Geochimica et Cosmochimica Acta Vol. 44, pp. 1543 to 1577
Pergamon Press Ltd 1980. Printed in Great Britain

Carbonaceous chondrites—II. Carbonaceous chondrite phyllosilicates and light element geochemistry as indicators of parent body processes and surface conditions

T. E. BUNCH and S. CHANG
Ames Research Center, NASA, Moffett Field, CA 94035, U.S.A.

(*Received* 11 *December* 1978; *accepted in revised form* 12 *June* 1980)

Abstract—Petrographic observations and analyses of CM matrices are consistent with their origin as *in situ* low temperature (<400°K) aqueous alteration products in a parent body regolith. At least four different phyllosilicates were tentatively characterized in Murray and Murchison meteorites, in addition to Fe- and Mg-serpentines in Nogoya. In comparison with bulk meteorite compositions, all phyllosilicates and bulk matrices show enrichment of K relative to Na. Possible loss of Na and possibly some Cl, with addition of H_2O and CO_2 and water-soluble organic compounds during alteration, indicates a partially open system during alteration. Poorly characterized phases (PCP) are fine-grained ($<1 \mu m$) admixtures of variable proportions of phyllosilicates, carbonaceous matter and opaque oxides of sulfur with high Fe, Ni and Cr contents. Calcite and some magnetite show paragenetic overlap with PCP and phyllosilicates. Carbonaceous matter is largely associated with PCP in altered CM matrices. In the unaltered CV Allende, carbonaceous matter is concentrated on olivine surfaces as a micromounded coating, particularly in the dark haloes that surround some chondrules and aggregates. Precursive alteration material may have been analogous to similarly coated olivine mixed with smaller amounts of metal and sulfides.

Synthesis of the water soluble organic compounds found in CM matrices may have occurred prior to or in the same environment as did aqueous alteration of the precursive phases. Preservation or partial preservation of this organic matter may reflect the degree of overlap in episodes of synthesis and alteration.

Nogoya is 95% altered and has a bulk carbon content of 5.2 wt%, which is higher than any meteorite. In addition, it has the lowest measured $^{13}C/^{12}C$ ratio of any other carbonaceous chondrite, except for Karoonda.

INTRODUCTION

CARBONACEOUS CHONDRITES are some of the most studied objects in the solar system, principally because of the presence of water-bearing phases (in CI and CM types), organic compounds, relatively high noble gas concentrations, and possible high-temperature condensates (CM, C3(V), and C3(O) types), and apparently primitive bulk compositions. Moreover, ZELLNER and BOWELL (1977) have observed that C-type asteroids (material closely resembling carbonaceous chondrites) are the most abundant main belt asteroid. Thus, carbonaceous chondrites may serve as a 'Rosetta Stone' in unraveling the mysteries of the origin and early history of the solar system.

Results to date from the study of so-called high-temperature condensates in carbonaceous chondrites have been very useful in strengthening theoretical models of the chemical behavior of a cooling gas of solar composition (LORD, 1965; LARIMER, 1967; GROSSMAN, 1972). Matrices of carbonaceous chondrites are considered to represent a low-temperature fraction of solar nebula condensation (ARRHENIUS and ALFVEN, 1971; GROSSMAN, 1972; WILKENING, 1978) or hydrothermal alteration, based on the observations of CI chondrites (FISH *et al.*, 1960; DUFRESNE and ANDERS, 1962; NAGY *et al.*, 1963; BOSTRÖM and FREDRIKSSON, 1966; KERRIDGE, 1977; RICHARDSON, 1978) and CM chondrites (DUFRESNE and ANDERS, 1962; BUNCH and CHANG, 1978; BUNCH *et al.*, 1979). Briefly, the condensation theory involves reaction of the residual solar nebula gas with earlier formed phases, mostly ferromagnesian minerals, NiFe metal, and FeS in the presence of water at 350–500°K. Nonorganic products from these reactions are layer–lattice silicates, magnetite, and pentlandite. The organic compounds in CI and CM meteorites have been attributed with both nebula and parent body origins; discussions of possible formation locations and reaction mechanisms are provided in reviews by ANDERS *et al.* (1973) and MILLER *et al.* (1976). Formation of CI chondrites by low-temperature aqueous alteration of pre-existing materials is generally accepted by the condensation proponents, although similar alteration-like components present in CM meteorites are thought by many to be strictly preplanetary condensation products.

