Redox environment estimation of Ryugu's parent body by analysing valence of multiple redox-sensitive elements

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Inside the parent body of asteroid Ryugu, it is estimated that water ice melted due to the decay of radioactive nuclide, creating an alkaline and reducing aqueous environment through reactions with reducing materials such as metallic iron (Nakamura et al., 2023; Yokoyama et al., 2023). It is possible that organic compounds synthesized abiotically in environments like Ryugu's parent body may have contributed to the synthesis of organic matter and origins of lives on Earth today, making it important to estimate the aqueous environment in which these compounds were formed.

To constrain the Eh environment of Ryugu's parent body, we prepared powdered samples, resin-embedded and polished samples, and bulk samples of the Ryugu samples brought by Hayabusa2 mission (A0477, C0120, C0289) and the CI chondrite Orgueil meteorite. For the resin-embedded samples of Ryugu, whole processes from handling to their measurement were conducted under an anaerobic condition to prevent oxidation of the samples. X-ray Absorption Near Edge Structure (XANES) of chromium (Cr), titanium (Ti), vanadium (V), arsenic (As), and iron (Fe) in phyllosilicate region, and sulfur (S), which is mainly present as pyrrhotite in Ryugu, were measured at the following synchrotron radiation facilities: Photon Factory BL-9A, BL-12C, and BL-15A at High-Energy Accelerator Research Organizatio, Japan (bulk measurement and analysis in 20µm spatial resolution); BL17SU and BL-37XU at SPring-8, Japan (µ-XRF-XAFS analysis); and PetraIII P-06 at DESY, Germany (µ-XRF-XAFS analysis with image-stack method).

The aqueous environment during water-rock alteration on Ryugu's parent body was estimated by comparing the analysed valences for the elements shown above with the thermodynamically calculated Eh-pH diagrams. In addition, the results from the Orgueil meteorite and that of Ryugu were compared to examine the changes in the valence states of various elements caused by terrestrial atmospheric oxidation.

The valence of Cr in the Ryugu samples was determined by μ -XRF-XAFS analysis, showing that Cr(III) predominated in the samples, though Cr(II) was also observed in some areas. The latter is considered to be an olivine relict, suggesting a reducing environment in the parent body during water-rock alteration. Titanium in Ryugu sample was found to be distributed between two phases, ilmenite enrichment spots and phyllosilicates, with Ti(IV) predominating in both. Arsenic was distributed in the phyllosilicate matrix and phosphate spot. As(III) and As(V) were observed in the former, and arsenides in the latter. Vanadium was uniformly distributed as V(III) in the whole Ryugu sample texture. XANES spectra from bulk and local analyses were found to be consistent. Analysis of the V valence based on the height of the pre-edge peak (Sutton et al., 2005) revealed that V in Ryugu is mainly V(III), which is a more reduced state than the V(V) observed in the Orgueil meteorite.

From these results, we infer that the distribution and valence of Cr, Ti, and As are constrained by the rock texture of the samples. Particularly, the presence of As(V) observed in phyllosilicates of Ryugu sample appears to contradict the previous research and V(III) observed in this work. This As state in phyllosilicates is thought to be constrained by the structure of phyllosilicate such as substitution of Si(IV) by As(V) (Takahashi et al., 2003; Hattori et al., 2005). Instead, information on the water-rock alteration environment of Ryugu's parent body is recorded in the V state found throughout the samples, supporting previous studies suggesting that the alteration environment on Ryugu's parent body was highly reducing.

References: Hattori K. et al. (2005) Occurrence of arsenic (V) in forearc mantle serpentinites based on X-ray absorption spectroscopy study. *Geochim Cosmochim Acta* **69**, 5585-5596.; Nakamura T. et al. (2023) Formation and evolution of carbonaceous asteroid Ryugu: Direct evidence from returned samples. *Science* **379**.; Sutton S. R. et al. (2005) Vanadium K edge XANES of synthetic and natural basaltic glasses and application to microscale oxygen barometry. *Geochim Cosmochim Acta* **69**, 2333-2348.; Takahashi Y. et al. (2003) Introduction Determination of the As(III)/As(V) Ratio in Soil by X-ray Absorption Near-edge Structure (XANES) and Its Application to the Arsenic Distribution between Soil and Water. *Analytical Sciences* **19** 891-896.; Yokoyama T. et al. (2023) Samples returned from the asteroid Ryugu are similar to Ivuna-type carbonaceous meteorites. *Science* **379**.