

## MINERALOGICAL ALTERATION OF CM CARBONACEOUS CHONDRITES: A REVIEW

Kazushige TOMEOKA<sup>1</sup>, Harry Y. MCSWEEN, Jr.<sup>2</sup> and Peter R. BUSECK<sup>3</sup>

<sup>1</sup>*Mineralogical Institute, Faculty of Science, University of Tokyo,  
3-1, Hongo 7-chome, Bunkyo-ku, Tokyo 113*

<sup>2</sup>*Department of Geological Sciences, University of Tennessee,  
Knoxville, Tennessee 38996, U.S.A.*

<sup>3</sup>*Departments of Geology and Chemistry, Arizona State University,  
Tempe, Arizona 85287, U.S.A.*

**Abstract:** CM carbonaceous chondrites have been considerably affected by aqueous alteration, probably on the regolith of their parent body or bodies. The aqueous alteration resulted in the alteration of anhydrous silicates, metal, and sulfides, producing a complex mixture of Fe-Mg serpentines, Fe-Ni-S-O phase (tochilinite), and minor Fe-rich oxides and Fe-Ni sulfides. We here present a review of recent petrographic and mineralogical studies of CM carbonaceous chondrites and interpretations of their mineralogical alteration process.

### 1. Introduction

The compositions of carbonaceous chondrites are similar to elemental abundances in the solar photosphere (HOLWEGGER, 1977); thus these chondrites are commonly regarded as primitive solar system material that formed directly from the solar nebula. However, some chondrites, CI and CM type carbonaceous chondrites in particular, show abundant evidence of secondary aqueous alteration. Whether such alteration occurred before or after accretion to parent bodies has long been a subject of controversy, although it is now widely accepted that the alteration occurred in the parent bodies.

Numerous fracture-filling veins of carbonates and hydrous sulfates in CI chondrites provide strong evidence that aqueous alteration occurred by the activity of liquid water in the regolith of their parent bodies (DUFRESNE and ANDERS, 1962; NAGY *et al.*, 1963; BOSTRÖM and FREDRIKSSON, 1966; RICHARDSON, 1978). Disequilibrium assemblages of sulfides and clusters of magnetite grains with polygonal morphology in CI chondrites likely formed by such alteration (KERRIDGE *et al.*, 1979a, b). Olivine and pyroxene in chondrules and aggregates of CM chondrites are commonly replaced by phyllosilicates (BUNCH and CHANG, 1980; KOJIMA *et al.*, 1984; also see Fig. 1). Thus, the phyllosilicates that constitute the bulk of the CI and CM chondrite matrices probably resulted from aqueous alteration of fine-grained olivine and pyroxene. The brecciated textures and the fracture-filling veins suggest that the alteration at least partly overlapped a period of regolith gardening by meteoroid impacts, although shock effects were gentle enough not to alter the primary mineralogy (RICHARDSON and MCSWEEN, 1978; KERRIDGE and BUNCH, 1979). Oxygen isotopic

study of CI and CM chondrites (CLAYTON and MAYEDA, 1984) suggests that such alteration occurred at low temperatures ( $<20^{\circ}\text{C}$ ) and at high water: rock ratios ( $>44$  volume % of water). The low degree of ordering in poorly graphitized carbon in CM chondrites (RIETMEIJER and MACKINNON, 1985) supports the view that alteration occurred at relatively low temperatures.

The major fraction of CM carbonaceous chondrites is optically opaque matrix consisting of extremely fine-grained minerals. Knowledge of matrix mineralogy is crucial for determining the conditions under which these meteorites formed and the degree to which they retain their primitive character. The fine-grained and complexly mixed nature has long hindered detailed characterization of the matrix minerals. Recently, however, analytical transmission electron microscopes (ATEMs) have been used for studying the fine-grained minerals, and their results combined with those from petrographic studies have greatly improved our understanding of the matrix mineralogy of the CM carbonaceous chondrites.

## 2. Mineralogy of CM Matrix

The CM chondrites are regolith breccias composed of matrix, chondrules, aggregates (inclusions), isolated olivine and pyroxene grains, carbonates, and opaque phases — mostly Fe-Ni sulfides, kamacite (Fe-Ni metal), magnetite, and PCP (an intergrowth that was originally described as “Poorly Characterized Phase (PCP)” by FUCHS *et al.*, 1973). The modal abundance of matrix varies widely (57 to  $\sim 100$  vol%) among the CM chondrites (McSWEEN, 1979a). Aqueous alteration resulted in the replacement of chondrules and aggregates by phyllosilicates (Fig. 1) and converted them into optically obscure material, thus increasing the modal abundance of materials optically counted as “matrix”. Therefore, McSWEEN (1979a) proposed that the degree of alteration of CM chondrites could be proportional to and thus be measured by the relative proportion of matrix.

CM matrices are complex assemblages consisting mainly of phyllosilicates and an Fe-Ni-S-O phase (tochilinite, a constituent of PCP), with lesser amounts of Fe-Ni sulfides, magnetite, chromite, calcite, and organic compounds. Matrix minerals and their approximate chemical formulas are listed in Table 1.

