AQUEOUS ALTERATION OF CARBONACEOUS CHONDRITES: NEW INSIGHTS FROM COMPARATIVE STUDIES OF TWO UNBRECCIATED CM2 CHONDRITES, Y-791198 AND ALH81002.

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Introduction: Carbonaceous chondrites are an important resource for understanding the physical and chemical conditions in the early solar system. In particular, a long-standing question concerns the role of water in the cosmochemical evolution of carbonaceous chondrites. It is well established that extensive hydration of primary nebular phases occurred in the CM and CI chondrites, but the location where this alteration occurred remains controversial. In the CM2 chondrites, hydration formed secondary phases such as serpentine, tochilinite, pentlandite, carbonate and PCP. There are several textural observations which suggest that alteration occurred before the accretion of the final CM parent asteroid, *i.e.* preaccretionary alteration [1,2]. Conversely, there is a significant body of evidence that supports parent-body alteration [3-5].

In order to test these two competing hypotheses further, we studied two CM chondrites, Y-791198 and ALH81002, two meteorites that exhibit widely differing degrees of aqueous alteration. In addition, both meteorites have primary accretionary textures, *i.e.* experienced minimal asteroidal brecciation [1,5]. Brecciation significantly complicates the task of unraveling alteration histories, mixing components that have been altered to different degrees from different locations on the same asteroidal parent body. Alteration in Y-791198 is mostly confined to chondrule mesostases, FeNi metal and fine-grained matrix and rims. In comparison, the primary chondrule silicates in ALH81002 have undergone extensive replacement by secondary hydrous phases [4,6].

This study focuses on compositional and textural relationships between chondrule mesostasis and the associated rim materials. Our hypothesis is: both these components are highly susceptible to aqueous alteration and should be sensitive recorders of the alteration process. For parent body alteration, we expect systematic coupled mineralogical and compositional changes in rims and altered mesostasis, as elemental exchange between these components occurs. Conversely, for preaccretionary alteration, there should be no clear relationships between the rims and mesostases.

Results: 62 type I and 22 type II chondrules were examined by SEM and electron microprobe (EMP) in Y-791198. In addition, 25 type I and 3 type II chondrules were investigated in ALH81002 [4,5].

Fine-grained rim compositions and textures: There are notable differences between Y-791198 and ALH81002 rims; Y-791198 rims are texturally more distinct, showing significantly more fine scale heterogeneity. Large particles (*e.g.* PCP) tend to occur in outer portion of the rims, whereas large particles are more homogeneously distributed in ALH81002. Rims on these two meteorites are also mineralogically distinct. TEM studies [7] reveal that Y-791198 rims are mostly composed of amorphous/ poorly-crystalline materials, while those of ALH81002 consist dominantly of serpentine and sulfides. Phyllosilicates are typically more coarser-grained and more abundant in ALH81002. Two distinct types of regions dominate Y-791198 rims: sulfide-rich and sulfide-poor. The sulfide-rich regions contain a myriad of sulfide particles ranging down to <10 nm in size, while sulfide-rich regions and fine-grained sulfides are much less common in ALH81002 rims. Fine scale textural characteristics of ALH81002 rims are more homogeneous.

Compositionally, fine-grained rim in ALH81002 are richer in Mg, Al, Na, Ca and K, and lower in Fe and S compared to Y-791198. Mg/(Mg+Fe) ratios and S contents most strikingly illustrate the differences between Y-791198 and ALH81002 rims. In Y-791198 rims, Mg/(Mg+Fe) ratios and S contents have Gaussian distributions, while those of ALH-81002 do not, favoring higher Mg/(Mg+Fe) ratios and lower S values. The average Mg/(Mg+Fe) of Y-791198 rims is 0.31 compared with 0.41 for ALH81002, with average S contents of 3.37 and 3.00, respectively.

Chondrule mesostasis compositions: In both chondrites, secondary hydrous phases (serpentines) have completely replaced the chondrule glass in all chondrules studied; we found no evidence of unaltered chondrule glass. There are distinct differences between altered mesostases in the two meteorites. Y-791198 mesostasis alteration products show a continuum in Mg/(Mg+Fe) ratios from 0.2-0.6, whereas two distinct serpentine populations occur in ALH81002, ranging from 0.28-0.38 and 0.48-0.63, respectively. Overall, Y-791198 mesostases have consistently higher Al, Ti and Ca contents compared to ALH81002, but have lower Fe, S and Ni contents.

