Non-Destructive Elemental Analysis of Carbonaceous Chondrites with High-Intensity Muon Beam.

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Introduction: The muon is a lepton with a mass of 105.7 MeV/c^2, approximately 200 times heavier than the electron. So far, electron-induced characteristic X-ray analysis has been widely used to determine chemical compositions of materials in Earth and Planetary Science. In recent years, analysis of characteristic X-rays from muonic atoms, in which a muon is captured, has attracted attention because both a muon beam and a muon-induced characteristic X-ray have high transmission abilities, of which energies are about 200 times higher (e.g., muonic carbon-Kα is 75keV, whereas electron-induced carbon-Kα is 0.3 keV)[1, 2]. It is known that muonic X-ray analysis has great advantages in several ways; (1) non-destructive elemental analysis from light to heavy elements, (2) depth profile analysis, (3) isotopic measurement for heavy elements and (4) investigation of chemical condition (redox-state). Such a non-destructive muonic X-ray analysis has a great potential to characterize precious extraterrestrial samples returned by spacecrafts such as Hayabusa2 and OSIRIS-REx in 2020's.

Results: ***Non-destructive bulk elemental analysis of carbonaceous chondrite.*** A 3 cm × 3 cm × 0.6 cm chip of Jbilet Winselwan meteorite was prepared for the bulk elemental analysis at the MuSIC facility (Fig. 1).

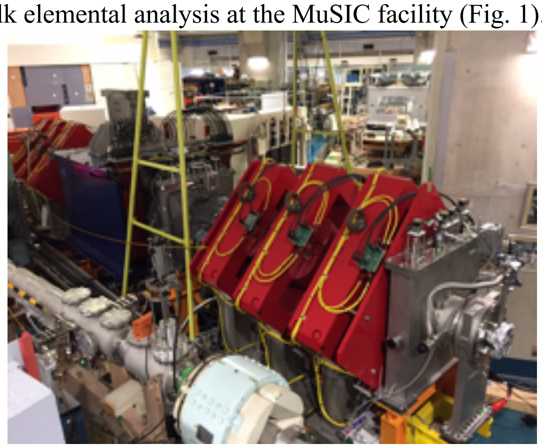

Fig.1. Photograph of Direct Muon beam line

The meteorite chip was exposed to the muon beam with the momentum of 60 MeV/*c* for about 20 hours, and a muonic X-ray spectrum emitting out from the ~3-mm depth of the sample was obtained with a high-purity germanium detector. A comparison of the muonic X-ray spectrum from Jbilet Winselwan with the background spectrum (Fig. 2) shows clear detection of muonic X-rays of Mg, C, Si, O and Fe and marginal detection of those of Ca and S from the meteorite sample. We especially note that the C-K_α signal at 75 keV from 2 wt% of carbon in the sample is clearly distinct from that of Si-L_α at 77 keV. We also note that peaks for Al, Sn, and N were mainly from a sample holder, a masking shield, and the atmosphere, respectively.

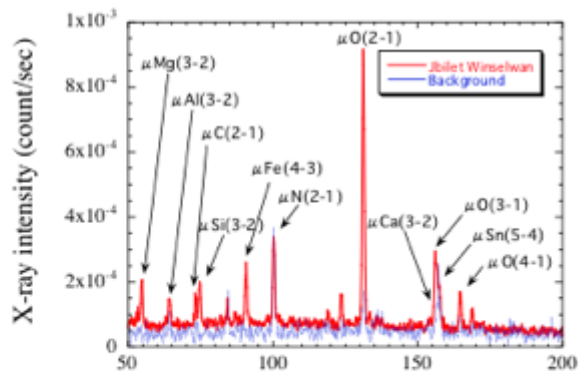

Fig.2. X-ray spectra of Jbilet Winselwan (CM2)

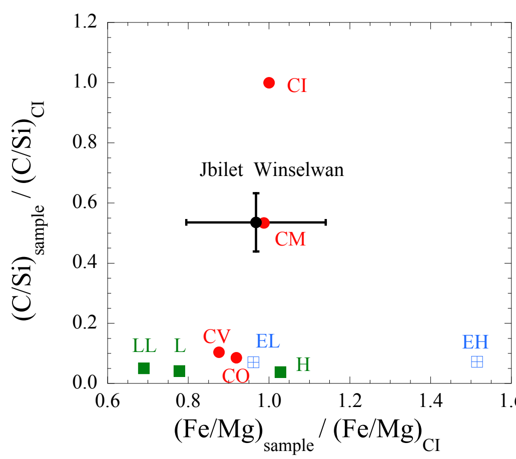

Fig.3. Comparison of CI-normalized C/Si and Fe/Mg ratios of Jbilet Winselwan with those of different chemical groups of chondrites

The depth profiling of light elements from a layered sample. The muon beam analysis of a four-layered sample consisting of SiO_2 glass, graphite (C), boron nitride (BN), and SiO_2 glass was generated to obtain a depth profile of light elements. The sample is held by an Al holder in an Al vacuum chamber with a beam-entrance window composed of polyimide foil.