### 2.1. Phyllosilicates

The phyllosilicates in CM matrix are predominantly Fe-Mg serpentine minerals\* that commonly contain small amounts of Al (FUCHS *et al.*, 1973; MACKINNON and BUSECK, 1979a, b; MCKEE and MOORE, 1979; MÜLLER *et al.*, 1979; AKAI, 1980, 1982; BUNCH and CHANG, 1980; MACKINNON, 1980, 1982; BARBER, 1981; TOMEOKA and BUSECK, 1983, 1985a; AKAI and KANNO, 1986). Although these minerals show a wide range of Fe/Mg ratios, they can be divided into two compositionally distinct groups corresponding to magnesian cronstedtite and ferroan serpentine; representative compositions are shown in Table 2. Members of the first group are Fe-rich and

\* The phyllosilicates have been called “septechlorites” by several investigators. However, the AIPEA nomenclature committee has recommended that the name “septechlorite” should not be used (BAILEY, 1980). Thus, we here call those minerals Fe-Mg serpentines.

Table 1. Matrix minerals of CM carbonaceous chondrites.

Mineral	Chemical formula
Ferroan serpentine	$(\text{Mg}_3\text{Fe}_3)\text{Si}_4\text{O}_{10}(\text{OH})_3$
Magnesian serpentine	$(\text{Mg}_5\text{Fe})\text{Si}_4\text{O}_{10}(\text{OH})_3$
Magnesian cronstedtite*	$(\text{Fe}_4\text{Mg}_2)(\text{Fe}^{3+}\text{Si}_3)\text{O}_{10}(\text{OH})_3$
Fe-Ni-S-O phase (tochilinite)*	$\text{Fe}_{1.3}\text{Ni}_{0.1}\text{SO}_{1.4}(\text{OH})?$
Troilite	$\text{FeS}$
Pyrrhotite	$\text{Fe}_{1-x}\text{S}$
Pentlandite	$(\text{FeNi})_9\text{S}_8$
Magnetite	$\text{Fe}_3\text{O}_4$
Chromite	$\text{FeCr}_2\text{O}_4$
Calcite	$\text{CaCO}_3$
Organic compounds	$\text{C}_l\text{-H}_m\text{-O}_n\text{-N}_p$

\* Major constituents of PCP.

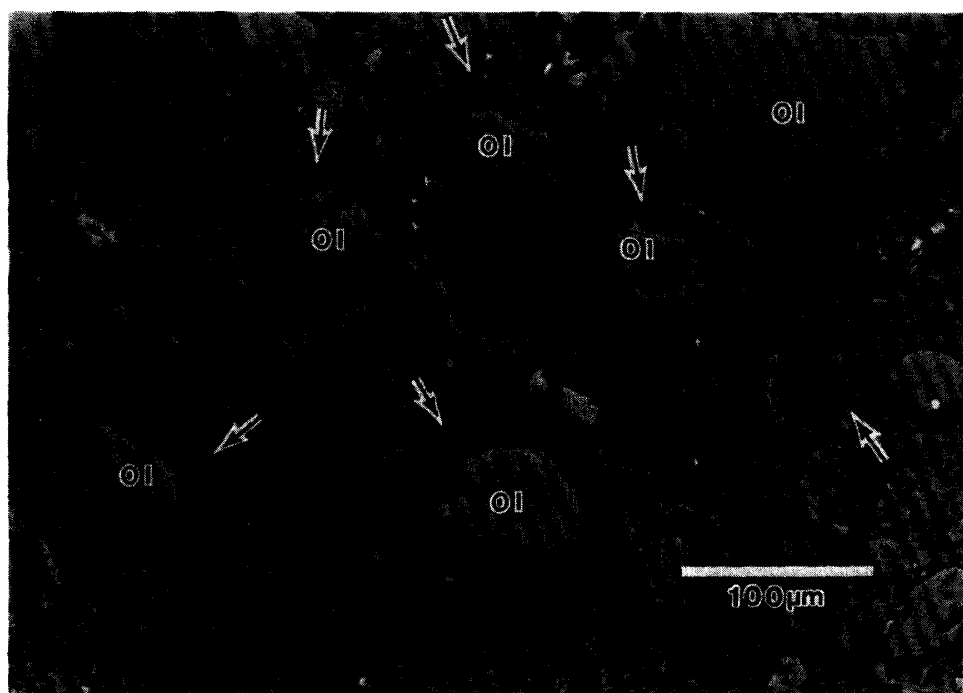


Fig. 1. Image by reflected-light optical microscopy of part of a chondrule that has been extensively replaced by phyllosilicates (Murray CM meteorite). Outlines of partially and completely replaced olivine (OI) grains (indicated by arrows) are well preserved.

have  $\text{Fe}^{3+}$  substituting for Si in tetrahedral sites, in addition to the  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  in octahedral sites. Their compositions are similar to that of cronstedtite, which has an ideal end-member composition of  $(\text{Fe}_4^{2+}\text{Fe}_2^{3+})(\text{Si}_2\text{Fe}_2^{3+})\text{O}_{10}(\text{OH})_3$ , but they differ from ideal cronstedtite in containing less  $\text{Fe}^{3+}$  and substantial Mg. The ferroan serpentine group is more Mg-rich and has a composition intermediate between end-member serpentine and greenalite; its chemical formula can be expressed as  $(\text{Fe, Mg})_9\text{Si}_4\text{O}_{10}(\text{OH})_3$ .