In a recent report (BUNCH and CHANG, 1978), we suggested that the matrices of CM meteorites developed in response to low-temperature hydrothermal alteration, similar to CI chondrites but to a lesser degree. We report here more detailed work on the matrices of Jodzie, Murray, Murchison, and Nogoya, in which we find ample evidence to confirm our earlier contention that the 'low-temperature' matrices of carbonaceous chondrites formed in aqueous media under low temperature (<400 K) on or in the near surface of a planetary body. Our observations of some C3(V) and C3(O) matrices also suggest they were partially altered and that *in situ* alteration of carbonaceous chondrites was more prevalent that previously recognized. How and where the low-temperature fraction (matrices) formed is important to understanding the evolution of organic compounds, entrapment and retention of noble gases (the carrier is a matrix phase) and environmental constraints on parent body surfaces.

A growing number of investigators are becoming more convinced that all carbonaceous chondrites are not representative of pristine condensation materials, either the 'high' or 'low' temperature fractions, but are examples of a segment of primordial composition altered by thermal, mechanical and hydrothermal processes (KERRIDGE and BUNCH, 1979; BUNCH and CHANG, 1979; MCSWEEN, 1979a, b).

In this report, we use the revised carbonaceous chondrite classification (VAN SCHMUS, 1969) that subdivides C3 meteorites in C3(V) (Vigarano type) and C3(O) (Ornans type). The C2 designation is replaced by CM.

GENERAL DESCRIPTION OF MURCHISON–MURRAY–JODZIE TYPE MATRICES

Standard and ultrathin sections of meteorites used in this investigation were prepared for petrographic, electron microprobe and scanning electron microscope (SEM) analyses. In addition, a bulk sample of Murchison was subjected to multiple cycles of freeze–thaw–ultrasonication treatment with subsequent grain-size separation to purify and concentrate phyllosilicates. Phyllosilicate separates were investigated by SEM, transmission electron microscopy (TEM) and diffraction, and X-ray diffraction methods.

Two CM meteorites, Murchison and Murray and CM clasts in the Jodzie howardite were studied in detail for their matrix characteristics. Because Jodzie clasts (BUNCH *et al.*, 1979) and Murchison (FUCHS *et al.*, 1973) have been adequately described, most of the observations and data presented here are from Murray. We emphasize that matrix characteristics of all three meteorites are very similar.

The CM matrices consist of phyllosilicates, 'poorly characterized phases' (PCP), sulfides [pentlandite, pyrrhotite, 'Q' sulfide (LEWIS *et al.*, 1975)], magnetite and hydrated iron oxides, calcite, solvent-and-acid-insoluble carbonaceous (SAIC) matter, solvent-and-acid-extractable organic compounds, and mineral and lithic clasts. Phyllosilicates, PCP and carbonaceous matter are intimately associated and serve as host matrix for the other components.

Figure 1 shows an extensively altered area in Murray. The overall textures of most areas in CM meteorites are similar to terrestrial altered volcanic tuffs and tuff breccias in which precursive components are pseudomorphed by alteration minerals and the original shape outlines are sharply preserved. Nearly all clasts $< 100\,\mu$ are altered to OH-bearing phases and PCP. Metal and sulfides in the matrix show various degrees of alteration and those in chondrules and clasts are often less altered. Figures 1 through 7 show typical examples of *in situ* alteration of precursive materials to phyllosilicates and PCP. Mechanical movement of components in possibly a water (vapor or liquid) medium is indicated by partial rotation of chondrules and aggregates, schlieren of comminuted minerals, clasts and phyllosilicate patches, boudinage-like structures, and subtle foliated zones, which together given an appearance of protomylonite, but with much less intense dynamic metamorphism. An alternative explanation is alteration of a preexisting compaction or shock lithification fabric similar to that described for Allende by KING and KING (1978).

