg}^{2+}$ ion images of Area 6. (d) Line profile for $^{56}\text{Fe}^{3+}$ along traverse A–B, representing the yellow line in (b). (e) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (c).

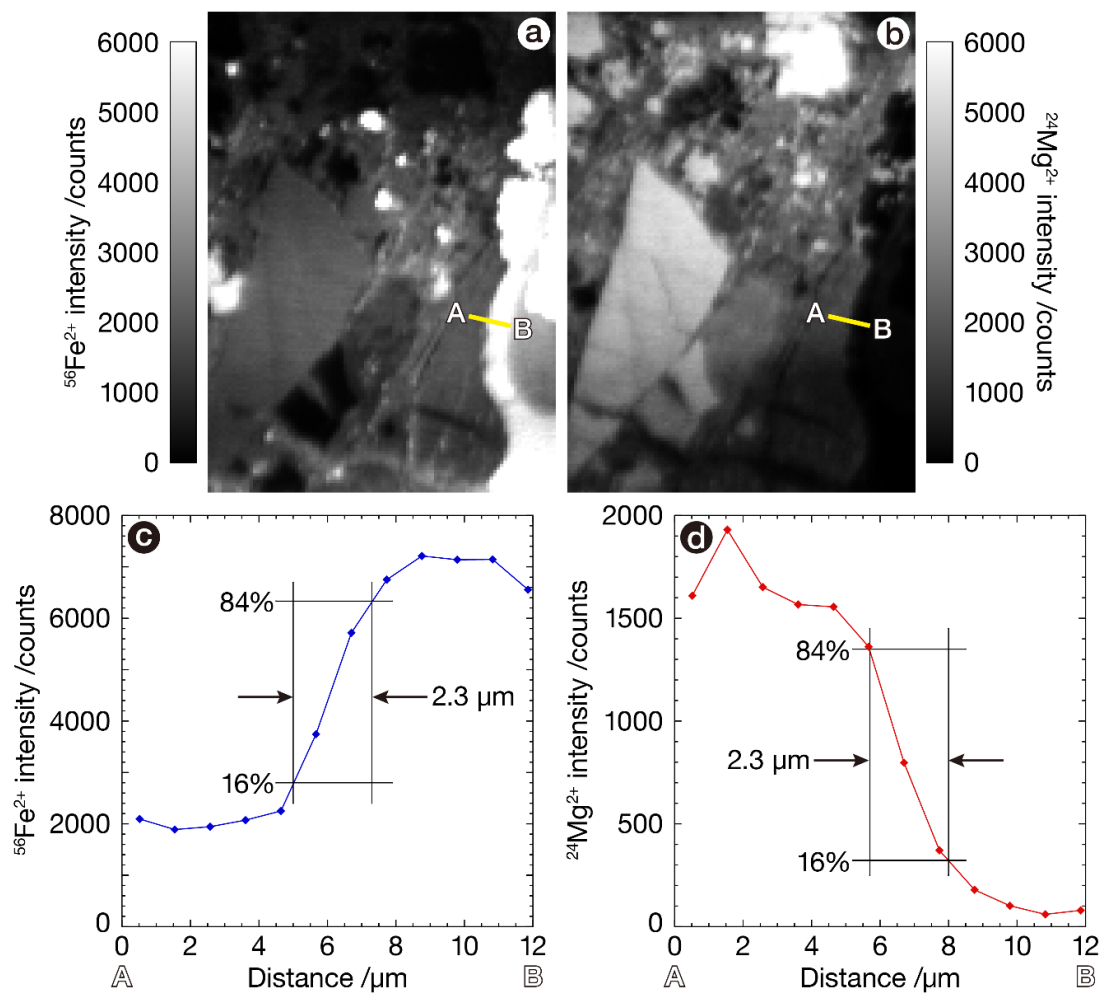


Figure S7. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 1. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

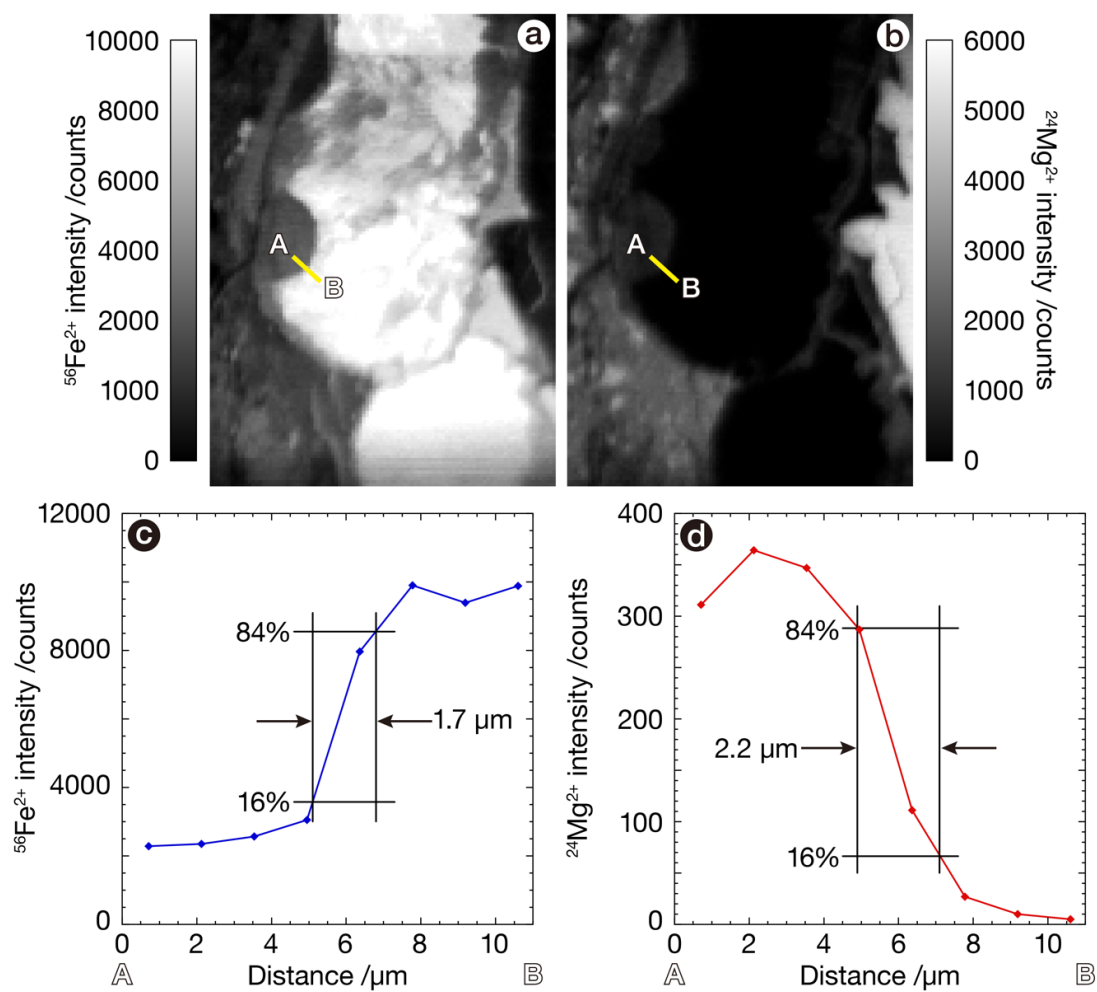


Figure S8. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 2. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

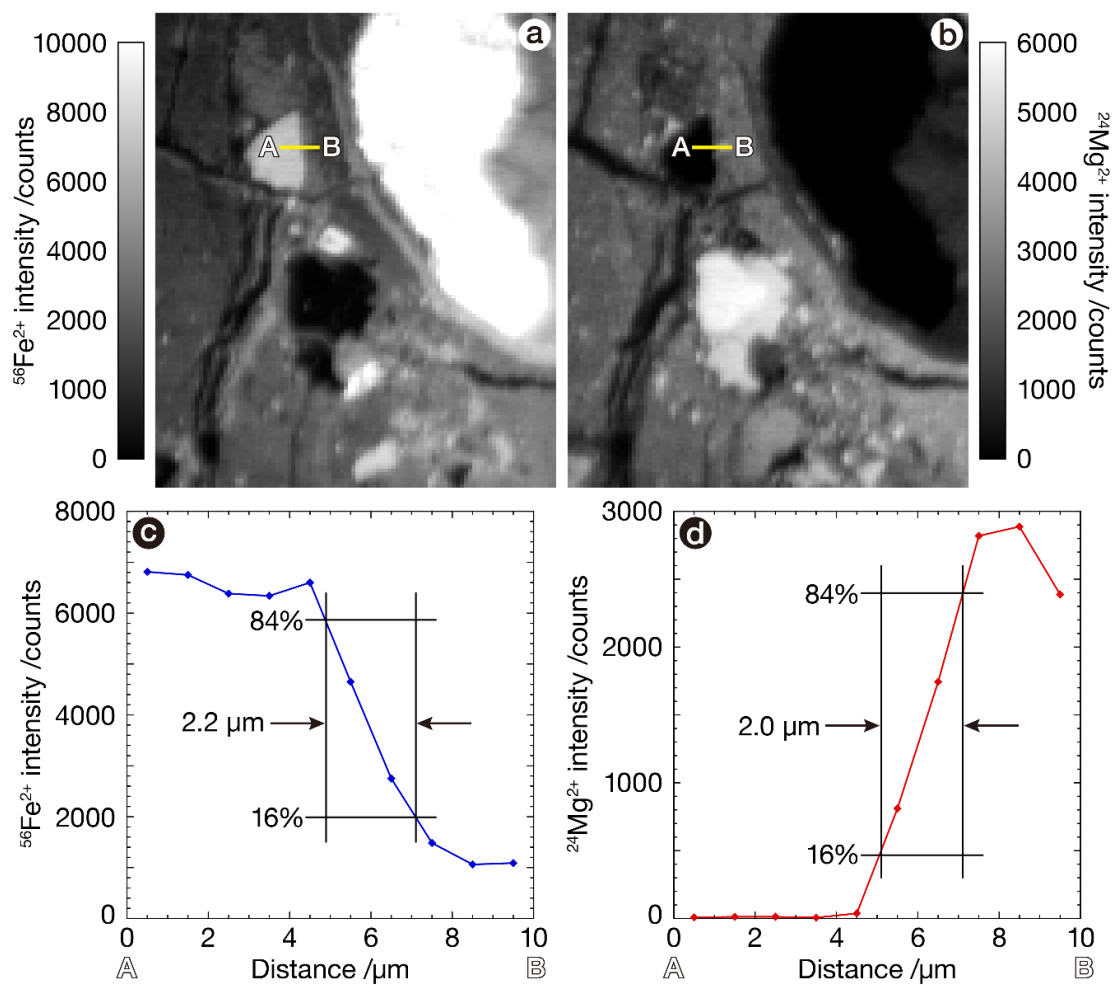


Figure S9. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 3. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