The magnesian cronstedtite and ferroan serpentine display a wide range of morphologies and degrees of crystallinity, and they are intimately mixed and intergrown

Table 2. Representative electron microprobe analyses of major constituent minerals of CM matrix.\*

	Fe-Ni-S-O phase <sup>1)</sup> (tochilinite)	Magnesian <sup>1)</sup> cronstedtite	Ferroan <sup>2)</sup> serpentine	Magnesian serpentine
SiO <sub>2</sub>	0.8	21.2	33.5	38.4
Al <sub>2</sub> O <sub>3</sub>	3.8	2.2	3.3	2.1
Cr <sub>2</sub> O <sub>3</sub>	0.0	0.0	0.2	0.3
FeO	62.6	54.2	25.3	13.8
MgO	4.4	7.6	16.8	33.0
CaO	0.0	0.0	0.3	0.2
P <sub>2</sub> O <sub>5</sub>	0.0	0.0	0.2	0.2
S	20.8	2.0	2.5	1.6
NiO	4.8	0.2	1.9	1.5
Total	97.2	87.4	84.0	91.1

\* Because of intimately intergrown nature of the matrix, the analyses include minor amounts of other phases.

1) From Murray (TOMEOKA and BUSECK, 1985).

2) From Nogoya (BUNCH and CHANG, 1980).

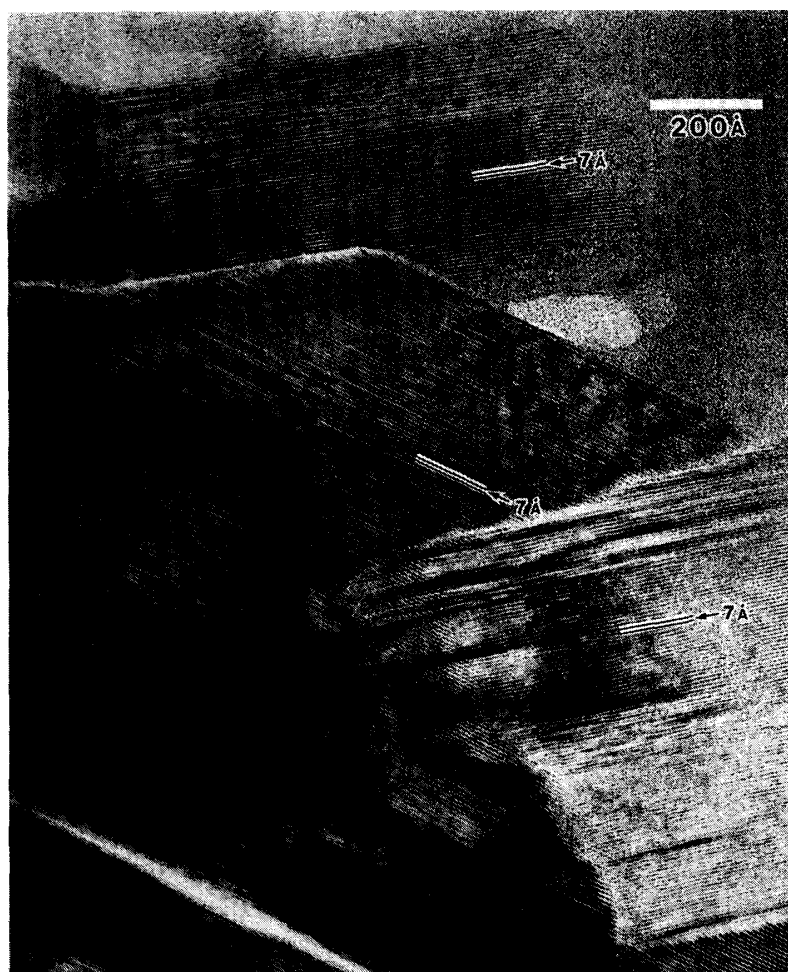


Fig. 2. High-resolution TEM image of platy cronstedtite crystals showing interpenetrating texture in the matrix of the Murchison CM meteorite. Fringes indicated by arrows correspond to cronstedtite layers having  $\sim 7\text{\AA}$  interlayer spacing.

