THE SETTINGS OF AQUEOUS ALTERATION IN THE EARLY SOLAR SYSTEM: AN X-RAY SPECTROMICROSCOPY INVESTIGATION OF THE MURCHISON CM2 CHONDRITE

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Introduction: Many studies have described petrographic evidence for *in situ* aqueous alteration on the asteroid parent bodies of CM chondrites [1]. However, a component of CM chondrites that remains controversial are the fine-grained rims (FGRs) of phyllosilicates surrounding pristine coarse-grained fragments. The textures of the FGRs suggest that they formed through accretion onto their host objects, but it's not clear whether hydration of the dust occurred in a nebula environment [2, 3] or after incorporation into the parent body [4]. We have been exploring the possibility that information associated with the settings of aqueous alteration in the early solar system may be gleaned from differences in the inter- and intra-granular crystal chemistry to be found within the sub-micron mineralogy of FGRs and matrix within the CM chondrites [5]. In doing so we have performed a synchrotron-based spectromicroscopy study on FGRs and matrix within the Murchison CM2 carbonaceous chondrite. We have collected spatially resolved X-ray fluorescence (XRF), X-ray micro-diffraction (μXRD), Fe-X-ray Absorption Near Edge Structure (XANES) spectroscopy and scanning transmission X-ray microscopy (STXM) data at the micro- and nano-scale to characterize the chemistry, mineralogy and Fe oxidation state of FGRs and matrix.

Methodology: Synchrotron experiments were performed at Diamond Light Source, UK. XRF, μXRD and Fe-XANES data were acquired on beamline I18 [6] with a beam spot size of 3 μm and step size of 3 μm. XRF and μXRD maps 90 × 90 μm in size were acquired from both FGRs and matrix. Within these areas Fe-XANES linescans were acquired. STXM experiments were performed on beamline I08 using FIB sections ~80 nm thick extracted from FGRs and matrix. Fe L-edge and Mg and O K-edge XANES stacks were acquired with a pixel size of < 100 nm.

Results: XRF maps reveal a slight decrease in Fe content towards the inner regions of the FGRs. From Fe-XANES we found that the bulk $Fe^{3+}/\Sigma Fe$ of the FGRs is essentially the same as that of the surrounding matrix material, in agreement with [8]. There is, however, a systematic variation in the pre-edge, edge and white line energies across the FGRs that does not exist in the matrix. Analysis of µXRD maps from the same areas that we collected Fe µXANES data shows that the mineral assemblages identified within the FGRs and the matrix correlate well with our in-house bulk analysis of CM chondrites [7, 9]. However, there is no clear correlation between the µXRD and the systematic changes of the Fe µXANES across the FGRs suggesting that the variation may be associated with the crystal-chemistry of the grains within the FGRs.

From STXM imaging, subtle changes in spectral detail for the Fe L- and O K-edges from matrix areas can be attributed to the presence of these elements within oxide and phyllosilicate minerals. Within the matrix the $Fe^{3+}/\Sigma Fe$ ratio is consistently ~0.85-0.90 in both the Fe and Mg phyllosilicates, in agreement with [10] but more oxidized than other reports [e.g. 8]. More spectral variation is observed in the FGRs, with changes in the relative intensities of the main peaks in the Fe L_3 edge suggesting changes in the $Fe^{3+}/\Sigma Fe$ ratio across the FGR. Indeed the Fe μXANES reveals grain-scale variations of $Fe^{3+}/\Sigma Fe$ in the outer FGR ranging between ~0.75 and ~0.90, which reduces to two distinct groupings in the inner FGR with $Fe^{3+}/\Sigma Fe$ ratios of ~0.60 and ~0.80. Furthermore, greater grain-scale chemical variability is observed in the FGR than in the matrix. For example Mg XANES suggests that the Mg is structurally bound within both Fe- and Mg-rich phyllosilicate hosts in the matrix. In the FGR however, the Fe and Mg phyllosilicates are not distinguishable at the 100 nm scale but pristine forsterite is clearly present within the inner part of the FGR. Additionally, Fe-sulphides with oxidised reaction rims are detected in the outer FGR, and patches of intermixed phyllosilicate and nanoscale Fe oxides are evident in the matrix and the outer FGR, but not the inner FGR.

Settings of Aqueous Alteration: Our data show that the composition of FGRs is mineralogically distinct from the matrix, suggesting they were surrounding the chondrules in their nebular sojourn prior to accretion and sampled a different reservoir of dust to matrix. A possible explanation for the data is the incorporation of ice and/or carbonaceous grains in the FGRs which would cause local variability in redox states when these phases reacted on the asteroidal parent body.

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