

## Two chondrule groups each with distinctive rims in Murchison recognized by cathodoluminescence

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**Abstract**—Two groups of chondrules in the Murchison CM chondrite, which have previously been identified on the basis of FeO in the chondrule grains, are readily identified from cathodoluminescence (CL) and belong to those of the ordinary chondrite group A and B chondrules of Sears *et al.* (1992a). All chondrules are surrounded by fine-grained rims containing forsterite with bright red CL, but on group A chondrules an outer thin rim grades into a much thicker rim, with a lower density of forsterite grains, which in turn grades into the central chondrule. Group B chondrules have only the thin outer rim with a high density of small forsterite grains. This is the first time an unequivocal correlation has been observed between chondrule rim thickness and the composition of the object on which the rim is located. We suggest that while all objects in the meteorite (group B chondrules, refractory inclusions, mineral and chondrule fragments, clasts) acquired a very thin rim during processing in a wet regolith, the thick rims on group A chondrules were formed by aqueous alteration of precursor metal- and sulfide-rich rims which are a characteristic of group A chondrules in ordinary chondrites.

### INTRODUCTION

The primitive meteorites have many properties in common, one of them being a fine-grained matrix containing abundant finely dispersed forsterite grains. These forsterite grains generate a highly distinctive and ubiquitous red cathodoluminescence (CL; Sears *et al.*, 1989; Steele, 1986, 1989). Since

this CL is readily destroyed by metamorphism, which enables diffusion of Fe into the grains, the phenomenon is restricted to ordinary and CO chondrites of types 3.0 and 3.1 (Sears *et al.*, 1989; 1991). The formation of this forsterite is of considerable interest in itself (Scott *et al.*, 1984; Nagahara, 1984; Alexander *et al.*, 1989), but more important for our present purposes is that the profusion of fine-grained forsterite grains with their bright

red CL provides a new means of examining the wide scale petrography of primitive meteorites and the relationship between their chondrules, rims, and matrix.

Opinions are divided concerning the origin of the matrix of primitive chondrites and the forsterite grains it contains. Some authors argue that both are primary nebular materials which, for the ordinary chondrites at least, suffered little or no alteration on their parent bodies (Scott *et al.*, 1984; Nagahara, 1984). Others suggest that the matrix, even of ordinary chondrites, was derived primarily from chondrules (Alexander *et al.*, 1989) by a variety of processes which may have included aqueous alteration. Aqueous alteration was extremely important for CM chondrites (Tomeoka and Buseck, 1985; McSween, 1987), so it is quite clear that many properties of the matrix, including its abundant forsterite grains, are related to this process.

The chondrules of primitive ordinary and carbonaceous chondrites also show many similarities (Scott and Taylor, 1983), and they often contain fine-grained rims consisting of phyllosilicates, with inclusions of olivine, pyroxene, sulfides, metal, magnetite and calcite (Bunch and Chang, 1980; King and King, 1981; Allen *et al.*, 1980; Metzler *et al.*, 1992). The mineralogy and petrology of the rims is similar to that of the matrix, although they are somewhat finer grained, and perhaps relatively Fe-rich (Zolensky *et al.*, 1988). Sometimes the rims show concentric textural and compositional layering (Metzler *et al.*, 1992). The present study reports results of an examination of the chondrules and rims in Murchison meteorite using their unusual cathodoluminescence properties.

#### EXPERIMENTAL

A mosaic of the CL of a section of Murchison (AMNH 4377-2) was prepared using a Nuclide Corporation (now MAAS) "Luminoscope" operated at 13kv and 7  $\mu$ A. The section was  $\sim 2 \times 1$  cm and required about 40 images, each capable of resolving  $\sim 5 \mu$ m grains. Kodacolor Gold (ASA 400) film with the C-41 development process was used. Each chondrule was classified according to whether or not it produced CL; this is equivalent to sorting the chondrules by the FeO of the olivine grains. The sizes of the chondrules and their "rims" were measured in two perpendicular directions and the results averaged. We took as our operating definition of "chondrule rims", the fine-grained material, somewhat similar to matrix, that completely enclosed the chondrule, where a "chondrule" was any object, circular or slightly elliptical, with internal structure involving  $\geq 50 \mu$ m grains.