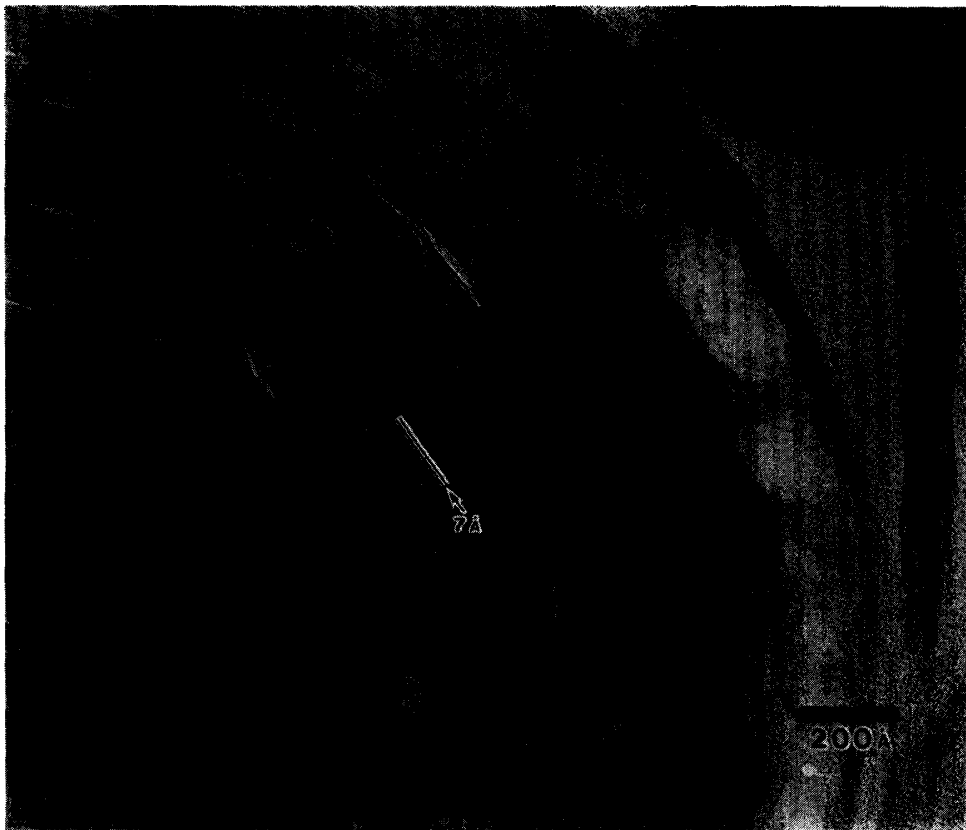


Fig. 3. Part of a cross section of tubular serpentine in the matrix of the Mighei CM meteorite (TEM image).

on a submicron scale. Interlocking and interpenetrating textures are common (e.g., Fig. 2), indicating that the phyllosilicates developed through complex replacement reactions. Some of them show curved, tubular, and polygonal structures that are characteristic of chrysotile serpentine (BARBER, 1981; also see Fig. 3).

The relative proportions, Fe/Mg ratios, compositional homogeneity, and crystallinity of the phyllosilicates vary considerably among CM chondrites and can be correlated with degree of alteration. Magnesian cronstedtite is abundant in CM chondrites that were relatively mildly altered, where it commonly occurs as well-developed platy crystals. In more altered CM chondrites, ferroan serpentine is more abundant.

Chondrules and aggregates also contain phyllosilicates that have a characteristic transparent green-to-brown color; these phases were described by FUCHS *et al.* (1973) as "spinach". This material shows broad compositional variations in Fe, Mg, and Al contents (RICHARDSON, 1981; IKEDA, 1983; KOJIMA *et al.*, 1984). While most spinach consists of either magnesian cronstedtite or ferroan serpentine, some spinach contains up to 18 wt%  $\text{Al}_2\text{O}_3$  and is chemically similar to berthierine, a ferroan aluminous serpentine. The aluminous spinach is probably an early alteration product of primary mesostasis glass in chondrules and aggregates. During subsequent aqueous alteration, this spinach lost Al and gained Fe by elemental exchange with the matrix (RICHARDSON, 1981; KOJIMA *et al.*, 1984). A chlorite-like phyllosilicate was

also reported from chondrules in an Antarctic CM chondrite (AKAI and KANNO, 1986). In highly altered chondrules and aggregates, olivine and pyroxene phenocrysts are also replaced by phyllosilicates (Fig. 1). Therefore, the phyllosilicates in chondrules and aggregates show, like matrix phyllosilicates, a tendency to increase in both Si/Fe and Mg/Fe ratios with increasing degrees of alteration.

## 2.2. Fe-Ni-S-O phase (tochilinite)

PCP has been one of the more problematical materials in the CM chondrites

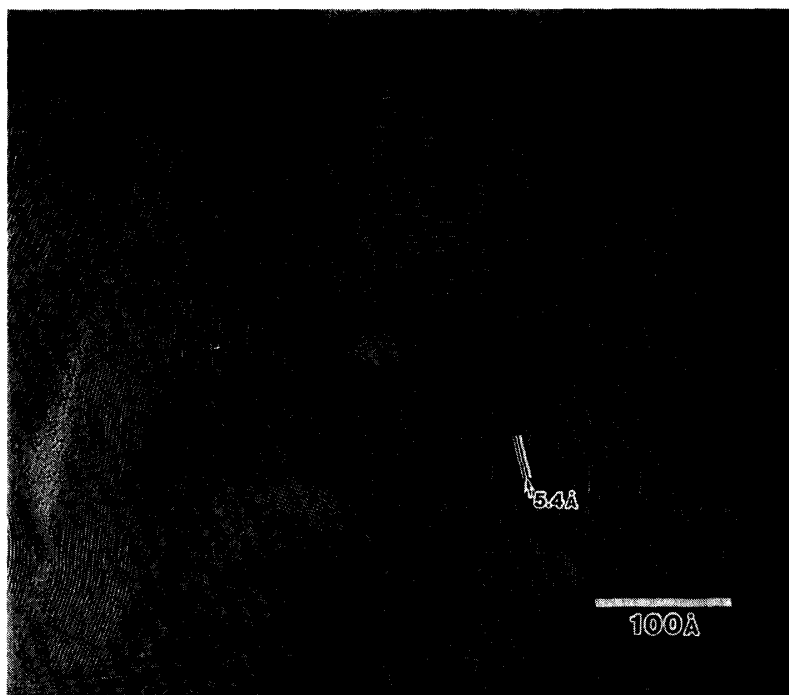


Fig. 4a.