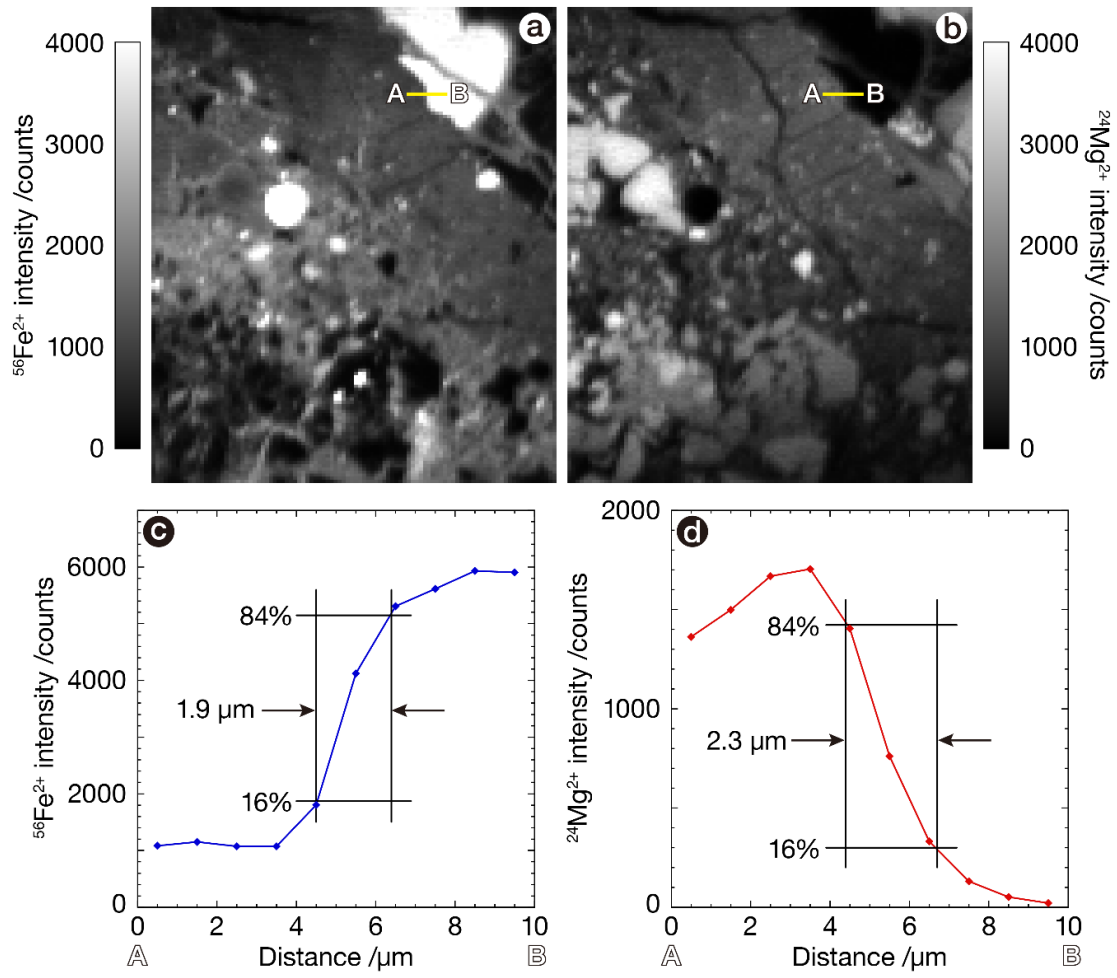


Figure S10. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 4. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

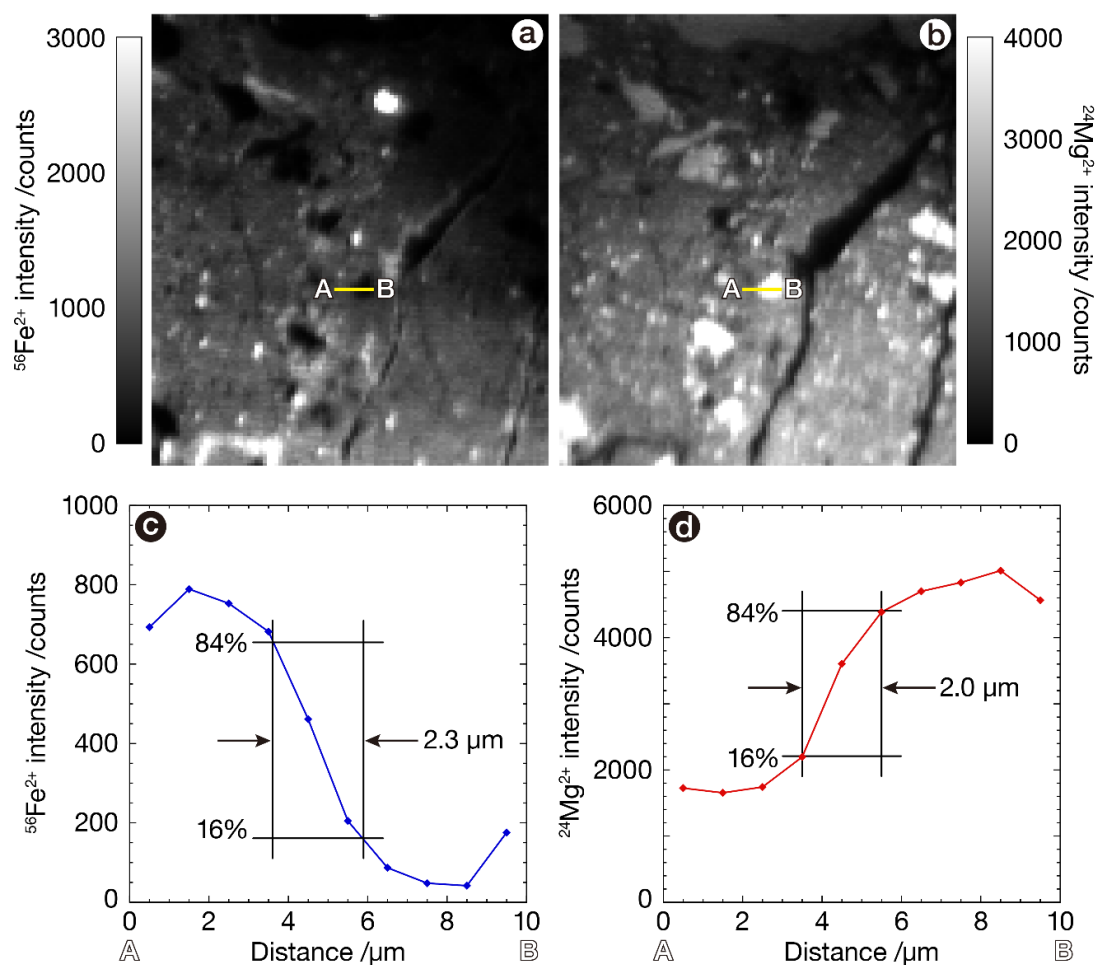


Figure S11. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 5. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