#### RESULTS

##### Cathodoluminescence Properties of Murchison

Plate I shows the CL of five regions of the Murchison meteorite. All of the important textures observed in our section are represented in the Plate. A color reproduction of the whole mosaic, compared with those of other primitive chondrites, was shown in Sears *et al.* (1991). The profusion of fine grains with bright red CL give the whole section a bright red appearance. We and others have shown that this fine-grained, highly dispersed mineral phase with red CL is forsterite (Steele, 1986;

Marshall, 1988; Sears *et al.*, 1991;). Additionally, many of the chondrules in these meteorites contain olivine grains with red CL, sometimes with Fe and CL zoning where the Fe is lowest and the CL brightest at the center of the grain. A number of refractory inclusions with complex rims in Murchison also show yellow CL.

Two kinds of chondrules may be readily identified depending on whether or not they show any CL. For reasons which will become apparent below, and following the precedent of Sears *et al.* (1992a) and DeHart *et al.* (1992), those which contain phases which luminesce are referred to as group A and while those which do not are referred to as group B.

##### The Chondrule Groups in Murchison

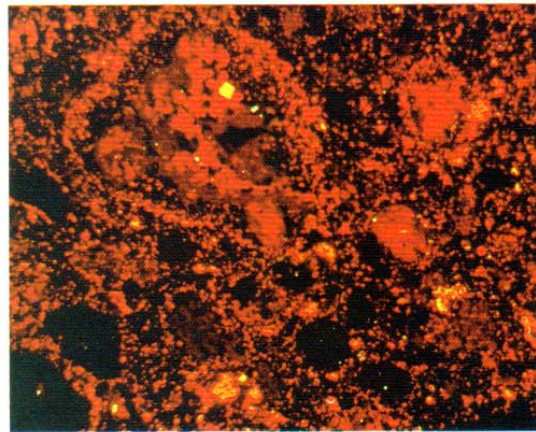
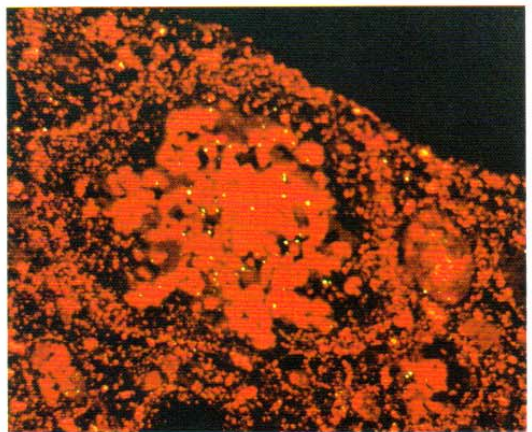
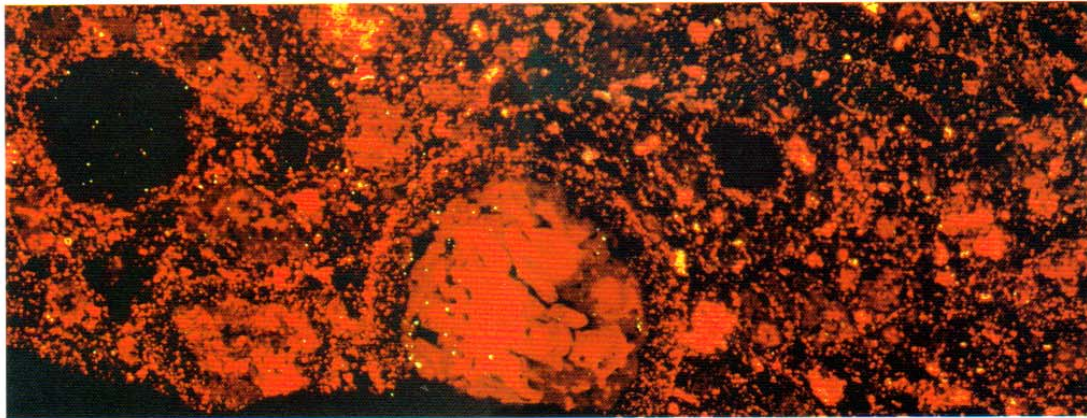
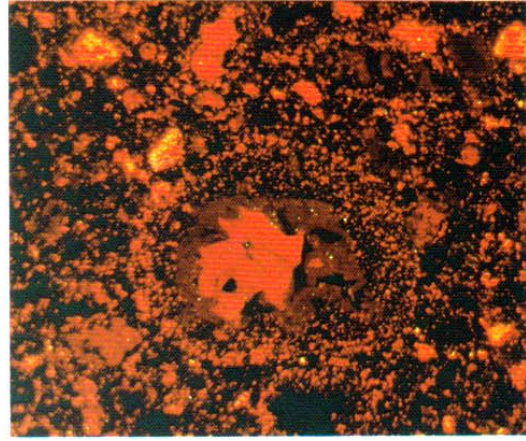
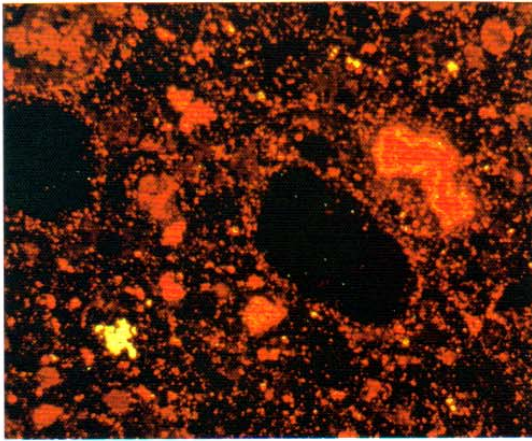
Chondrule olivine grains often display regions of dull-red CL surrounding intense bright CL areas (Plate Ib-e). Steele (1986) and DeHart (1989) have shown that about 2 wt% FeO is sufficient to greatly reduce the intensity of the CL of olivine while 4 wt% FeO reduces the CL to negligible levels. Thus the CL intensity is a sensitive indicator of the FeO content. About 62% of the chondrules (by number) in Murchison show bright red CL in their interior grains, while 38% do not. The chondrules without CL appear as the conspicuous black objects on the CL mosaic (Plate Ia and Ic). Consistent with this, about 68% of the olivine grains in Wood's (1962) survey of Murray, which resembles Murchison in many respects, have olivine compositions with  $< 2$  wt% FeO, while the 32% have 2–55 wt% FeO. Similarly, Scott and Taylor (1983) showed that Murray contains two distinct types of chondrule and, judging from their Fig. 6, about 62% have olivine grains with  $< 2$  wt% FeO and 38% contain olivine grains with 20–60 wt% FeO.