Fig. 4b.

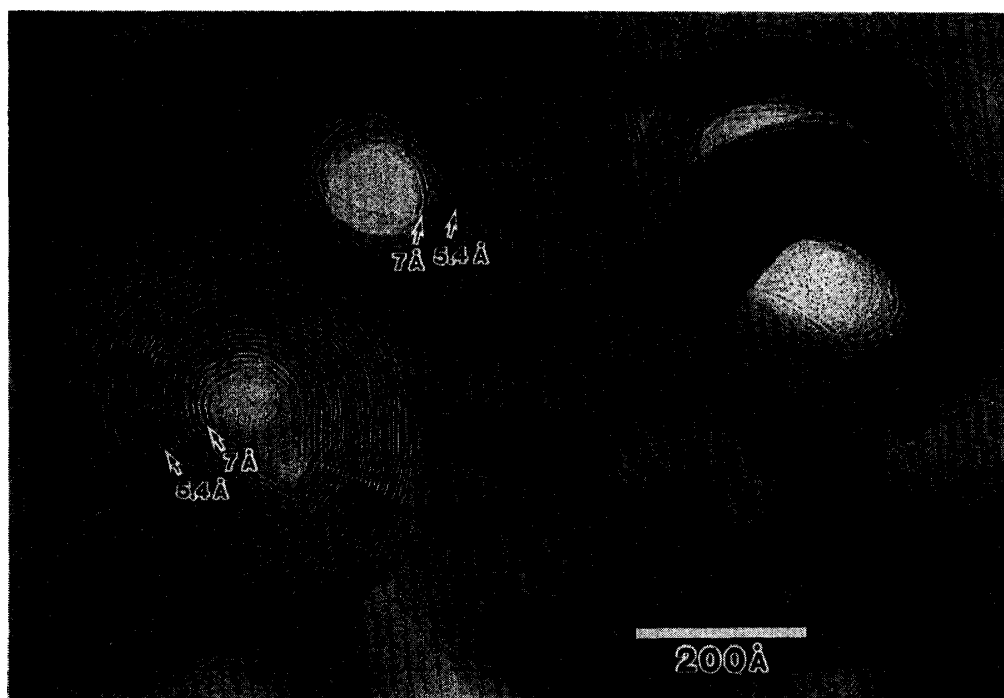


Fig. 4c.

Fig. 4. (a), (b) High-resolution TEM images of the Fe-Ni-S-O phase (probably tochilinite) in PCP (Murray CM meteorite). The images are cross sections of tubular bands of layers. The spacing between the layers is approximately  $5.4\text{Å}$ . Both bands have irregular fringes roughly perpendicular to the layer directions; they resulted from layer corrugation. (c) Tubular, coherent intergrowths of the  $5.4\text{Å}$  layers (Fe-Ni-S-O phase) and  $7\text{Å}$  layers (cronstedtite) (Murchison CM meteorite).

(RAMDOHR, 1963; FUCHS *et al.*, 1973; BUNCH and CHANG, 1980). It occurs in chondrules and aggregates as rounded inclusions, and also in matrix as aggregates of platelets and acicular fibers. It is rich in Fe and O and contains variable amounts of S, Ni, Si, Mg, Cr, and P. TOMEOKA and BUSECK (1983, 1985a) studied PCP in three CM chondrites using transmission electron microscopy and found that it is a complex intergrowth of 1) an Fe-Ni-S-O layered mineral, 2) cronstedtite, and 3) minor amounts of submicron grains of magnetite, chromite, and a mineral containing Fe, Ni, Cr, and P. PCP is particularly abundant in the CM meteorites that are relatively unaltered and that show high Fe contents in their matrices (*e.g.*, Mighei, Murchison, and Murray), and it is responsible for a large portion of this Fe.

