Redox conditions during aqueous alteration of CM chondrites: A combined study of Fe-K\(\alpha\) XANES and Fe Mössbauer spectroscopy

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Introduction: In CM chondrites, anhydrous phases such as olivine and pyroxene, Fe-Ni metal, Fe-Ni sulfide, and chondrules glass are replaced by hydrous phases, serpentine and tochilinite (e.g. [1] and references therein). Temperatures during aqueous alteration have been estimated to be from \(\sim 0^\circ\)C to \(<170^\circ\)C [2]. However, compared with temperatures, redox conditions during aqueous alteration have not been understood well. Fe\(^{3+}\)/ΣFe ratio of cronstedtite (Fe\(^2+\)\(\times\)Fe\(^{3+}\)\(\times\)(Fe\(^{3+}\)\(\times\)Si\(^2\))O\(_{10}\)(OH)\(_8\) in matrix and chondrule rims were measured by TEM-EELS [3]. Two Fe K\(\alpha\) XANES studies measured Fe\(^{3+}\)/ΣFe ratios of matrices and serpentine in CM chondrites, respectively [4, 5]. We performed a combined study of Fe K\(\alpha\) XANES and \(^{57}\)Fe Mössbauer spectroscopy to estimate Fe\(^{3+}\)/ΣFe ratios of serpentine in CM chondrites more quantitatively.

Samples and methods: We used cronstedtite from two different localities (Aude, France and Bohemia, Czech). \(^{57}\)Fe Mössbauer spectroscopy was measured in a room temperature by using 370 MBq \(^{57}\)Co in Pd as a source at Shimane University. Forty and 200 mg aliquots of the samples were used as absorbers. Thin sections of Mighei, Bells, Murray, El Quess Ab Said, Nogoya, Sayama, LEW85311, Murchison, Y75293, Essebi, Cold Bokkeveld, and Y82042 were used for this study. To determine the degrees of aqueous alteration according to [6], we observed texture and mineralogy by using both an optical microscope and SEM at Ibaraki University. Chemical compositions of the serpentine-group minerals in chondrules were measured by electron microprobe at National Institute of Polar Research. Fe K\(\alpha\) XANES was measured at Beamline 4A, KEK-PF.

Results and discussion: It has been expected that Mg content in phyllosilicates increases as aqueous alteration proceeds [7, 8]. The average Fe\(^*/(\text{Mg+Fe}^*)\) ratio of serpentine in Sayama CM 2.1 was 0.35 and that of LEW 85311 CM 2.1 was 0.73 (Fe\(^* =\text{Fe}^{2+} + \text{Fe}^{3+}\)). However, it is hard to say that average Fe\(^*/(\text{Mg+Fe}^*)\) ratios of serpentine correlate positively with the degrees of aqueous alteration by considering large \(1\) \(\sigma\) of each average value. We calculated Fe\(^{3+}\)/ΣFe ratios of serpentine based on a combination of Fe K\(\alpha\) XANES and \(^{57}\)Fe Mössbauer spectroscopy. The average values of Fe\(^{3+}\)/ΣFe of serpentine increase from 0.57 in LEW 85331 CM 2.1 to 0.71 in Sayama CM 2.6. However, similarly these variations may be statistically insignificant because the average values overlap by considering \(1\) \(\sigma\) of these values. Ratios between integrated and area intensities of (Fe\(^{3+}\)/ΣFe) in each meteorite do not show remarkable differences by considering \(1\) \(\sigma\). These results indicate that the redox state of aqueous alteration in CM chondrite parent body did not change considerably. Because standard deviation of Fe\(^{3+}\)/ΣFe ratios within a meteorite is large, it is suggested that redox states may have been locally heterogeneous. Comparison between Fe\(^*/(\text{Mg+Fe}^*)\) and (Fe\(^{3+} + \text{Al}\))/[(Fe\(^{3+} + \text{Al}\) + (Si + Mg + Fe\(^{2+}\))] ratios revealed that Fe\(^{3+}\) and Al\(^{3+}\) cations in serpentine that replaced mesostasis of chondrules decreased as aqueous alteration proceeded. In contrast, Fe\(^{2+}\) and Mg\(^{2+}\) in the serpentine increased. This result supports the reaction of replacing elements proposed by [8]. Fe\(^{2+}\) and Mg\(^{2+}\) were probably supplied from phenocrysts in chondrules that replaced by serpentine. Fe\(^{3+}\) and Al\(^{3+}\) cations expelled from serpentine that replaced mesostasis were probably incorporated in abundant serpentine that were formed by replacement of
phenocrysts as aqueous alteration proceeded.


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