McSween (1977) has described these two groups of chondrule as type I and type II, using both texture and composition as a criterion. To a first approximation, type I chondrules are equivalent to group A and type II to group B, but there is a major difference in that the present groups are entirely compositionally defined while the McSween "types" are also textural. As such, the present groups are much broader and diverse in many properties, but  $\geq 95\%$  of the chondrules can be classified. Also unlike the types of McSween, a number of subdivisions of the A and B compositional groups are possible, especially when changes in composition and CL caused by metamorphism are taken into account (Sears *et al.*, 1992a; DeHart *et al.*, 1992).

##### Textures of Group A Chondrules

We are able to make three generalizations about the textures of the group A chondrules in Murchison. (1) They show great variation in their internal integrity. Some are intact and highly distinctive, while others appear to be falling apart and in some sense merging with the matrix. (2) Group A chondrules have much thicker rims. (3) The boundary between the thick rims and the surrounding matrix is usually much more

Plate 1 (Opposite). Cathodoluminescence (CL) images of structures in the Murchison CM chondrite. (a, top left) Matrix region including two group B chondrules and an irregular refractory inclusion with red and yellow CL. (b, top right) Group A chondrule with a thick fine-grained rim and CL zoning in the large central olivine. (c, center) Large group A chondrule, surrounded by a thick fine-grained rim. (d, bottom left) Large somewhat irregular group A chondrule with a thick fine-grained rim which, in places, seems to grade into the surrounding matrix. (e, bottom right) Another irregular group A chondrule with a thick fine-grained rim and mesostasis which has one small region with yellow CL. Several small group B chondrules are in the lower half of the image, each with thin fine-grained rims. All images are to the same scale, the horizontal width of the four smaller figures is 150  $\mu$ m.





regular and more readily delineated than the boundary between the rim and the group A chondrule which it is enclosing. The four particular chondrules included in Plate I were chosen to illustrate these subjective generalizations.