The Fe-Ni-S-O phase has a pronounced layering repeat of  $\sim 5.4\text{Å}$  and a weak repeat of  $\sim 10.8\text{Å}$ . It shows peculiar curved, rolled, and wavy structures (Figs. 4a, b, c). It and cronstedtite commonly form alternating intergrowths in ordered and disordered arrangements (MACKINNON and BUSECK, 1979a, b; AKAI, 1980, 1982; MACKINNON, 1982; TOMEOKA and BUSECK, 1983, 1985a; also see Fig. 4c). Assuming that it contains no hydroxyl, the Fe-Ni-S-O phase has an approximate chemical formula of  $\text{Fe}_{1.3}\text{Ni}_{0.1}\text{SO}_{1.4}$  (TOMEOKA and BUSECK, 1985a; also see Table 2). BARBER *et al.* (1983) reported it to be  $\text{Fe}_{1.4}\text{SO}_{1.3}$ , in which Ni/Fe ratios vary between 0.03 and 0.04. BARBER *et al.* (1983) suggested that it may be a hydrous sulfide. MACKINNON and ZOLENSKY (1984) proposed that it is equivalent to an Fe- and Ni-rich member of tochilinite,  $6\text{F}_{0.9}\text{S} \cdot 5[(\text{Mg}, \text{Fe})(\text{OH})]_2$ , which consists of coherent interstratified

makinauwite-type sulfide sheets and brucite-type sheets (ORGANOVA *et al.*, 1974). An electron microscope study of ZOLENSKY and MACKINNON (1986) showed that tochilinite has structures and textures similar to the Fe-Ni-S-O phase, supporting its identification as tochilinite. It appears to have considerable variations in structure and composition, and its more detailed characteristics remain to be studied.

Because PCP turned out to be an intergrowth of several phases, the term "PCP" in the sense originally described is now inappropriate. PCP was redefined as "Partly Characterized Phases" by TOMEOKA and BUSECK (1985a). We still need more work to make it "Perfectly Characterized Phases".

### 2.3. Sulfides, magnetite, and other minerals

Small grains ( $<1 \mu\text{m}$  in diameter) of troilite, pentlandite, and magnetite are dispersed through the matrix (BARBER, 1981; TOMEOKA and BUSECK, 1985a). In places, magnetite occurs in clusters of grains with polygonal morphology, similar to the framboidal magnetite that is common in CI chondrites (TOMEOKA and BUSECK, 1985a; HYMAN *et al.*, 1985). The sulfides apparently formed from Fe, S, and Ni that migrated out of PCP. In addition to those formed during alteration of Fe-rich olivine, many magnetite grains were probably also derived from PCP. Calcite occurs as individual and mosaic clusters of small grains in association with phyllosilicates and PCP. Needles of PCP commonly intrude calcite, suggesting simultaneous formation (FUCHS *et al.*, 1973; KERRIDGE and BUNCH, 1979; BUNCH and CHANG, 1980).

## 3. Aqueous Alteration Process

The mineralogy of matrix prior to alteration may be approximated by some of the CV chondrites, which are little affected by alteration and are probably petrologically more primitive than CI and CM chondrites (McSWEEN, 1979b, 1987). The CV matrices consist largely of small grains (0.1–10  $\mu\text{m}$  in diameter) of Fe-rich olivine ( $\text{Fo}_{40-80}$ ) (PECK, 1983). In contrast, the olivines and pyroxenes that form chondrules, aggregates, and isolated grains are much more coarse-grained and mostly Mg-rich ( $\text{Fo}_{90-100}$ ;  $\text{En}_{90-100}$ ). Because the matrix is fine-grained, it is more permeable to fluids and thus more reactive than the coarser olivines and pyroxenes in chondrules and aggregates. Therefore, in the initial stages of aqueous alteration, the fine-grained fayalitic olivine in the matrix must have been preferentially altered to phyllosilicates, forming an initial Fe-rich matrix.

TOMEOKA and BUSECK (1985a) proposed that PCP developed through a series of alteration processes, and these are also directly related to the formation of the matrix phyllosilicates. The alteration can be understood as the result of reactions among the four major matrix minerals: (1) Fe-Ni-S-O phase (tochilinite), (2) magnesian cronstedtite, (3) ferroan serpentine, and (4) magnesian serpentine; representative compositions are shown in Table 2. The proposed process is summarized below and schematically illustrated in Fig. 5.

During Stage I, PCP, which consists mainly of tochilinite, formed by a reaction between kamacite and a S-bearing fluid that moved through cracks in chondrules and aggregates. During Stage II, the meteorites were subjected to aqueous alteration that



