Structural and compositional changes of constituents in Murchison CM chondrite by He^+ ion irradiation

Takaaki Noguchi[1], Yuuji Miyake[2], Ryuji Okazaki[2], Takahito Osawa[3], Hiroyuki Serizawa[3], Hikaru Yabuta[4], Tomoki Nakamura[5]

[1]*Faculty of Arts and Science, Kyushu University (tnoguchi@artsci.kyushu-u.ac.jp),* [2]*Department of Earth andPlanetary Science, Kyushu University,* [3]*Japan Atomic Energy Agency,* [4]*Department of Earth Systems Science, Hiroshima University,* [5]*Department of Earth Science, Tohoku University*

Introduction: In 2020, Hayabusa 2 spacecraft will bring back samples from a C-type asteroid (162173) Ryugu to the earth. We will have an opportunity to investigate materials from a C-type asteroid. Because Ryugu is an airless body, it is expected that the surface materials of Ryugu have been affected by solar wind irradiation and micrometeoroid impacts (space weathering). In general, CM chondrites that experienced aqueous alteration are thought to be an analog material of C-type asteroids. In the case of Ryugu, its reflectance spectra are basically featureless but show a drop-off at <0.45 µm, suggestive of the presence of hydrated minerals with a certain degree of thermal alteration, and hence the best analog material of Ryugu is thermally altered CM chondrites [1].

Because reflectance spectra contain the information of the uppermost surface (on the orders of the wavelength of light) of the materials on celestial bodies, it is not necessarily easy to distinguish homogeneous partially dehydrated CM chondrite-like material, space weathered (in other word, thinly coated by anhydrous material) CM chondrite-like material, and fine-grained mixture of dehydrated and hydrated CM chondrite-like materials. In reality, most CM chondrites are regolith breccias and contain solar gases and abundant subangular mineral and lithic fragments (e.g. [2], [3], [4] and references therein). However, although solar noble gases are restricted to the clastic matrix [2], [3], textural and mineralogical changes related to the solar wind irradiation and/or micrometeoroid impacts have not been identified among CM chondrites yet.

In this study, we try to understand what happens on CM chondrites during low energy solar wind irradiation (1 keV/nucleon), which may lead to the discovery of space weathering products in CM chondrites and to will serve to understand the samples returned from Ryugu. As compared with many spectroscopic studies of CM chondrites (e.g. [5]), few studies are focused on the modifications of texture, mineralogy, and organic materials related to the micrometeoroid impacts and solar wind irradiation on CM chondrites (e.g. [6], [7]). We try to perform spectrum measurements, field-emission scanning electron microscope (FE-SEM), transmission electron microscopy (TEM) observation, C Kα X-ray absorption near-edge structure (XANES) of the irradiated samples.

Materials and Methods: Two chips of Murchison CM chondrite were lapped by using emery papers to obtain roughly flat surfaces without lubricants at Nuclear Science Research Center, Japan Atomic Energy Agency (JAEA). They were irradiated by 4 keV He^+ ions at Takasaki Advanced Radiation Research Institute, JAEA. The doses of He^+ ion beams were 5×10^{16} and 5×10^{17} He^+/cm^2, respectively. These values correspond to the He^+ ion doses in solar wind when Ryugu is exposed to solar wind at 1.1 AU (the averaged orbital radius of Ryugu) for $\sim 10^2$ and $\sim 10^3$ years, respectively. It is highly likely that H^+ that occupy ~96% of positive ions in solar wind particles [8] plays an important role on space weathering. However, we did not perform serial irradiation of H^+ and He^+ due to the functional limitation of the facility. After irradiation, reflectance spectra of the irradiated surface were measured at JASCO Co. Ltd. by using JASCO V-670 absorption spectrometer with an integrating sphere and by using JASCO MSV-5200 UV-VIS-NIR micro-spectrometer. After spectral measurements, each sample was cut by a wire-saw cutter without lubricants. These samples were observed by JEOL JEM-7600F field-emission scanning electron microscope (FE-SEM) at JAEA as well as JEOL JEM-7100F FE-SEM at Kyushu University. We observed the samples by using 2 or 3 kV acceleration voltage to avoid structural changes during observation. A pair of samples irradiated by 10^{16} and 10^{17} He^+ doses (hereafter, we call them M16 and M17 samples, respectively) was coated by C then coated by Pt. Another pair was coated by Os. From the C and Pt coated samples, thin (~150 nm thick) sections were prepared by using scanning electron microscope-focused ion beam sample preparation machine JEOL JIB-4501 at Kyushu University. These thin sections were further process by low acceleration voltage Ar milling machine Fischione model 1050 NanoMill at Kyushu University. They were used for observation and analysis of the irradiated surface by using transmission electron microscope (TEM) FEI Tecnai-20F at Kyushu University. Chemical compositions of phases in the samples were measured by energy dispersive spectrometer (EDS) equipped on FEI Tecnai-20F. For quantitative analysis, many mineral standards were used to determine k factors. Similarly, thin sections were prepared from the Os coated samples. One of them was used to measure C Kα XANES of carbonaceous material in the irradiated surface. Scanning transmission X-ray microscope (STXM) at BL 5.3.2.2 at the Advanced Light Source, Lawrence Berkeley National Laboratory, USA was used to measure the CKα XANES.