The chondrule shown in Plate Ib is one of the most distinctive in our section, showing an approximately circular outline with a large intact olivine grain. The olivine displays bright red CL in the central region, indicating <2 wt% FeO, but dull-red CL in the outer regions reflecting the slightly higher FeO content in the outer region. It is quite common for chondrule olivine grains in Murchison to show this CL- and Fe-zoning and the effect has been observed in low-FeO olivine grains in all classes (Miyamoto *et al.*, 1986; Jones and Scott, 1989; Steele, 1986; 1989; Jones, 1990). The interface between the central object and the rim in Plate Ib is very irregular in contrast to the rim/matrix boundary which is relatively sharp. There appears to be a region near the outer perimeter of the rim where the density of fine-grains is especially high. The chondrule shown in Plate Ic is also well-delineated in all its features and resembles the former chondrule in the FeO and CL zoning, the sharpness of the rim/matrix boundary compared with the boundary between the rim and the central object and the increased density of fine grains at the outer edge. However, there are many cracks separating the central olivine grains which also appear less jagged than in the chondrule in Plate Ib.

The chondrules shown in Plates Id and e are difficult to discern in the section because they resemble surrounding matrix. Their outlines are much less regular. Nevertheless the outline of the entire object (central object plus rim) is sharper than the interface between the fine grain mantle and the central cluster of loosely aggregated olivine grains. The olivines are a random mixture of the highly luminescent low-FeO grains and weak or non-luminescent grains. The CL does not indicate that the individual grains are symmetrically zoned in their FeO contents, as in the large olivine in the chondrule in Plate Ib.

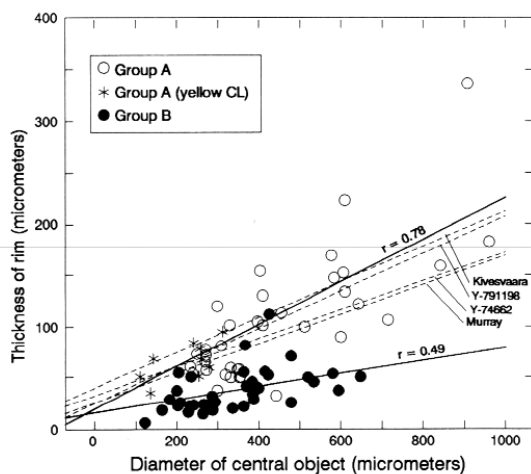


Fig. 1. Plot of the thickness of the rims vs. the central chondrule diameter to rim diameter for chondrules in the Murchison meteorite for group A and group B chondrules. Regression lines and their correlation coefficients are also indicated. Also shown are comparable regression lines for four other CM chondrites from Metzler *et al.* (1992).

### Rim Sizes and Chondrule Groups

Figure 1 compares the thickness of the fine-grained rims with the diameter of the central object (*i.e.*, chondrule), determined from our CL images. While there is overlap in the data for the diameters of group A and group B chondrules, and although group A chondrules extend to larger sizes, there is relatively little overlap in the thickness of the rims. This is despite uncertainty introduced into the data by the section not cutting the center of each chondrule. In fact, numerical modelling indicates the scatter we observe is equal to that predicted from this effect (E. Olsen, *per. comm.*). This confirms that *rims around group A chondrules are much thicker than those around group B chondrules of comparable diameter*. This is the first time an unequivocal relationship between rim thickness and chondrule type has been observed. The ratio of the rim thickness to the diameter of the entire object, Fig. 1 in Sears *et al.* (1992b), shows an even greater difference between the two chondrule groups. Our data for group A chondrules agree well with those of Metzler *et al.* (1992), which are also shown in Fig. 1, from which we conclude that their results concern only the relatively thick rims around group A chondrules.