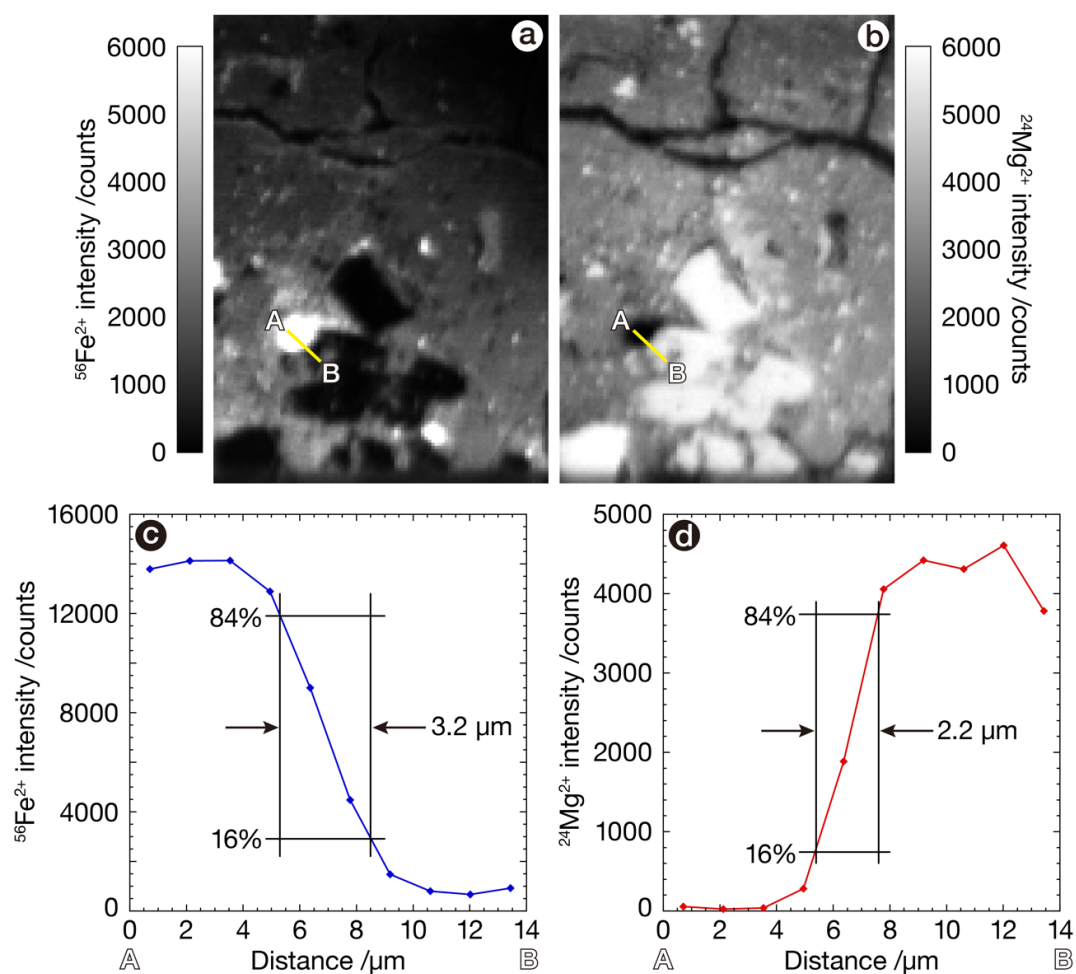


Figure S12. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 6. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

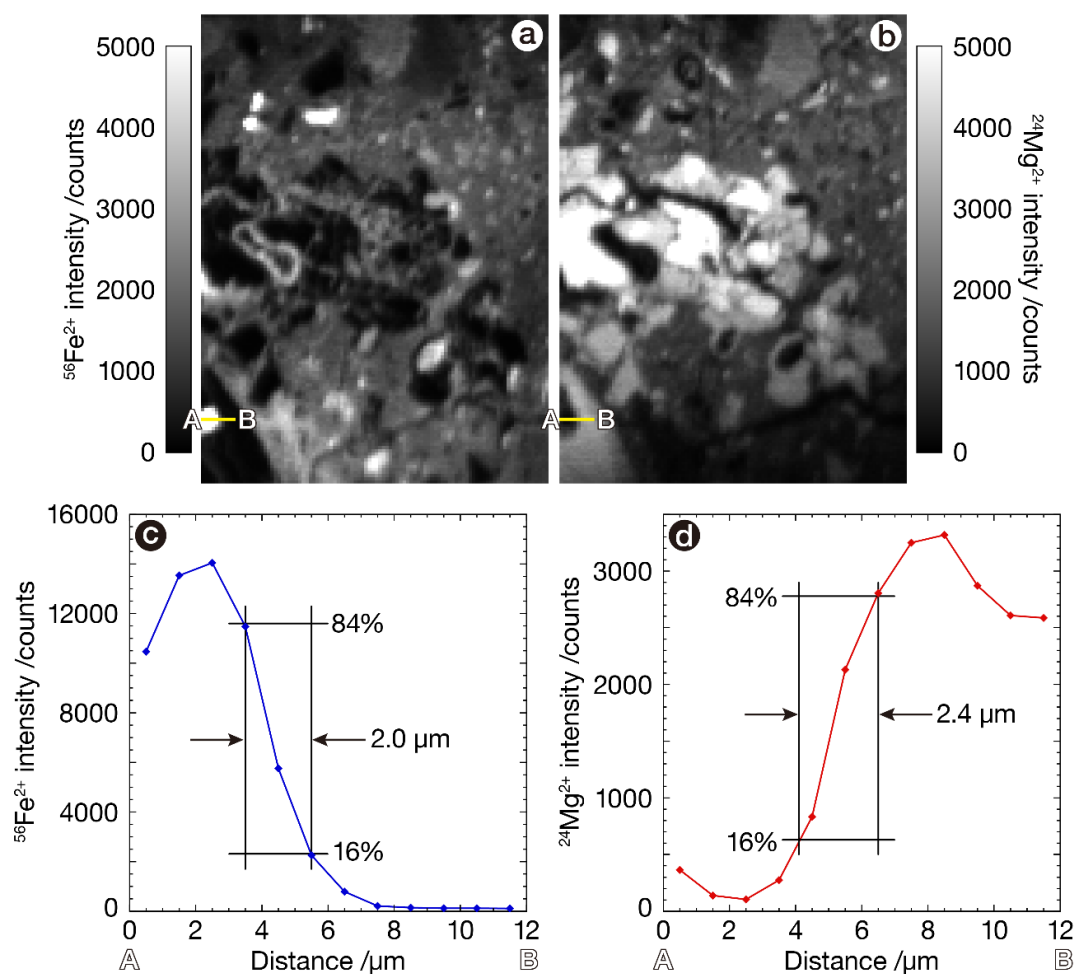


Figure S13. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 7. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

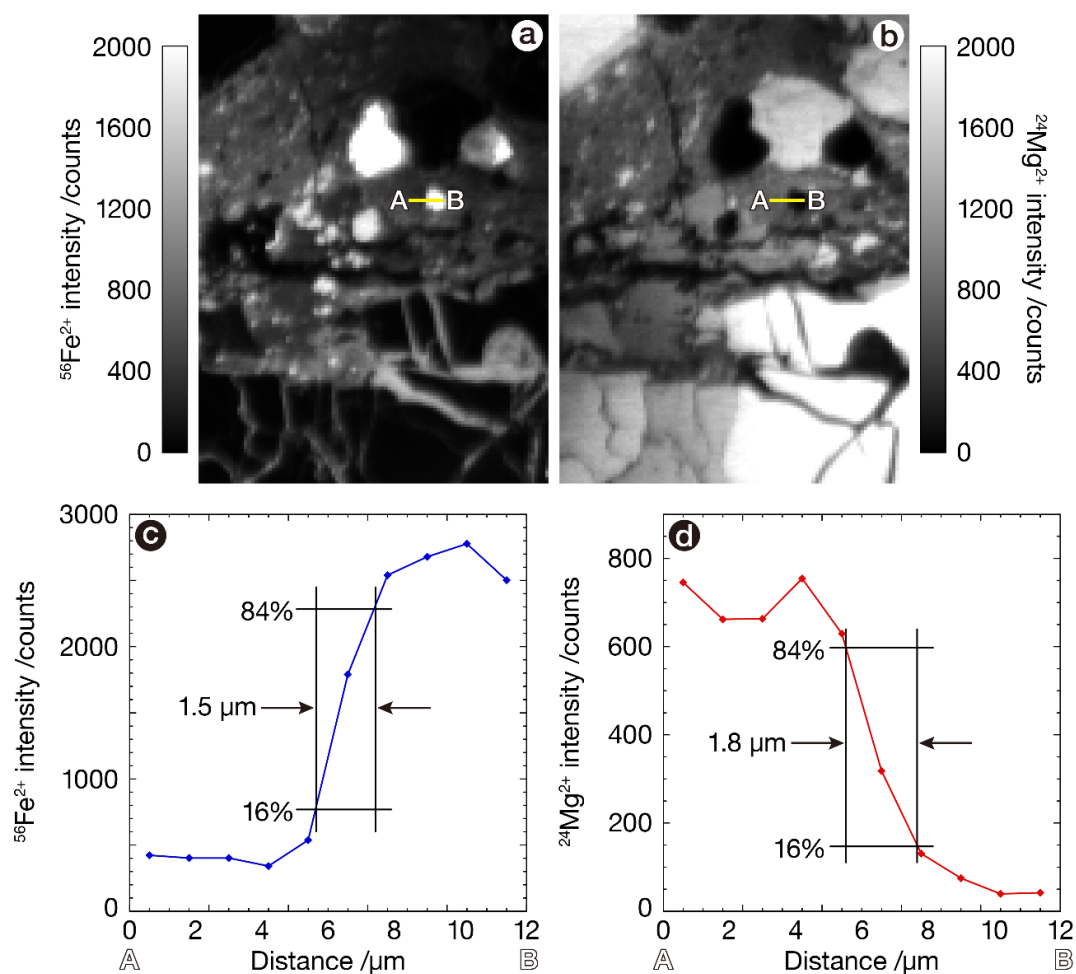


Figure S14. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 8. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