Results: Both reflectance spectra of the bulk samples by using an integrating sphere and the averaged reflectance spectra of 5 areas of the fine-grained matrix do not show remarkable changes after 5 × 10^{16} He^+ irradiation. By contrast, a broad absorption from 0.7 to 1.4 µm, related to the absorption by Fe-rich serpentine group minerals, is disappeared after 5 × 10^{17} He^+ irradiation. FE-SEM observation found no remarkable surface morphological changes in the M16 sample. However, TEM observation of the M16 samples revealed that the uppermost surface of the matrix (~20 nm thick) was amorphous. In the case of the uppermost surface of an olivine crystal in a chondrule, there are a ~50-nm amorphous layer on the top and a ~25-nm damaged olivine showing a contrast due to the concentration of shear strain. Therefore, these thin surface modifications could not be detected by reflectance spectra. By contrast, surfaces of both matrix and chondrules in the M17 sample show abundant dome-shaped swells (length <200 nm) formed on the surface, which are confirmed to be radiation blistering based on the TEM observation (Fig. 1). In the uppermost amorphous layers in the matrix, nanocrystals showing 0.20 nm lattice fringes were observed. Their spacing suggests that they are nanophase Fe^0. On the other hand, in the M17 samples, remarkable blisters were observed in the surfaces of both matrix and chondrules (Fig. 1). The thickness of the surface modification ranges from ~50 to ~100 nm (including vesicles). Below the heavily vesiculated amorphous surface layers, lattice fringes of minerals (serpentine and tochlinite in the case of matrix, olivine and low-Ca pyroxene in the case of chondrules) were observed. The heavily vesiculated surface layers on the matrix contain abundant nanoparticles showing 0.20-nm and 0.25-nm lattice fringes, suggestive of the presence of both nanophase Fe^0 and Fe oxide. Chemical compositions of the surface modification layers on the matrix in the M17 samples tend to be more homogeneous than the original matrix minerals and slightly depleted in Mg. By contrast, those on the ferromagnesian silicates in chondrules are considerably depleted in Mg relative to Si and Fe (up to ~70%) compared to the original silicates.

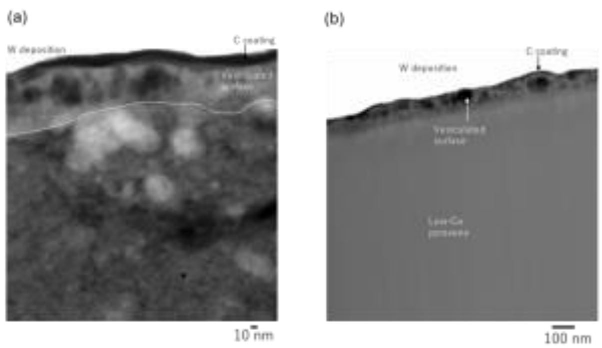

Figure 1. HAADF-STEM images of surface modification in (a) fine-grained matrix and (b) low-Ca pyroxene in a chondrule in Murchison CM chondrite after 5 × 10^{17} He^+ irradiation. Remarkable blistering is readily recognized on the surfaces.

Discussion: $Helium^+$ irradiation with 10^{17} dose changes the shape of reflectance spectra, which is similar to the effect by dehydration at 600 ºC [4]. Therefore, ~1000-year solar wind irradiation seems to cause spectral changes indistinguishable from those caused by 600-ºC heating and subsequent dehydrated by micrometeoroid impacts. Our observations suggest that we could distinguish thermal alterations by shock and micrometeoroid impacts and ion irradiation based on texture and mineralogy including formation of nanophase Fe^0 and Fe oxide and compositions of the uppermost surfaces. Although a previous study [5] reported nanophase Fe^0, we observation identified both nanophase Fe^0 and Fe oxide. The differences may be related to the differences of irradiated ions between [5] and our study, because [5] irradiated H^+ along with He^+ ions, which might the irradiated surface in a more reduced condition. Remarkable Mg depletion in anhydrous ferromagnesian silicates irradiated to low energy ions was reported if the samples were exposed to humid air (70% relative humidity, 30.5 ºC) for 20.8 days although depletion mechanism is unclear [8]. Although we have not exposed to humid air, it is likely that H_2O molecules and/or OH^- would have been released from the fine-grained matrix during He^+ irradiation, which may have serve to deplete Mg from the surface of ferromagnesian silicates. Identification and investigation of Mg depleted materials in CM chondrites may lead to the understanding of space weathering of C-type asteroids.

References

[1] D. Perna et al. 2017. Astron. Astrophys. 599, L1. [2] Nakamura T. et al. (1999a) Geochim. Cosmochim. Acta 63, 241-255. [3] Nakamura T. et al. (1999b) Geochim. Cosmochim. Acta 63, 257-273. [4] Krot A. et al. In: Meteorites and the early solar system II, pp. 679-712. [5] Hiroi T. et al. (1993) Science 261, 1016-1018. [6] Matsuoka M. et al. (2015) Icarus 254, 135-143. [7] Keller L. P. et al. (2015) LPSC 46, Abstract #1913. [8] Dukes and Baragiola. 2011. MRS Proceedings, 1298: mrsf10-1298-r04-05.