A plot of the thickness of the fine-grained rim against the diameter of the entire object, that is, the central object (chondrule) plus rim (not shown), is qualitatively similar to Fig. 1. However, the correlation coefficients are much higher ( $r = 0.91$  and  $0.68$  for group A and group B chondrules, respectively). This is quantitative confirmation of our observation that the rim/chondrule boundary is less well-delineated than the rim/matrix boundary.

### DISCUSSION

#### Chondrule Groups and a Comparison with the Semarkona Ordinary Chondrite.

In addition to containing abundant fine-grained forsterite in their matrix and chondrule rims, primitive chondrites are similar in their populations of chondrules (Scott and Taylor, 1983). Thus the two chondrule groups we observe in Murchison are the equivalents of Semarkona's group A and B chondrules (we here neglect Semarkona's group A5 chondrules, which are relatively rare).

The main distinction concerns the mesostasis CL of the group A chondrules which in Murchison generally lack the bright yellow CL characteristic of Semarkona group A chondrules. Objects with yellow CL are present in Murchison, and a few of these may be regions of chondrule mesostasis, however the refractory inclusions with zones of yellow CL tend to be larger and more numerous (see Plates Ia and Ie). The lack of yellow mesostasis in Murchison chondrules is almost certainly because extensive aqueous alteration has caused the chondrule glass to be converted to non-luminescent phyllosilicates (Fuchs *et al.*, 1973; McSween, 1977; Bunch and Chang, 1980; Tomeoka and Buseck, 1985). A second distinction between the chondrule groups in Murchison and Semarkona concerns their relative abundance. The ratio (by numbers) of group A to group B chondrules in Murchison is  $\sim 2:1$ , while for Semarkona it is  $\sim 1:2$ .

#### The Rims, Their Origins and Their History

We can identify essentially three types of process whereby group A chondrules could have obtained much thicker rims than

group B chondrules. (1) If the rims are accreted dust, then the two chondrule groups may have formed in environments which differed greatly in the availability of dust. (2) If the rims are a secondary product, produced by alteration of the host chondrule, then differing compositions of chondrules somehow caused the formation of rims of differing thickness. (3) The rims reflect different degrees of zoning in the structure and composition of the precursor chondrules.

**Accretionary Origin for the Rims**—Allen *et al.* (1980) and King and King (1981) suggested that the rims formed by the accretion of nebular dust because the rims they examined were compositionally heterogeneous and yet not concentrically zoned. However, they also observed that the textures of the outer parts of the rims were suggestive of vapor deposition. In some cases, this might reflect redistribution of volatiles during metamorphism. (Their sample with the highest apparent "accretion temperature", Tieschitz, type 3.6, is relatively highly metamorphosed, cf. Prairie Dog Creek, 3.7; Inman, 3.3; Bishunpur, 3.1; Murray, 2; and Allende, probably 3.2-3.4). However, some of their samples are little-metamorphosed.

Several authors have now described rims with concentrically zoned compositions; such chondrules normally—but not always—have higher Fe/Si in the outer layers than in the inner layers and sometimes the zoning is quite complex (Bunch and Chang, 1980; Metzler *et al.*, 1992). We also see evidence for concentric zoning in the rims of group A chondrules in virtually all of our CL images, since the grain density around the outer edges of the rims is much higher than throughout the rim.

There are other arguments against the accretion of solid nebular dust by the chondrules prior to incorporation in the meteorite. Allen *et al.* (1980) themselves were concerned about the absence of nuclear track evidence for irradiation of the chondrule surfaces in free-space, but this might simply mean a short time in free-space. Formation in discrete environments, one dust-free and the other dust-rich, implies considerable transport of material in the early solar system, which has not appealed to many authors (e.g., Wood, 1985). Different chondrule size distributions and different proportions of group A to group B chondrules for each class are sometimes cited as evidence for little transport. Especially compelling is Wood's (1985) argument that despite considerable departures from cosmic composition shown by certain chondrules and the matrix, they always seem to end up in the appropriate proportions to give remarkably constant solar bulk compositions. It also seems unlikely to us that the accretion of dust around an already completely formed chondrule would be consistent with the rim/matrix boundary being sharper than the chondrule/rim boundary. *We suggest that the relationship between rim thickness and the host chondrule implies that it is the nature of the chondrule being rimmed which directly governs the formation of the rim.*