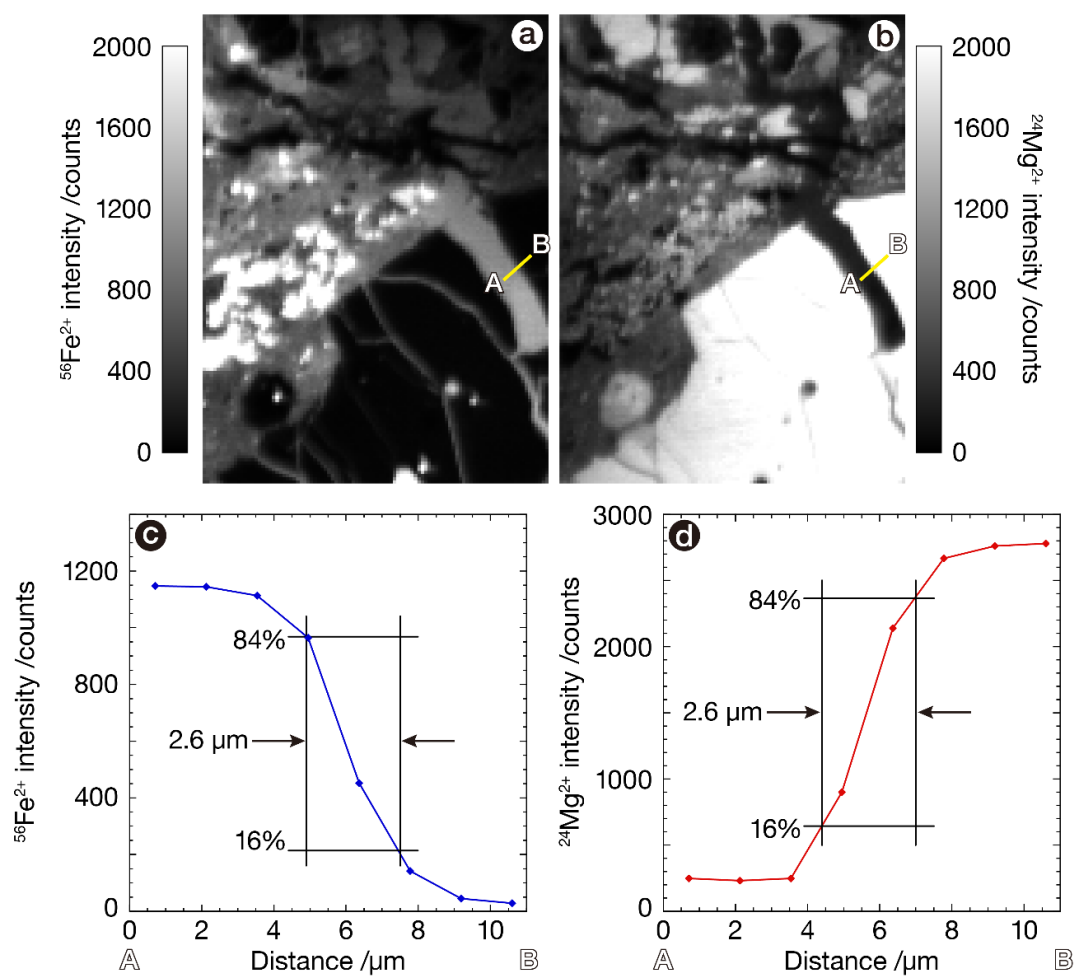


Figure S15. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 9. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

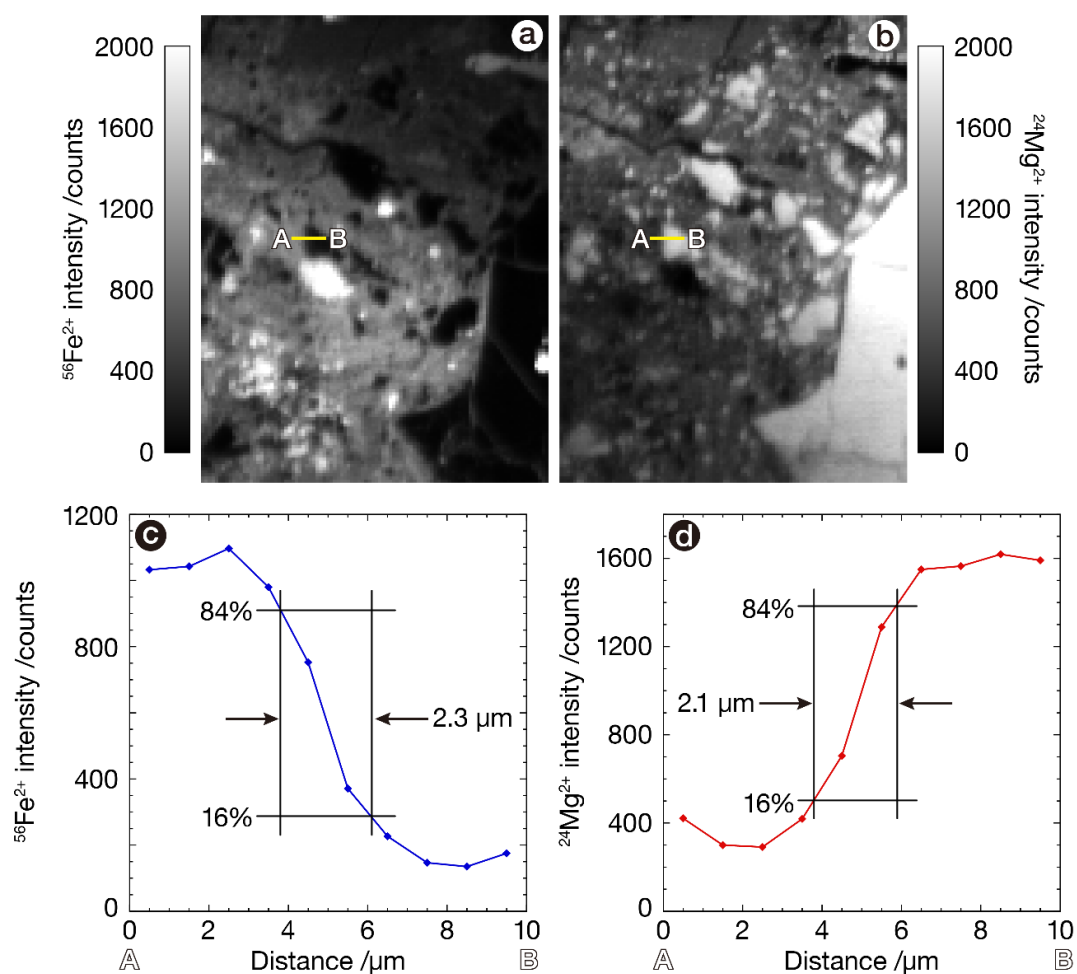


Figure S16. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 10. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

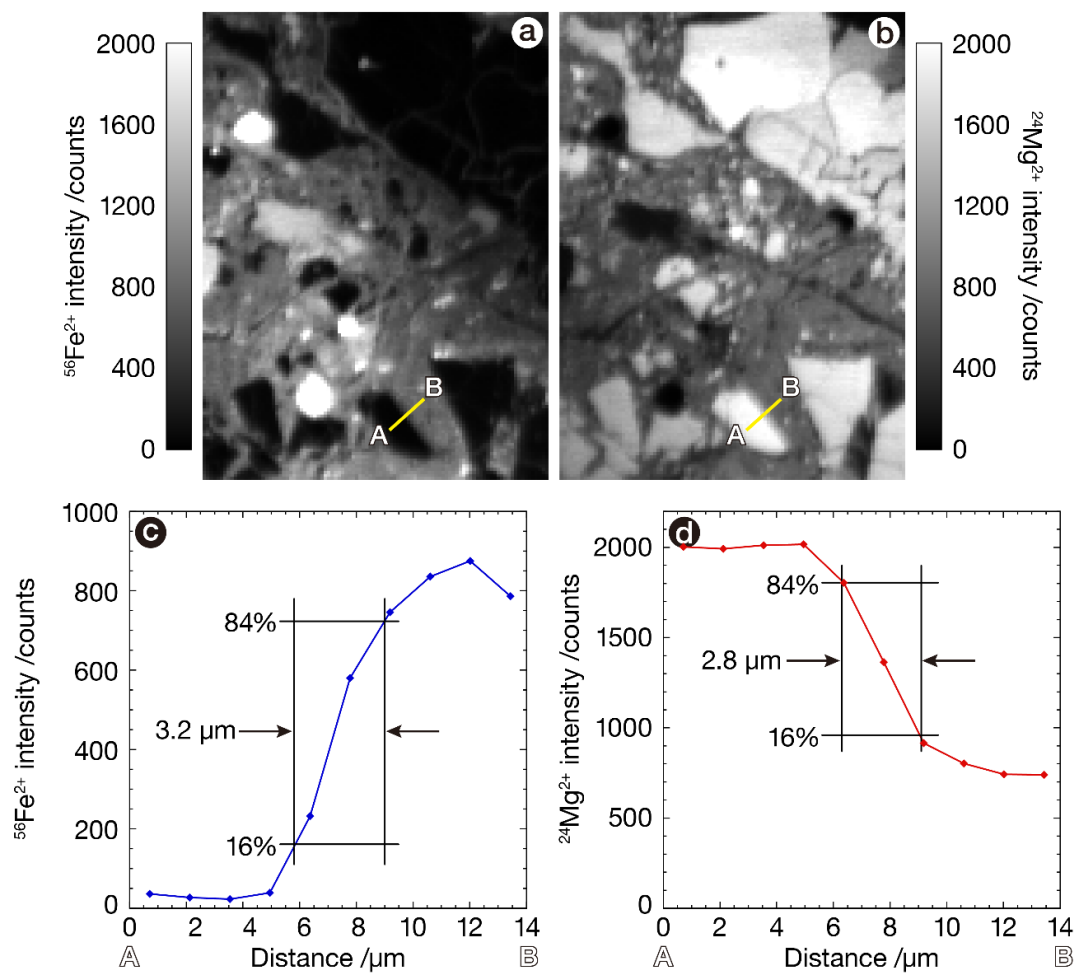


Figure S17. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 11. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