Murray is a good example of a regolith breccia because it contains abundant clasts of other carbonaceous chondrites (CM and C3) that range in size from tens of microns to 0.7 cm. The largest clast observed is 0.7 × 0.5 cm and represents an unrecognized CM subtype meteorite; it contains large amounts of magnetite (3.6 vol%) and pyrrhotite–pentlandite (8.7 vol%). A detailed description of this clast will be presented elsewhere. Most of these xenolithic inclusions are consistent with CM characteristics, although <10% are akin to C3's based on mineral contents of spinels, melilite, grossular, perovskite, rhonite, nepheline and sodalite. Many of the C3 clasts contain phyllosilicates in replacement of olivine and pyroxenes so that the less-alterable minerals are remnant 'islands' in an altered matrix of phyllosilicates, which are indistinguishable from those in the host meteorite. The observation offers strong support to the *in situ* alteration model of CM phyllosilicates.

Phyllosilicates

Phyllosilicates collectively represent the most abundant mineral type in CM meteorites (of those studied). Laboratory investigations show that at least four phyllosilicates occur in Murray and three in Nogoya. Adequate identification in terms of terrestrial phyllosilicate nomenclature is precluded by the unique composition and environmental conditions of the meteorite phyllosilicates, slight compositional changes and thermal events. Therefore, it is not surprising that meteoritic phyllosilicates possess their own unique characteristics. For convenience, we refer to the various CM phyllosilicates in terms of their closest terrestrial mineral analogs.

Phyllosilicates in Murray were separated into 4 groups by petrographic characteristic, although a considerable compositional range exists for each of the phyllosilicates. The compositions given in Table 1 are only an attempt to compositionally characterize what we think are distinct phyllosilicate species based partly on petrographic and X-ray diffraction data. Murray matrix consists mainly of these phyllosilicates and intimately mixed PCP on micron to submicron scale so that accurate microprobe analyses are difficult. Samples of the phyllosilicates in Table 1 were analyzed with the microprobe and these compositions (element ratios) were confirmed with scanning electron microscope energy dispersive analysis (SEM–EDA). Despite the small electron beam of the SEM (<200 Å), there was possible contamination by S from PCP and observed sulfides $< 1\,\mu$ in size, because the electron-beam-excited sample volumes may reach $1–2\,\mu$ dia. Many small sulfide particles have been observed (SEM) to cling to phyllosilicate grains. Most sulfur and some Fe and Ni in the analyses are probably not coordinated in the phyllosilicate structure. The same problem exists for C by microprobe analyses; thus, it is not included in the analyses totals. In some phyllosilicate regions of thin sections where PCP appears to be admixed in low abundance, the C concentration is <0.67%. Almost all (>90%) of the phyllosilicate separates from Murchison contained about 2% C (CHANG *et al.*, 1978), a concentration comparable to the whole rock value. MCKEE and MOORE (1979) and MACKINNON and BUSECK (1979) have reported coatings of amorphous material, which may be carbonaceous, on phyllosilicate obtained from CM matrices, which suggests that C may be more abundant in PCP phases than in phyllosilicates.

Table 1. Electron microprobe analyses of phyllosilicates in CM meteorites.