We agree with Metzler *et al.* (1992) that these thick group A rims did not form by picking up dust in a dry regolith since similar mantles are not seen around components in lunar or meteoritic regoliths. This is despite the considerable evidence that virtually all the CM chondrites, like most chondrites, underwent considerable brecciation, some in a regolith layer. Even the apparently unbrecciated Y-791189 may have been brecciated on a scale larger than a typical thin section. On the other hand, the fact that everything in Murchison has a thin rim of the sort seen on group B chondrules implies that their

formation post-dated the agglomeration of the components in their present form on the parent body. Unlike lunar and most meteorite regoliths, the CM chondrite regolith was apparently very wet and possibly the thin ubiquitous rims formed by the objects sloshing about in the mud (H.Y. McSween, per. comm.). Of course, this process alone would not explain why group A chondrule rims are so much thicker than group B chondrule rims.

**Secondary Origin for the Rims**—Since aqueous alteration has been such a major process in the history of the CM chondrites, it is reasonable to explore the idea that the thick group A chondrule rims in Murchison were produced by aqueous alteration of the host chondrule. This process might produce rims whose thickness was dependent on the composition of the host object, so that the calcic mesostases of group A chondrules—which are relatively susceptible to hydrous alteration—would yield thick rims while the SiO<sub>2</sub>-rich mesostases normally associated with group B chondrules (DeHart *et al.*, 1992) would produce no rims or only thin rims.

An argument against the formation of the thick group A rims entirely by aqueous alteration of the host chondrule is the uniformly fine-grained texture of the rim, with no evidence for grains of partially destroyed olivine and pyroxene (I. Steele, per. comm.).

**Precursor Zoning**—Rims are common around Semarkona chondrules and Huang *et al.* (1993b) recently found that metal- and sulfide-rich rims are more abundant around group A chondrules than group B chondrules. They also found the plot of rim thickness against chondrule size for Semarkona was remarkably similar to the Murchison data in Fig. 1. Semarkona group A chondrules show much evidence for reduction and evaporation of major elements during chondrule formation (Lu *et al.*, 1991; Huang *et al.*, 1993a), and Huang *et al.* (1993b) propose that the rims were formed by recondensation of the volatiles, which included metal and sulfide, in the outer regions or on the surface of the chondrules.

Whatever their origin, the thick group A rims could be the aqueously altered analogs of the metal and sulfide-rich rims seen in Semarkona. Like the Semarkona rims, the CM chondrule rims are volatile-rich (King and King, 1981; Bunch and Chang, 1980), but this might reflect the redistribution of volatile elements during aqueous alteration. Whether by interaction with gases during chondrule formation, or with fluids during aqueous alteration, one might also expect variations in the oxygen isotope ratios through the rims. Although there are no data for Murchison chondrule rims, the rims around Allende chondrules are invariably <sup>18</sup>O-rich and the olivines FeO-rich relative to the host chondrule (Rubin *et al.*, 1986).

**Parent Body Aqueous Alteration of Zoned Precursors**—We thus suggest that the two types of rim and their association with chondrules of different classes is described by the cartoon in Fig. 2. Prior to aqueous alteration, group A chondrules contained a thick rim with abundant metal and sulfide similar to that observed by Huang *et al.* (1993b) on the group A chondrules of Semarkona. Once on the parent body, aqueous alteration affected these rims in the way described by Tomeoka and Buseck (1985); metal and sulfide was converted to tochilinite, then olivine altered to cronstedtite and finally both these phases were converted to serpentine. Thus, as McSween (1987) observed, the chondrule to matrix ratio and Fe/Si of the matrix decreased during aqueous alteration as chondrules, metal and sulfide were destroyed and the products added to the matrix.