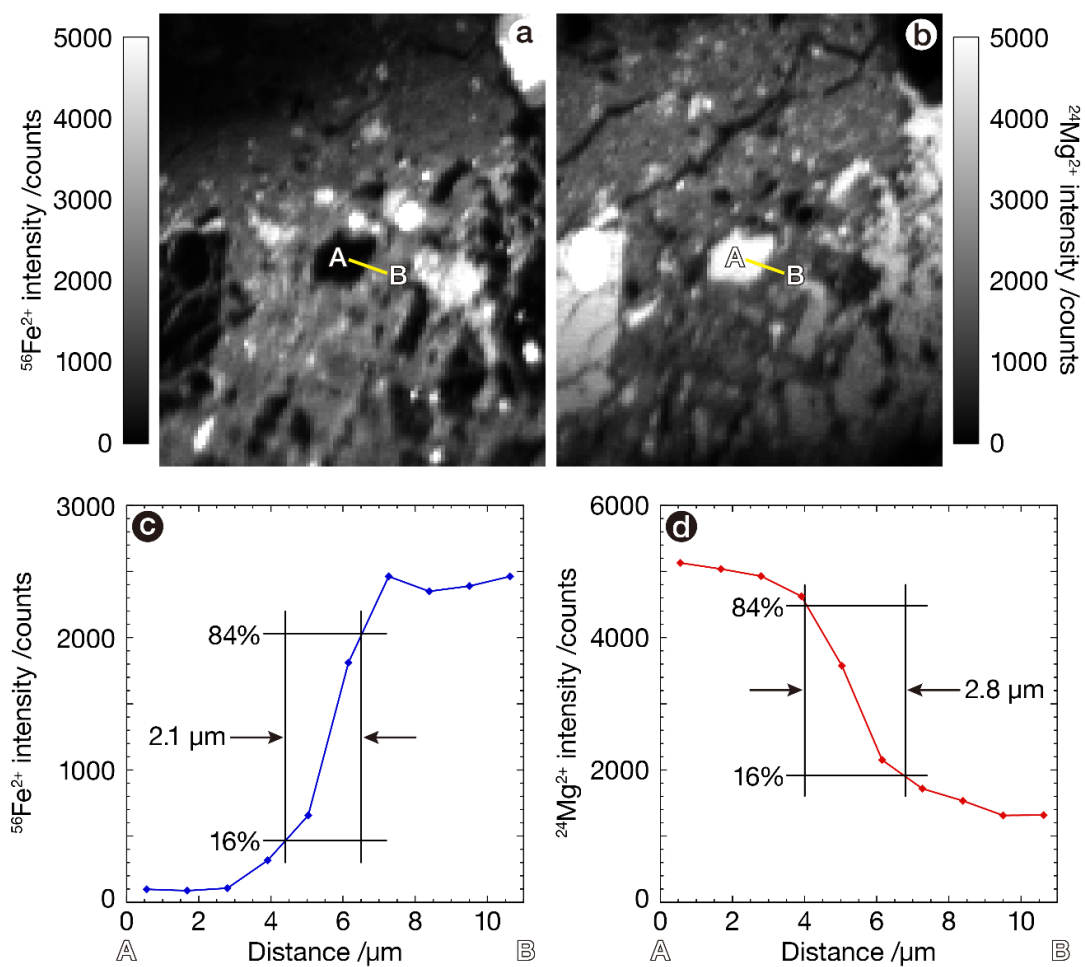


Figure S18. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 12. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

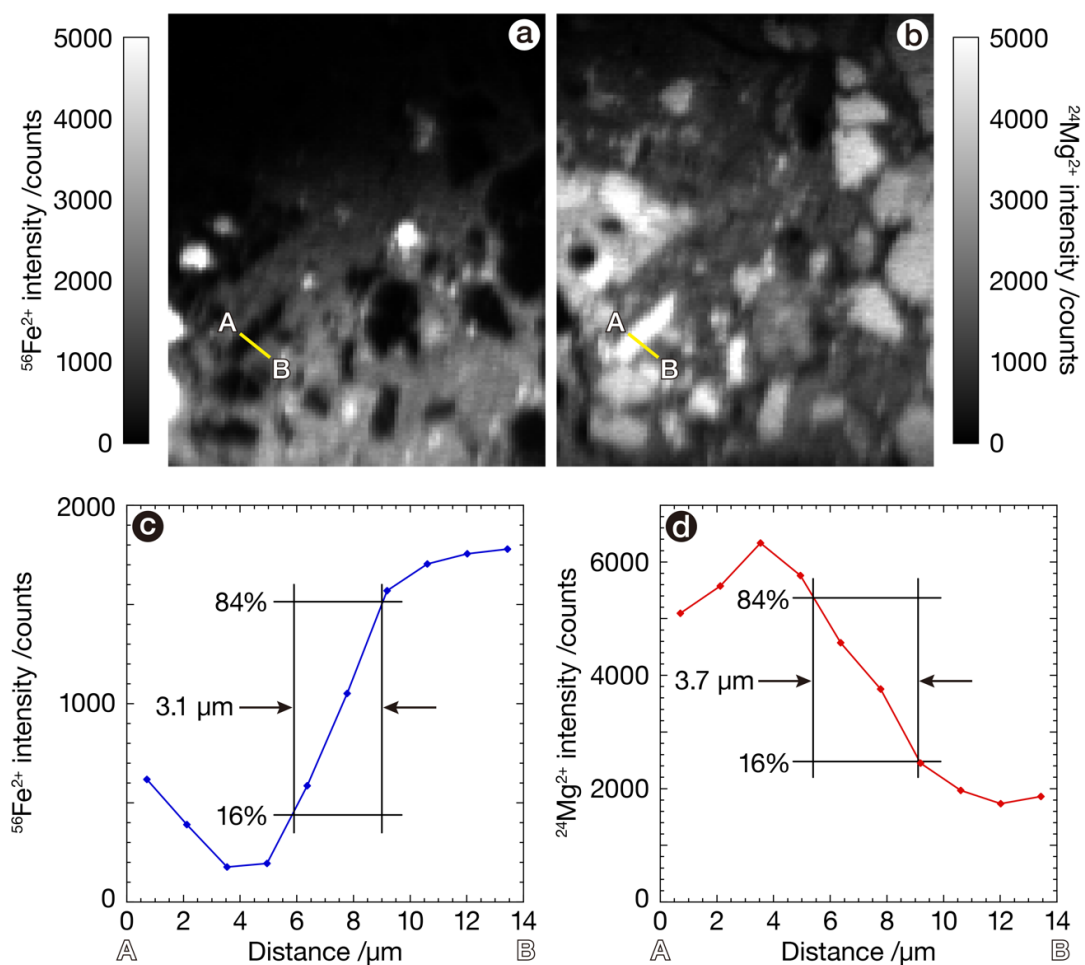


Figure S19. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 13. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

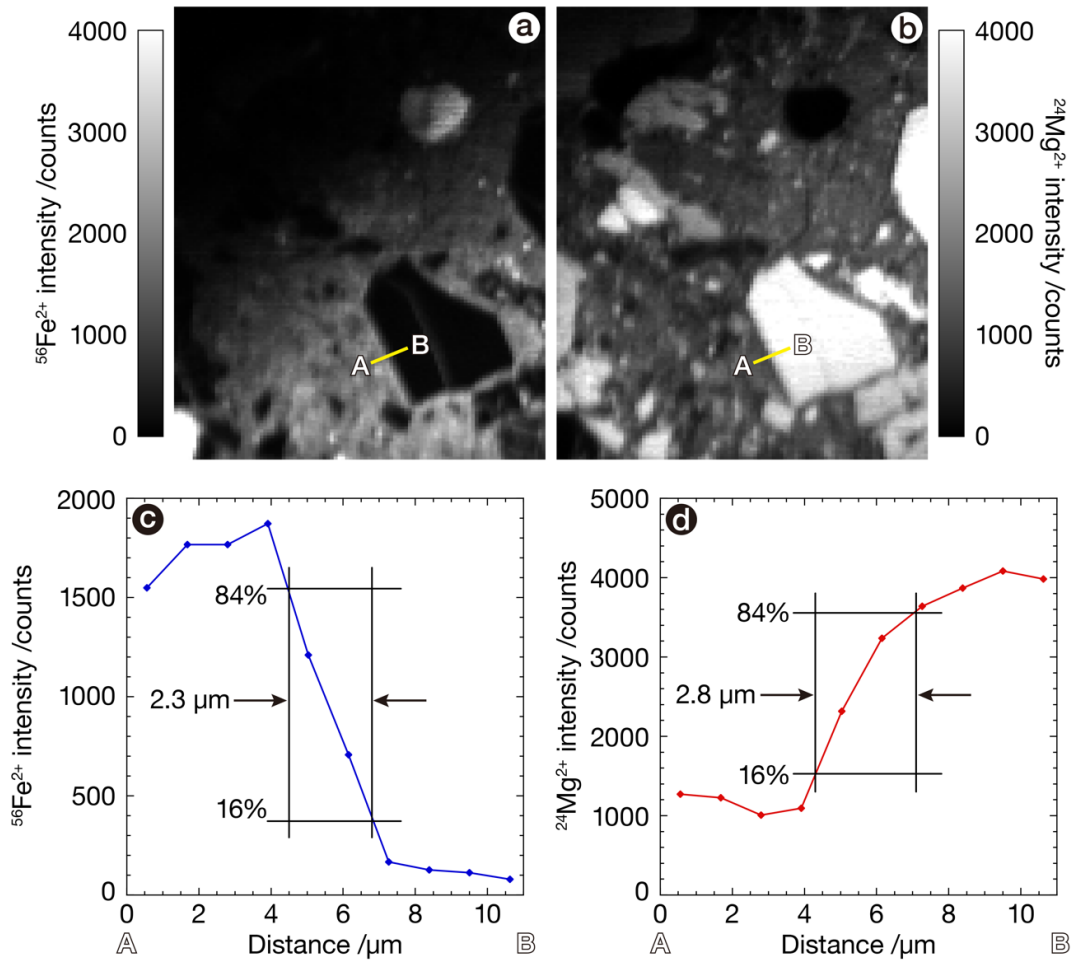


Figure S20. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 14. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