Compositional relationships between rims and chondrules: Y-791198 altered mesostases show enrichments in Mn, Al, Ti and Ca and depletions in Fe, Mg, K, Na, Ni and S, normalized to Si, relative to the rims. ALH81002 mesostases are slightly enriched in Mg and Mn and depleted in Fe, Ni, S, Al, Ca and Ti, normalized to Si, compared to the rims. Histograms show that Y-791198 rims are more Fe and S-rich than

ALH81002, while the reverse is true of the altered mesostasis. In both meteorites, rims show less variation in composition than mesostasis.

Discussion: Our data show that Y-791198 and ALH81002 differ in characteristics most consistent with progressive aqueous alteration involving elemental exchange between chondrules and matrix, *i.e.* parent body alteration. (1) In Y-791198, EMP analyses show the fine-grained rim Mg/(Mg+Fe) ratios have a Gaussian distribution, indicating that all rims accreted from material that had essentially the same average composition. If different chondrules sampled dust regions with different average compositions, a Gaussian distribution is unlikely. In ALH81002, the distribution is not Gaussian, but skewed to higher Mg/(Mg+Fe) ratios, an effect that would not be expected from a random sample of nebular dust, but more indicative of a process that has modified the compositions of the rims. According to the preaccretionary model, rims accreted onto variably altered chondrules in a turbulent nebula [1]. Because CM chondrites have rather constant bulk compositions, it is reasonable to assume that each CM chondrite sampled the same region of dust, *i.e.* a reservoir with similar compositional properties. Sampling of dust regions with different bulk compositions would lead to significantly variable CM bulk compositions which are not observed. The distinct differences in rim compositions between Y-791198 and ALH81002 implies that modification of rim materials occurred after accretion of rims to the chondrules, *i.e.* during parent-body alteration. (2) Mesostasis compositions also support such a scenario. Y-791198 mesostases show a broad range in Mg/(Mg+Fe) ratios which is very distinct from that in ALH81002, where two compositionally distinct alteration products occur in chondrules. This demonstrates that the alteration histories experienced by chondrules in these two meteorites were distinct. (3) The textural differences between rims in the two meteorites can be best attributed to the effects of advanced alteration. Overall, there is clearly evidence of textural homogenization in ALH81002 rims compared with Y-791198. ALH81002 rims are very similar, regardless of chondrule type; any discernable layers are discontinuous and compositionally very similar to each other. (4) The compositional differences between rims and mesostasis are consistent with closed system elemental exchange between chondrules and rims. Prior to alteration, Mg, Al, Ca, Na and K are all largely concentrated in chondrules, whereas Fe, Ni and S are enriched in the rims. Redistribution of these elements during alteration would tend to equilibrate the elemental concentrations between chondrules and matrix. Assuming that rim-compositions in both meteorites were very similar prior to alteration, our data confirm that redistribution of elements has occurred, as predicted. ALH81002 rims show increases in Mg, Al, Ca, Na and K and depletions in Fe and S, compared to Y-791198. Mesostases show increases in Ni, S and decreases in Al, Ca and Ti. (5) There is an overall equilibration of elemental concentrations with increasing degree of alteration.

It is often assumed that the earliest stage in CM chondrite alteration involved an Fe-rich fluid, caused by alteration of phases such as metal, sulfide and Fe-rich olivines in the matrix [3-5,7]. However, our studies show that this is may not be the case. Y-791198 mesostases are actually more Mg-rich than their associated fine-grained rims. One possible explanation for this effect is the probable misconception that rim mineralogy was dominated by Fe-rich olivine, prior to alteration. TEM studies of Y-791198 rims suggest instead that amorphous material may have been the dominant precursor. Alteration of Y-791198 initially hydrated this amorphous material and drove crystallization of abundant nm-sized sulfides. This process may have stabilized Fe into the matrix as sulfides and actually produced a fluid that was relatively Mg-rich, resulting from partial dissolution of the amorphous material. Thus, the early pulse of fluid was more Mg-rich than assumed by [3-5,7] and it was this fluid that altered glassy mesostases in chondrules. Further aqueous interaction resulted in alteration of FeNi metal grains embedded in the interiors of the chondrules, releasing Fe that is, at least in part, responsible for the formation of Fe-rich serpentines that are present in altered mesostases in ALH81002 and other CM chondrites.

Parent-body vs. preaccretionary alteration: The observed equilibration with higher degrees of alteration, coupled with the exchange of soluble elements between mesostases and fine-grained rims strongly supports the parent-body alteration model. However, the current model [3-5,7] argues that the first pulse of aqueous fluid formed Fe-rich hydrous phases. As seen in the altered mesostasis of Y-791198, the first pulse appears to have been Mg, rather than Fe-rich. A modification to the parent-body alteration model that includes an earlier period of Mg-rich fluid alteration may be necessary.

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