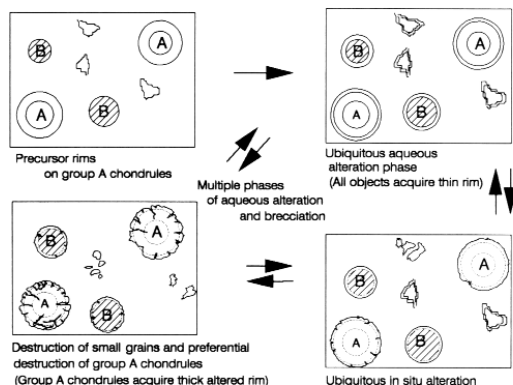


Fig. 2. Sketch of the proposed sequence by which thick-rimmed group A chondrules and all other objects in Murchison are altered and brecciated and acquire thin rims.

On the basis of such criteria, Murchison appears to be among least-altered CM chondrites (McSween, 1987).

Most authors consider that the aqueous alteration occurred on the parent body (Bunch and Chang, 1980; Tomeoka and Buseck, 1985), although Metzler *et al.* (1992) have suggested that some aqueous alteration was pre-accretionary occurring either in the nebula or on previous surfaces of the parent body. We also think it unlikely that much aqueous alteration occurred in the dispersed nebula. The considerable variability in composition of dust mantles, the small differences in the composition of the rims compared to the bulk meteorite, the close proximity (within  $\sim 50 \mu\text{m}$ ) of altered and unaltered material, the presence of serpentine apparently inside a chondrule (Fig. 21 in Metzler *et al.*, 1992), and the fact that altered material (grains of PCP) and individual mineral grains are surrounded by fine-grained rims, suggests to us that multiple episodes of aqueous alteration and brecciation on the parent body overlapped in time. Similarly, the evidence described by Ikeda and Prinz (1993) for nebular reactions is actually evidence that water was in the vapor state when it reacted with the silicates in CM chondrites, and this could also apply to shock and brecciation on the parent body since both involve localized excursions to high temperatures. Fegley and Prinn (1989) also argue that nebular aqueous alteration is unlikely on kinetic grounds.

Thus Fig. 2 tries to depict the effect of multiple phases of aqueous alteration and brecciation on a variety of starting materials. The ubiquitous phase of aqueous alteration resulted in all objects in Murchison having a thin fine-grained rim, either by the solidification of 'mud' or some other more complex process. The group A chondrules were altered to varying degrees depending on the duration of exposure to water and their original structure and composition. During the process, the thick-rimmed group A chondrules with complex multilayered sulfide and metal structures were altered to the complex rims described by Zolensky *et al.* (1988) and Metzler *et al.* (1992), while relatively metal- and sulfide-poor rims or the simple blocky structures sometimes observed (Huang *et al.*, 1993b; Allen *et al.*, 1980) resulted in less complex structures.

## SUMMARY AND CONCLUSIONS

We have found that chondrules in Murchison may be divided into two groups on the basis of their CL which reflect differences in the FeO content of the chondrule olivine grains. The groups, which we term group A and group B, are approximately equivalent to McSween's type I and type II. The CL images show that group A chondrules invariably have thick fine-grained rims which grade outwards into thin rims similar to those observed around all objects in Murchison, including group B chondrules, refractory inclusions, mineral and chondrule fragments and clasts. Most previous literature discussions of fine-grained rims around chondrules in CM chondrites have concerned these thick group A chondrule rims.

We suggest that these very thick group A chondrule rims were formed by aqueous alteration of the host chondrule's metal- and sulfide-rich rims which Huang *et al.* (1993b) show are characteristic of group A chondrules in ordinary chondrites. The greater clarity of the rim/matrix boundary compared with the rim/central object boundary is a result of this process. We see a variety of textures which appear to reflect progressive destruction of chondrules and their rims by aqueous alteration. We are, in effect, observing the process of chondrule destruction and the addition of the products to the matrix in the manner described by Tomeoka and Buseck (1985). It seems to us that most of the observations described in the literature can be reconciled with such a scenario, especially since most of these meteorites are breccias which have probably experienced multiple stages of alteration and brecciation. The frequent complex zoning in these chondrule rims is a result of the metal and sulfide distribution in the fine-grained rims prior to aqueous alteration.

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