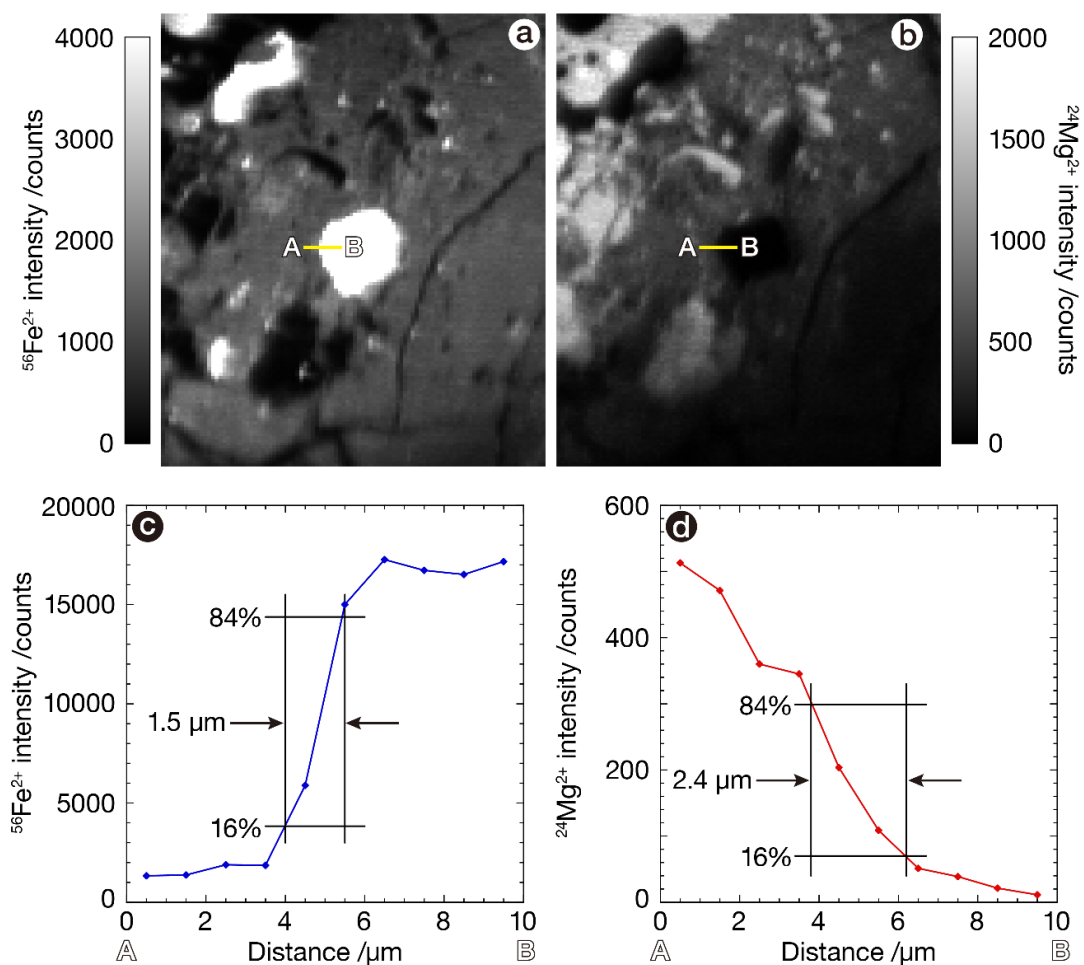


Figure S21. (a) $^{56}\text{Fe}^{2+}$ ion, and (b) $^{24}\text{Mg}^{2+}$ ion images of Area 15. (c) Line profile for $^{56}\text{Fe}^{2+}$ along traverse A–B, representing the yellow line in (a). (d) Line profile for $^{24}\text{Mg}^{2+}$ along traverse A–B, representing the yellow line in (b).

Table S1.

Mineral	Grain size	Grain number	$^4\text{He}^+$ counts	$^4\text{He}^+$ cpms	^4He concentrations / 10^{20} atoms cm^{-3}
Troilite	1–5 μm	01-0318-01-tro1-5um-01	3.0	0.015	0.7
		01-0318-01-tro1-5um-02	9.0	0.045	2.1
		01-0318-01-tro1-5um-03	18.8	0.094	4.3
		01-0318-01-tro1-5um-04	9.1	0.046	2.1
		01-0318-01-tro1-5um-05	3.5	0.018	0.8
		01-0318-01-tro1-5um-06	57.7	0.288	13.2
		01-0318-01-tro1-5um-07	7.5	0.038	1.7
		01-0318-01-tro1-5um-08	7.0	0.035	1.6
		01-0318-01-tro1-5um-09	2.0	0.010	0.5
		01-0318-01-tro1-5um-10	3.0	0.015	0.7
		01-0318-01-tro1-5um-11	4.5	0.023	1.0
		01-0318-01-tro1-5um-12	6.7	0.034	1.5
		01-0318-01-tro1-5um-13	6.0	0.030	1.4
		01-0318-01-tro1-5um-14	7.5	0.038	1.7
		01-0318-01-tro1-5um-15	7.0	0.035	1.6
		01-0318-01-tro1-5um-16	58.0	0.290	13.3
		01-0318-01-tro1-5um-17	2.0	0.010	0.5
		01-0318-01-tro1-5um-18	25.0	0.125	5.7
		01-0318-01-tro1-5um-19	8.0	0.040	1.8
		01-0318-01-tro1-5um-20	5.0	0.025	1.1
		01-0318-01-tro1-5um-21	28.5	0.143	6.5
		01-0318-01-tro1-5um-22	5.6	0.028	1.3
		01-0318-01-tro1-5um-23	8.0	0.040	1.8
		02-0318-11-tro1-5um-01	2.3	0.012	0.5
		02-0318-11-tro1-5um-02	2.3	0.011	0.5
		02-0318-11-tro1-5um-03	6.0	0.030	1.4
		03-0318-12-tro1-5um-01	5.0	0.025	1.1
		03-0318-12-tro1-5um-02	3.5	0.018	0.8
		03-0318-12-tro1-5um-03	2.7	0.013	0.6
		03-0318-12-tro1-5um-04	7.1	0.036	1.6
		03-0318-12-tro1-5um-05	4.0	0.020	0.9
		03-0318-12-tro1-5um-06	16.5	0.083	3.8
		03-0318-12-tro1-5um-07	12.7	0.063	2.9
		04-0705-01-tro1-5um-01	5.7	0.028	1.3
		04-0705-01-tro1-5um-02	15.6	0.078	3.6
		04-0705-01-tro1-5um-03	5.8	0.029	1.3
		04-0705-01-tro1-5um-04	3.0	0.015	0.7
		04-0705-01-tro1-5um-05	11.6	0.058	2.7
		04-0705-01-tro1-5um-06	10.0	0.050	2.3
		04-0705-01-tro1-5um-07	14.0	0.070	3.2
		04-0705-01-tro1-5um-08	21.3	0.106	4.9
		04-0705-01-tro1-5um-09	7.7	0.038	1.8
		04-0705-01-tro1-5um-10	13.3	0.067	3.1
		05-0318-09-tro1-5um-01	22.0	0.110	5.0
		05-0318-09-tro1-5um-02	79.2	0.396	18.1
		05-0318-09-tro1-5um-03	9.8	0.049	2.2
		05-0318-09-tro1-5um-04	7.0	0.035	1.6
		05-0318-09-tro1-5um-05	8.7	0.043	2.0
		05-0318-09-tro1-5um-06	10.1	0.051	2.3
		05-0318-09-tro1-5um-07	12.3	0.062	2.8
		05-0318-09-tro1-5um-08	57.5	0.288	13.2
		05-0318-09-tro1-5um-09	48.5	0.243	11.1
		05-0318-09-tro1-5um-10	22.0	0.110	5.0

Table S1. (Continued)

Mineral	Grain size	Grain number	$^4\text{He}^+$ counts	$^4\text{He}^+$ cps	^4He concentrations / 10^{20} atoms cm^{-3}
Troilite	1–5 μm	05-0318-09-tro1-5um-11	15.3	0.076	3.5
(Continued)	(Continued)	05-0318-09-tro1-5um-12	3.8	0.019	0.9
		05-0318-09-tro1-5um-13	26.5	0.133	6.1
		05-0318-09-tro1-5um-14	10.0	0.050	2.3
		05-0318-09-tro1-5um-15	4.1	0.021	0.9
		06-0318-08-tro1-5um-01	43.6	0.218	10.0
		06-0318-08-tro1-5um-02	11.4	0.057	2.6
		06-0318-08-tro1-5um-03	17.5	0.088	4.0
		06-0318-08-tro1-5um-04	44.9	0.225	10.3
		06-0318-08-tro1-5um-05	27.3	0.137	6.3
		07-0705-02-tro1-5um-01	4.6	0.023	1.1
		07-0705-02-tro1-5um-02	23.3	0.117	5.3
		07-0705-02-tro1-5um-03	32.5	0.163	7.4
		08-0129-18-tro1-5um-01	2.0	0.010	0.5
		08-0129-18-tro1-5um-02	48.8	0.244	11.2
		08-0129-18-tro1-5um-03	18.8	0.094	4.3
		08-0129-18-tro1-5um-04	7.8	0.039	1.8
		08-0129-18-tro1-5um-05	5.2	0.026	1.2
		08-0129-18-tro1-5um-06	79.3	0.396	18.1
		08-0129-18-tro1-5um-07	16.1	0.081	3.7
		08-0129-18-tro1-5um-08	12.2	0.061	2.8
		09-0129-17-tro1-5um-01	5.0	0.025	1.1
		09-0129-17-tro1-5um-02	27.5	0.138	6.3
		09-0129-17-tro1-5um-03	9.7	0.048	2.2
		09-0129-17-tro1-5um-04	63.9	0.319	14.6
		09-0129-17-tro1-5um-05	10.9	0.054	2.5
		09-0129-17-tro1-5um-06	56.7	0.283	13.0
		09-0129-17-tro1-5um-07	5.8	0.029	1.3
		09-0129-17-tro1-5um-08	2.5	0.013	0.6
		10-0129-16-tro1-5um-01	3.8	0.019	0.9
		10-0129-16-tro1-5um-02	14.6	0.073	3.3
		10-0129-16-tro1-5um-03	72.4	0.362	16.6
		10-0129-16-tro1-5um-04	9.6	0.048	2.2
		10-0129-16-tro1-5um-05	9.4	0.047	2.2
		10-0129-16-tro1-5um-06	14.3	0.072	3.3
		10-0129-16-tro1-5um-07	14.3	0.071	3.3
		10-0129-16-tro1-5um-08	8.1	0.041	1.9
		10-0129-16-tro1-5um-09	22.0	0.110	5.0
		10-0129-16-tro1-5um-10	8.8	0.044	2.0
		10-0129-16-tro1-5um-11	27.7	0.138	6.3
		10-0129-16-tro1-5um-12	18.9	0.094	4.3
		10-0129-16-tro1-5um-13	3.6	0.018	0.8
		10-0129-16-tro1-5um-14	5.9	0.029	1.3
		10-0129-16-tro1-5um-15	11.0	0.055	2.5
		10-0129-16-tro1-5um-16	11.5	0.058	2.6
		10-0129-16-tro1-5um-17	6.8	0.034	1.6
		11-0129-19-tro1-5um-01	14.6	0.073	3.3
		11-0129-19-tro1-5um-02	3.5	0.018	0.8
		11-0129-19-tro1-5um-03	3.0	0.015	0.7
		11-0129-19-tro1-5um-04	8.8	0.044	2.0
		11-0129-19-tro1-5um-05	18.0	0.090	4.1
		11-0129-19-tro1-5um-06	3.8	0.019	0.9
		11-0129-19-tro1-5um-07	54.2	0.271	12.4