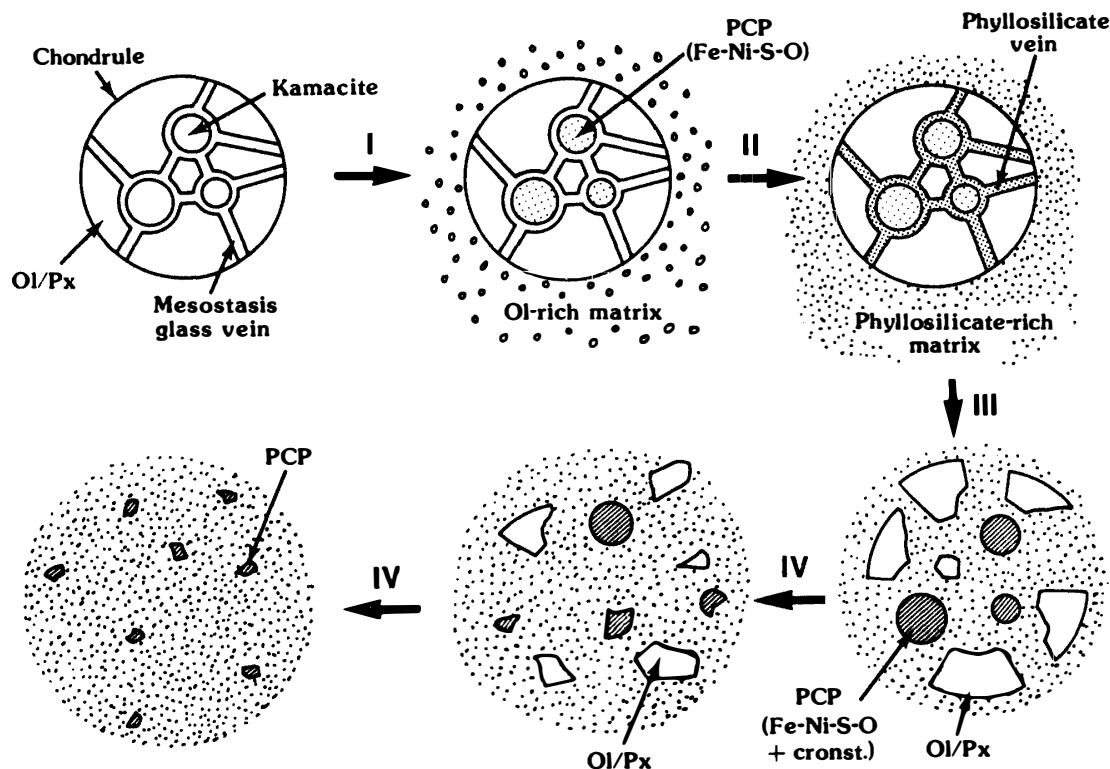


Fig. 5. Schematic illustration of the aqueous alteration process that probably occurred in the regolith of the CM carbonaceous chondrite parent body. The alteration converted chondrules and fine-olivine-grain-rich matrix to the phyllosilicate-rich matrix (modified from TOMOOKA and BUSECK, 1985a).

presumably was contemporaneous with regolith gardening. At this time, much of the fine-grained, Fe-rich matrix olivine was altered to phyllosilicates. Then, mesostasis glasses in chondrules and aggregates were attacked preferentially to form "spinach". The spinach formation probably reduced the mechanical strength of chondrules and aggregates and caused their disaggregation (RICHARDSON and MCSWEEN, 1978). By this mechanism, PCP grains were separated from chondrules and aggregates and were distributed into matrix.

During Stage III, Si, which was released during alteration of olivine and pyroxene, reacted with tochilinite in PCP, producing magnesian cronstedtite. As a result, the intergrowths of tochilinite and cronstedtite were produced, and they recrystallized into narrow fibers. In partially altered CM chondrites such as Mighei, Murchison, and Murray, the alteration was incomplete, thus leaving many heterogeneous mixtures of tochilinite and magnesian cronstedtite, *i.e.*, PCP, in the matrix.

As alteration proceeded (Stage IV), tochilinite was progressively consumed to form magnesian cronstedtite. Cronstedtite increased its Mg and Si contents by reacting with serpentine, altering to ferroan serpentine. With prolonged gardening, the matrix was stirred, and PCP was degraded and intermixed with the newly formed serpentine. Consequently, the CM matrix became more enriched in Mg (dominated by magnesian serpentine) and increasingly homogenized in its Mg/Fe distribution. Simultaneously, increasing amounts of Fe-Ni sulfides and magnetite were derived

from tochilinite and cronstedtite and were deposited as fine grains in the matrix.  $\text{Ca}^{2+}$  and  $\text{CO}_3^{2-}$  were also transported through solutions and deposited as calcite.

Advanced stages of alteration of the CM meteorites can be seen in the Nogoya and Yamato-82042 (Y-82042) meteorites. Both are far more altered than other CM chondrites. BUNCH and CHANG (1980) indicated that the serpentines in Nogoya contain more Mg and less Fe, on average, than those in other CM chondrites, and they are also compositionally homogeneous and structurally ordered, suggesting that they are approaching equilibrium. GRADY *et al.* (1987) also found that the Y-82042 matrix consists largely of magnesian serpentines. In this meteorite matrix, they noted that cronstedtite is absent and tochilinite is minor, whereas dispersed grains of Fe-Ni sulfides and carbonates are significantly concentrated. These mineralogies are consistent with the proposed progressive alteration process.

#### 4. Mineralogical Variations during Alteration

The relative proportions of secondary phases in matrix thus changed progressively during aqueous alteration. Such changes are reflected in the Fe-Mg-Si contents of CM matrices (MCSWEEN, 1979a, 1987). CM matrix became poorer in Fe with advancing alteration, as illustrated by the filled dots in Fig. 6. The Fe contents apparently decreased through the production of phyllosilicates with higher Mg/Fe ratios at the expense of olivine and pyroxene in chondrules and aggregates, consistent with the model based on textural observations noted above. The compositions of these phases (BUNCH and CHANG, 1980; TOMEOKA and BUSECK, 1985a) are shown for reference in Fig. 6.