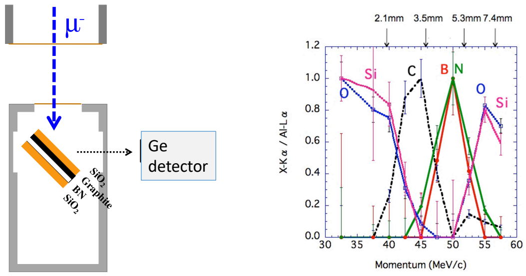

Fig.4: Geometry of the experiment and a depth-profile analysis of the four layered samples

The negative muon beam was collimated to approximately 2.7 cm in diameter and focused on the sample surface (50 mm x 75 mm), which was oriented at 45 degrees to the beam (Fig. 4). Adjusting the muon momentum from 32.5 MeV/c to 57.5 MeV/c, muonic X-rays from the sample were measured by a Ge detector. The accumulation time of X-ray signals was 3-4 hours for each muon momentum. The muonic X-ray intensities of B, C, N, O, and Si from the layered sample are plotted against the muon momentum in Fig.3. The muonic X-ray intensity of each element is normalized to the intensity of Al-K_α at each momentum and to its maximum count along the depth profile. Analytical uncertainties (1s) are estimated from the counting statistics of X-ray counts because the contributions of uncertainties in X-ray detection efficiency are much smaller than those of counting statistics (typically less than 5%). Thus, we successfully demonstrated that the non-destructive depth profile of light elements was successfully obtained up to ~7 mm in depth with a depth resolution of sub-mm. This result is hardly possible with other analytical techniques such as neutron activation analysis, X-ray fluorescence spectroscopy and electron probe microanalysis.

We also attempted to measure much smaller amounts of meteorite samples inside glass tubes to simulate non-destructive analyses of future return samples. Sealing extraterrestrial samples inside glass tubes was originally planned for samples from the asteroid Itokawa. Although Itokawa particles were not sealed in glass tubes due to their small sizes, sealing in a glass tube is one of the effective ways to avoid terrestrial contamination of organic materials and volatiles and thus could be used in future sample return missions. Powdered Murchison meteorite (610 mg) was placed in a 5-cm-long SiO_2 glass tube, in which the inner and outer diameters were 4 mm and 6 mm, respectively. The muon beam collimated to approximately 2.5 cm in diameter, and the apparent cross section of the sample was 4 mm x 25 mm. After exposure of the muon beam with the momentum of 37 MeV/c for approximately 24 hours, clear signals of Mg and marginally resolved signals of Fe were detected through the 1-mm thick glass wall (Fig. 4). Although O and Si are the major elements of rock samples, muonic X-rays of O and Si were emitted from the SiO_2 glass tube as well, which cannot be distinguished from the sample signals in this preliminary study. Although further developments in analytical techniques are required, such as detector setting and collimation of the incident muon beam, our first attempt to non-destructively measure an extraterrestrial sample inside a glass tube succeeded with the detection of Mg and Fe.

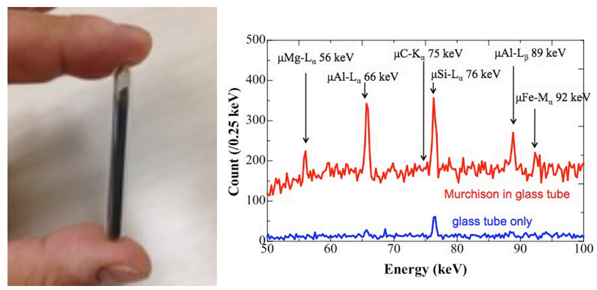

Fig.5: Muonic X-ray spectra from the powdered Murchison meteorite in an SiO_2 glass tube.

Discussion and Remarks: This study demonstrated that non-destructive elemental analysis of a carbonaceous chondrite with the muon beam can detect muonic X-rays from carbon at the 3-mm depth of the sample and that the semi-quantitatively estimated carbon abundance is consistent with that previously reported [3]. The on-going asteroidal sample return missions (Hayabusa2 and OSIRIS-REx) will collect millimeter- to centimeter-sized samples at near-Earth C-type and B-type asteroids Ryugu and Bennu [4, 5] and will return the samples in 2020 and 2023, respectively. The present study proved that the bulk elemental analysis with a muon beam could be a powerful non-destructive analytical technique to compare the returned samples with known chemical groups of chondrites and to determine the carbon content in the samples.

References: [1] Terada, K. *et al.* (2014) *Sci. Rep.* **4**, 5072. [2] Terada, K. *et al.* (2017) *Sci. Rep.* **7**, 15478. [3] Grady, M. M., *et al.* (2014) *LPI Contribution* **#1800**, id.5377. [4] Tachibana, S. *et al.* (2014) *Geochemical Journal* **48**, 571-587. [5] Lauretta, D. S. *et al. (2015) Meteoritics & Planetary Science* **50**, 834-849.