Table S1. (Continued)

Mineral	Grain size	Grain number	$^4\text{He}^+$ counts	$^4\text{He}^+$ cpms	^4He concentrations / 10^{20} atoms cm^{-3}
Troilite	1–5 μm	11-0129-19-tro1-5um-08	35.3	0.176	8.1
(Continued)	(Continued)	11-0129-19-tro1-5um-09	4.4	0.022	1.0
		11-0129-19-tro1-5um-10	51.0	0.255	11.7
		11-0129-19-tro1-5um-11	8.6	0.043	2.0
		11-0129-19-tro1-5um-12	38.8	0.194	8.9
		11-0129-19-tro1-5um-13	10.6	0.053	2.4
		11-0129-19-tro1-5um-14	6.5	0.033	1.5
		11-0129-19-tro1-5um-15	7.0	0.035	1.6
		11-0129-19-tro1-5um-16	7.1	0.036	1.6
		11-0129-19-tro1-5um-17	2.4	0.012	0.6
		12-0705-04-tro1-5um-01	21.2	0.106	4.8
		12-0705-04-tro1-5um-02	13.3	0.066	3.0
		12-0705-04-tro1-5um-03	27.0	0.135	6.2
		12-0705-04-tro1-5um-04	51.0	0.255	11.7
		12-0705-04-tro1-5um-05	8.5	0.043	1.9
		12-0705-04-tro1-5um-06	26.1	0.131	6.0
		12-0705-04-tro1-5um-07	13.7	0.068	3.1
		12-0705-04-tro1-5um-08	13.1	0.066	3.0
		12-0705-04-tro1-5um-09	11.8	0.059	2.7
		12-0705-04-tro1-5um-10	10.4	0.052	2.4
		12-0705-04-tro1-5um-11	9.8	0.049	2.2
		12-0705-04-tro1-5um-12	48.0	0.240	11.0
		12-0705-04-tro1-5um-13	5.0	0.025	1.1
		12-0705-04-tro1-5um-14	4.3	0.021	1.0
		12-0705-04-tro1-5um-15	3.5	0.018	0.8
		13-0705-11-tro1-5um-01	26.0	0.130	6.0
		13-0705-11-tro1-5um-02	10.1	0.051	2.3
		13-0705-11-tro1-5um-03	8.8	0.044	2.0
		13-0705-11-tro1-5um-04	7.6	0.038	1.7
		13-0705-11-tro1-5um-05	4.8	0.024	1.1
		13-0705-11-tro1-5um-06	8.5	0.043	1.9
		13-0705-11-tro1-5um-07	17.0	0.085	3.9
		13-0705-11-tro1-5um-08	6.7	0.033	1.5
		13-0705-11-tro1-5um-09	3.2	0.016	0.7
		13-0705-11-tro1-5um-10	22.2	0.111	5.1
		13-0705-11-tro1-5um-11	7.0	0.035	1.6
		13-0705-11-tro1-5um-12	17.7	0.088	4.0
		13-0705-11-tro1-5um-13	3.2	0.016	0.7
		13-0705-11-tro1-5um-14	4.3	0.022	1.0
		13-0705-11-tro1-5um-15	2.9	0.015	0.7
		14-0705-05-tro1-5um-01	4.5	0.022	1.0
		14-0705-05-tro1-5um-02	34.3	0.171	7.8
		14-0705-05-tro1-5um-03	15.0	0.075	3.4
		14-0705-05-tro1-5um-04	11.3	0.057	2.6
		14-0705-05-tro1-5um-05	17.7	0.088	4.0
		14-0705-05-tro1-5um-06	31.5	0.158	7.2
		14-0705-05-tro1-5um-07	18.3	0.091	4.2
		15-0705-07-tro1-5um-01	6.5	0.033	1.5
		15-0705-07-tro1-5um-02	8.8	0.044	2.0
		15-0705-07-tro1-5um-03	29.3	0.147	6.7
	5–10 μm	01-0318-01-tro5-10um-1	141.9	0.710	32.5
		01-0318-01-tro5-10um-2	35.2	0.176	8.1
		01-0318-01-tro5-10um-3	12.7	0.064	2.9

Table S1. (Continued)

Mineral	Grain size	Grain number	$^4\text{He}^+$ counts	$^4\text{He}^+$ cps	^4He concentrations / 10^{20} atoms cm^{-3}
Troilite (Continued)	5–10 μm (Continued)	03-0318-11-tro5-10um-01	7.7	0.039	1.8
		04-0705-01-tro5-10um-01	6.4	0.032	1.5
		06-0318-08-tro5-10um-01	54.2	0.271	12.4
		09-0129-17-tro5-10um-01	4.9	0.024	1.1
		09-0129-17-tro5-10um-02	47.7	0.239	10.9
	10–15 μm	03-0318-11-tro10-15um-01	3.9	0.020	0.9
		07-0705-02-tro10-15um-01	36.7	0.184	8.4
	15–20 μm	05-0318-09-tro15-20um-01	5.5	0.028	1.3
	20–25 μm	03-0318-11-tro20-25um-01	4.1	0.020	0.9
Ferrihydrite	1–5 μm	01-0318-01-fhy1-5um-01	3.7	0.019	0.9
		01-0318-01-fhy1-5um-02	34.0	0.170	7.8
		01-0318-01-fhy1-5um-03	4.0	0.020	0.9
		01-0318-01-fhy1-5um-04	8.0	0.040	1.8
		01-0318-01-fhy1-5um-05	9.0	0.045	2.1
		01-0318-01-fhy1-5um-06	7.0	0.035	1.6
		02-0318-11-fhy1-5um-01	4.0	0.020	0.9
		02-0318-11-fhy1-5um-02	3.4	0.017	0.8
		02-0318-11-fhy1-5um-03	2.0	0.010	0.5
		03-0318-11-fhy1-5um-01	7.3	0.036	1.7
		03-0318-11-fhy1-5um-02	4.2	0.021	1.0
		03-0318-11-fhy1-5um-03	7.7	0.038	1.8
		05-0318-09-fhy1-5um-01	8.8	0.044	2.0
		05-0318-09-fhy1-5um-02	14.9	0.074	3.4
		05-0318-09-fhy1-5um-03	27.3	0.136	6.2
		05-0318-09-fhy1-5um-04	18.0	0.090	4.1
		05-0318-09-fhy1-5um-05	90.8	0.454	20.8
		05-0318-09-fhy1-5um-06	49.3	0.246	11.3
		05-0318-09-fhy1-5um-07	19.3	0.097	4.4
		05-0318-09-fhy1-5um-08	26.4	0.132	6.0
		05-0318-09-fhy1-5um-09	23.6	0.118	5.4
		06-0318-08-fhy1-5um-01	27.1	0.136	6.2
		06-0318-08-fhy1-5um-02	16.7	0.083	3.8
		06-0318-08-fhy1-5um-03	7.2	0.036	1.6
		07-0705-02-fhy1-5um-01	11.2	0.056	2.6
		07-0705-02-fhy1-5um-02	12.5	0.062	2.9
		07-0705-02-fhy1-5um-03	20.3	0.101	4.6
		08-0129-18-fhy1-5um-01	3.4	0.017	0.8
		08-0129-18-fhy1-5um-02	5.9	0.029	1.3
		08-0129-18-fhy1-5um-03	9.0	0.045	2.1
		08-0129-18-fhy1-5um-04	4.0	0.020	0.9
		09-0129-17-fhy1-5um-01	7.9	0.039	1.8
		09-0129-17-fhy1-5um-02	6.1	0.031	1.4
		09-0129-17-fhy1-5um-03	3.0	0.015	0.7
		09-0129-17-fhy1-5um-04	4.0	0.020	0.9
		10-0129-16-fhy1-5um-01	10.0	0.050	2.3
		10-0129-16-fhy1-5um-02	14.0	0.070	3.2
		10-0129-16-fhy1-5um-03	84.2	0.421	19.3
		10-0129-16-fhy1-5um-04	4.3	0.021	1.0
		10-0129-16-fhy1-5um-05	30.6	0.153	7.0
		10-0129-16-fhy1-5um-06	7.8	0.039	1.8
		10-0129-16-fhy1-5um-07	4.9	0.025	1.1
		10-0129-16-fhy1-5um-08	14.3	0.072	3.3
		10-0129-16-fhy1-5um-09	11.0	0.055	2.5