Based on defocused electron beam analyses of matrices of over thirty non-Antarctic and Antarctic CM chondrites, MCSWEEN (1987) did mass balance calculations to specify the relative proportions of secondary phases in matrix and thus to evaluate the aqueous alteration model. The ratio of cronstedtite to tochilinite in PCP is approximately 3:1, as determined from application of the lever rule to the intercept point of the matrix trend line with the cronstedtite—tochilinite tie-line in Fig. 6. The same procedure gives an approximate 1:1 ratio of ferroan to magnesian

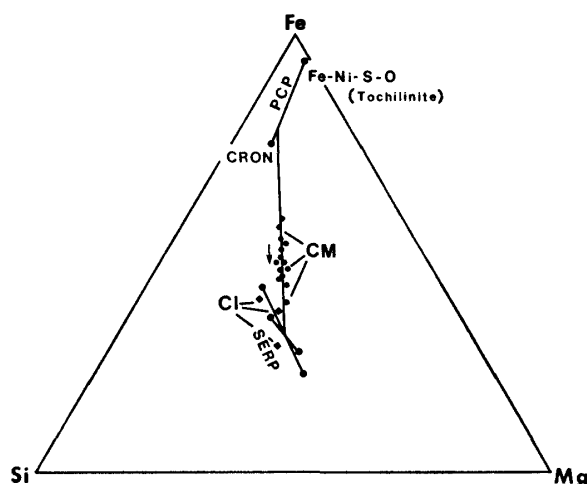


Fig. 6. Relative weight proportions of Fe, Si, and Mg in bulk matrices of CM and CI chondrites (data from MCSWEEN and RICHARDSON, 1977; MCSWEEN, 1979a, 1987). There is a trend that CM matrices (filled dots) become poorer in Fe with increasing degree of alteration; the trend is indicated by an arrow. The compositions of cronstedtite (CRON), Fe-Ni-S-O phase (tochilinite), and serpentines (SERP) (data from BUNCH and CHANG, 1980; TOMEOKA and BUSECK, 1985a) are shown for reference. Coexisting magnesian and ferroan serpentine in both Murray and Nogoya are illustrated; the steeper SERP tie-line corresponds to Nogoya. CM chondrite matrices are mixtures of PCP and serpentines that changed progressively to more serpentine-rich compositions with increasing alteration.

serpentes.  $SERP/(SERP+PCP)$  ratios for individual meteorite matrices can also be constrained by this procedure.

The results indicate that the compositional variability of CM matrices apparently reflects differing amounts of PCP and serpentes, and that the progressive alteration model is generally consistent with changes in their relative proportions. The alteration model predicts that  $PCP/(SERP+PCP)$  should vary inversely with increasing degree of alteration, thus with decreasing matrix Fe/Si ratios. McSween's calculations suggest that most data follow the predicted trend (see Fig. 6 in MCSWEEN, 1987). A more precise correlation may be obscured because PCP concentrations in some meteorites may be overestimated due to the presence of magnetite and sulfides. ZOLENSKY *et al.* (1988) suggested another mechanism which may explain this compositional trend. They noted that chondrule rim material was Fe-rich, and the intimate mixing of disaggregated rim material with matrix may produce the observed trend.

Despite their primitive chemical compositions, CM chondrites have been altered considerably by aqueous fluids. Thus, if we are to interpret the record in these meteorites, it is of obvious importance that we understand those alteration processes. However, the alteration of carbonaceous chondrites was extremely complex, and a more extensive survey and detailed data are needed to obtain a more precise understanding. We have just begun to document matrix mineralogy of some CM chondrites in sufficient detail to provide a possible alteration model.

## 5. Alteration in Other Types of Chondrites

CI matrix compositions lie near an extension of the proposed alteration trend for CM matrices (Fig. 6), thus suggesting that CI chondrites may have experienced more intense alteration than CM chondrites (MCSWEEN, 1979a, b). However, a recent TEM study showed that the matrix mineralogy of the Orgueil CI chondrite differs significantly from those of the CM chondrites and thus suggests that the CI and CM chondrites were derived from different primary materials and experienced distinct aqueous alteration (TOMEOKA and BUSECK, 1988). Thus, the petrographic differences between CI and CM chondrites may not simply be dependent on the extent of the aqueous alteration.

The presence of small amounts of phyllosilicates in some CV chondrites (TOMEOKA and BUSECK, 1982a, b, 1986; COHEN *et al.*, 1983; HASHIMOTO and GROSSMAN, 1987), CO chondrites (KELLER and BUSECK, 1988), and ordinary chondrites (HUTCHISON *et al.*, 1987) suggests that these meteorites have also experienced aqueous alteration, although to lesser degrees than CI and CM chondrites. Some interplanetary dust particles also contain phyllosilicates that appear to have been produced by aqueous alteration of anhydrous pyroxene and glass (TOMEOKA and BUSECK, 1985b; BRADLEY, 1988). Thus, aqueous alteration is clearly an important process that occurred in the early solar system.

Once the secondary mineralogy of these meteorites has been detailed, we must place constraints on the physical and chemical conditions necessary to cause the alteration. It seems clear that heavily altered chondrites will not provide mineralogical

data on processes in the early solar nebula, but they will be informative about chemical and geological aspects of primitive parent bodies or planetesimals.

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