Table S1. (Continued)

Mineral	Grain size	Grain number	$^4\text{He}^+$ counts	$^4\text{He}^+$ cpms	^4He concentrations / 10^{20} atoms cm^{-3}
Ferrihydrite	1–5 μm	11-0129-19-fhy1-5um-01	14.2	0.071	3.3
(Continued)	(Continued)	11-0129-19-fhy1-5um-02	6.2	0.031	1.4
		12-0705-04-fhy1-5um-01	15.7	0.078	3.6
		13-0705-11-fhy1-5um-01	3.9	0.019	0.9
		13-0705-11-fhy1-5um-02	18.6	0.093	4.3
		14-0705-05-fhy1-5um-01	26.7	0.134	6.1
		14-0705-05-fhy1-5um-02	21.0	0.105	4.8
		14-0705-05-fhy1-5um-03	2.8	0.014	0.6
	5–10 μm	04-0705-01-fhy5-10um-01	3.5	0.018	0.8
		11-0129-19-fhy5-10um-01	4.9	0.025	1.1
	15–20 μm	10-0129-16-fhy15-20um-01	9.7	0.048	2.2
Fe-Ni metal	1–5 μm	01-0318-01-met1-5um-01	8.0	0.040	1.8
		03-0318-11-met1-5um-01	5.7	0.028	1.3
		04-0705-01-met1-5um-01	14.4	0.072	3.3
		12-0705-04-met1-5um-01	2.7	0.013	0.6
		12-0705-04-met1-5um-02	3.3	0.016	0.7
	5–10 μm	11-0129-19-met5-10um-01	15.0	0.075	3.4
		12-0705-04-met5-10um-01	16.9	0.085	3.9
		13-0705-11-met5-10um-01	6.5	0.033	1.5
	10–15 μm	04-0705-01-met10-15um-01	5.6	0.028	1.3
	20–25 μm	14-0705-05-met20-25um-01	6.2	0.031	1.4
		15-0705-07-met20-25um-01	6.2	0.031	1.4
Magnetite	1–5 μm	01-0318-01-mag1-5um-01	19.1	0.095	4.4
		01-0318-01-mag1-5um-02	28.2	0.141	6.5
		07-0705-02-mag1-5um-01	9.2	0.046	2.1
Olivine	1–5 μm	01-0318-01-ol-1-5um-01	3.8	0.019	1.9
		01-0318-01-ol-1-5um-02	22.3	0.112	11.5
		01-0318-01-ol-1-5um-03	5.5	0.028	2.8
		01-0318-01-ol-1-5um-04	4.3	0.022	2.2
		01-0318-01-ol-1-5um-05	4.1	0.021	2.1
		01-0318-01-ol-1-5um-06	3.3	0.016	1.7
		02-0318-11-ol-1-5um-01	2.0	0.010	1.0
		02-0318-11-ol-1-5um-02	3.1	0.016	1.6
		04-0705-01-ol1-5um-01	3.2	0.016	1.6
		04-0705-01-ol1-5um-02	3.7	0.019	1.9
		05-0318-09-ol1-5um-01	7.0	0.035	3.6
		05-0318-09-ol1-5um-02	8.8	0.044	4.5
		05-0318-09-ol1-5um-03	3.1	0.016	1.6
		06-0318-08-ol1-5um-01	7.2	0.036	3.7
		09-0129-17-ol1-5um-01	6.9	0.035	3.6
		11-0129-19-Fa1-5um-01	3.8	0.019	0.9
		11-0129-19-Fa1-5um-02	3.7	0.019	0.9
	5–10 μm	01-0318-01-ol-5-10um-01	5.0	0.025	2.6
		01-0318-01-ol-5-10um-02	3.4	0.017	1.7
		02-0318-11-ol-5-10um-01	2.3	0.011	1.2
		02-0318-11-ol-5-10um-02	5.3	0.027	2.7
		02-0318-11-ol-5-10um-03	2.0	0.010	1.0
		02-0318-11-ol-5-10um-04	2.0	0.010	1.0
		04-0705-01-ol5-10um-01	4.6	0.023	2.3
		04-0705-01-Fa5-10um-01	3.7	0.019	1.9
		13-0705-11-ol5-10um-01	4.6	0.023	2.4
		14-0705-05-ol5-10um-01	4.3	0.022	2.2
	10–15 μm	05-0318-01-ol10-15um-01	6.8	0.034	3.5

Table S1. (Continued)

Mineral	Grain size	Grain number	$^4\text{He}^+$ counts	$^4\text{He}^+$ cpms	^4He concentrations / 10^{20} atoms cm^{-3}
Olivine (Continued)	15–20 μm	01-0318-01-ol-15-20um-01	5.9	0.029	3.0
		01-0318-01-ol-15-20um-02	3.8	0.019	2.0
	20–25 μm	11-0129-19-ol20-25um-01	3.8	0.019	1.9
	25–30 μm	01-0318-01-ol-25-30um-01	4.1	0.020	2.1
		06-0318-08-ol25-30um-01	2.9	0.015	1.5
Pyroxene	>30 μm	01-0318-01-Fa-100um-01	3.2	0.016	1.6
		14-0705-05-ol50-55um-01	7.3	0.037	3.8
		14-0705-05-ol50-55um-02	3.5	0.018	1.8
		01-0318-01-px-1-5um-01	4.3	0.021	2.2
	1–5 μm	01-0318-01-px-1-5um-02	3.6	0.018	1.9
		02-0318-11-px-1-5um-01	3.4	0.017	1.8
		02-0318-11-px-1-5um-02	10.0	0.050	5.2
		02-0318-11-px-1-5um-03	3.0	0.015	1.5
		02-0318-11-px-1-5um-04	4.6	0.023	2.4
		03-0318-11-px1-5um-01	3.8	0.019	2.0
		03-0318-11-px1-5um-02	4.6	0.023	2.4
		03-0318-11-px1-5um-03	3.1	0.016	1.6
		04-0705-01-px1-5um-01	2.4	0.012	1.3
		05-0318-09-px1-5um-01	4.8	0.024	2.5
		05-0318-09-px1-5um-02	4.8	0.024	2.4
		05-0318-09-px1-5um-03	5.5	0.028	2.8
		05-0318-09-px1-5um-04	4.7	0.023	2.4
		05-0318-09-px1-5um-05	10.1	0.051	5.2
		05-0318-09-px1-5um-06	9.0	0.045	4.6
		05-0318-09-px1-5um-07	6.7	0.033	3.4
		05-0318-09-px1-5um-08	26.3	0.132	13.6
		05-0318-09-px1-5um-09	12.2	0.061	6.3
		05-0318-09-px1-5um-10	4.3	0.021	2.2
		05-0318-09-px1-5um-11	4.6	0.023	2.4
		05-0318-09-px1-5um-12	5.9	0.029	3.0
		06-0318-08-px1-5um-01	41.5	0.208	21.4
		06-0318-08-px1-5um-02	5.6	0.028	2.9
		06-0318-08-px1-5um-03	6.4	0.032	3.3
		06-0318-08-px1-5um-04	4.7	0.024	2.4
		10-0129-16-px1-5um-01	2.3	0.012	1.2
		10-0129-16-px1-5um-02	4.0	0.020	2.1
		10-0129-16-px1-5um-03	3.6	0.018	1.9
		10-0129-16-px1-5um-04	5.2	0.026	2.7
		11-0129-19-px1-5um-01	6.5	0.033	3.3
		11-0129-19-px1-5um-02	5.5	0.027	2.8
		11-0129-19-px1-5um-03	7.8	0.039	4.0
		11-0129-19-px1-5um-04	3.0	0.015	1.5
		11-0129-19-px1-5um-05	4.9	0.024	2.5
	5–10 μm	01-0318-01-px-5-10um-01	4.4	0.022	2.3
		02-0318-11-px-5-10um-01	4.0	0.020	2.1
		03-0318-11-px10-15um-01	5.0	0.025	2.6
		05-0318-09-px5-10um-01	4.7	0.023	2.4
		05-0318-09-px5-10um-02	3.9	0.020	2.0
		08-0129-18-px5-10um-01	2.7	0.013	1.4
		10-0129-16-px5-10um-01	7.5	0.037	3.9
		10-0129-16-px5-10um-02	3.2	0.016	1.7
	10–15 μm	04-0705-01-px10-15um-01	5.0	0.025	2.6
		05-0318-09-px10-15um-01	4.2	0.021	2.2

Table S1. (Continued)

Mineral	Grain size	Grain number	$^4\text{He}^+$ counts	$^4\text{He}^+$ cpms	^4He concentrations / 10^{20} atoms cm^{-3}
Pyroxene	10–15 μm	05-0318-09-px10-15um-02	4.7	0.024	2.4
(Continued)	(Continued)	10-0129-16-px10-15um-01	4.3	0.021	2.2
		13-0705-11-px10-15um-01	4.0	0.020	2.1
	15–20 μm	09-0129-17-px15-20um-01	2.2	0.011	1.2
		10-0129-16-px15-20um-01	3.6	0.018	1.9
		10-0129-16-px15-20um-02	2.5	0.013	1.3
		13-0705-11-px15-20um-01	3.9	0.019	2.0
		13-0705-11-px15-20um-02	3.8	0.019	1.9
	20–25 μm	05-0318-09-px20-25um-01	3.4	0.017	1.8
		06-0318-08-px25-30um-01	4.8	0.024	2.5
		11-0129-19-px20-25um-01	3.5	0.017	1.8
		13-0705-11-px20-25um-01	3.1	0.015	1.6
Chondrule rim		09-0129-17-chd3-rim	4.3	0.021	2.2
		10-0129-16-chd3-rim	4.2	0.021	2.1

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