	PHYLLOSILICATES							
	Fe types							Mg type
	Jodzie* (Bulk)	Orgueil†	Nogoya	Murray				Nogoya
				Dark green	Yellow	Light green to brown	Gray	
SiO_2	32.1	33.87	33.5	22.8	22.6	36.2	37.2	38.4
Al_2O_3	2.85	2.72	3.3	5.3	2.4	3.1	3.2	2.1
TiO_2	0.09	0.04	0.08	0.04	0.10	0.07	0.05	0.15
Cr_2O_3	0.65	0.56	0.23	0.08	0.25	0.28	0.32	0.30
"FeO	19.0	10.13	25.3	58.1	51.1	23.0	20.9	13.8
Fe_2O_3	10.0	11.26						
MgO	18.0	19.62	16.8	2.2	7.9	20.1	19.0	33.0
CaO	1.15	0.16	0.28	0.17	0.35	0.19	0.24	0.15
MnO	0.11	0.21	0.20	0.58	0.27	0.19	0.10	0.15
Na_2O	0.03	0.32	0.33	0.05	0.09	0.10	0.20	0.24
K_2O	0.03	0.08	0.40	0.06	0.10	0.13	0.24	0.27
P_2O_5	0.39	0.35	0.17	<0.05	0.13	0.16	0.16	0.15
S	0.92	5.60	2.5	0.62	1.28	1.9	1.2	1.57
NiO	1.56	2.51	1.9	0.05	1.30	1.47	1.97	1.54
C‡	[4.0]	n.d.	6.1	6.4	7.2	3.0	8.3	<0.08
H_2O§	12.0	10.0	10.0	10.0	10.0	10.0	10.0	10.0
Sum	100.00	97.41	95.04	100.05	97.87	96.89	94.78	101.82

* Defocused beam analysis of bulk separated phyllosilicates (Bunch *et al.*, 1979): 1/3 FeO converted to Fe_2O_3, 12% assumed H_2O; normalized to 100%. Carbon not included.

† Kerridge (1976).

‡ Carbon not included in totals.

§ H_2O assumed to be 10%.

X-ray diffraction and electron diffraction of the phyllosilicates separated by density and settling techniques suggest that light green to brown, yellow and gray (colors in cross polarized light) varieties are probably septachlorites or Fe-serpentines. The d_{001} spacing is about 7.3 Å, which corresponds to septachlorites or serpentines. Compositional data and earlier conclusions by Fuchs *et al.* (1973) on the identification of Murchison phyllosilicates favor septachlorites, possibly chamosites. The dark-green phyllosilicate is also consistent with septachlorites although composition and diffraction data suggest that it is closest to cronstedtite ($SiO_2 = 19.0\%$; $FeO + Fe_2O_3 = 70\%$). Low Al_2O_3 contents of all phyllosilicates make further identification useless because site occupancies and structural characteristics are generally unknown. We prefer the classification of these phyllosilicates as Fe-serpentines based on the low Al and high Fe contents, which is consistent with Moody's classification scheme (Moody, 1976) and recent findings by McKee and Moore (1979). The four phyllosilicates in Murray will henceforth be referred to as dark-green, yellow, brown and gray Fe-serpentines.

Figure 3 shows a possible metacolloidal development of three Fe-serpentines and PCP. Gray Fe-serpentine developed from neighboring olivine (Fo_{85}) with additional Fe and Ni coming from included metal or sulfides. The remaining Fe-serpentines and PCP probably formed from alteration of ferromagnesian silicates, metal, and sulfides. This sequential development shows enrichment of S, C, Mn, Fe and Ni and depletion of Si, Cr, alkalis and P from the earliest formed gray Fe-serpentine through yellow Fe-serpentine to PCP; the dark-green Fe-serpentine is inconsistent with this compositional trend. Examples of this kind are common in Murray and are similar to alteration textures in water-saturated terrestrial mafic rocks. (See numerous initial reports of the JOIDES Deep Sea Drilling Program). McKee and Moore (1979) reported that their high resolution TEM studies of Murray and Nogoya Fe-serpentine indicate a close similarity with serpentine minerals synthesized hydrothermally from olivine (Yada and Iishi, 1977).

The most abundant phyllosilicate in Murray is the brown Fe-serpentine rather than cronstedtite, as we reported earlier (Bunch and Chang, 1978). This conclusion is based on petrographic observations, SEM-EDA analyses of phyllosilicate particles, and chemical balance of analyzed bulk samples of Jodzie and Murray phyllosilicates. Ultrathin section observations coupled with microprobe analyses indicate that brown Fe-serpentine is most abundant, followed by green, yellow and gray. Two hundred and thirty-eight phyllosilicate particles of Murchinson were analyzed by the SEM-EDA method and of these, 110 are similar to brown Fe-serpentine, 77 to green, 36 to yellow and 15 to gray. A bulk analysis of separated but mixed phyllosilicates from Jodzie is given in Table 1. If most of the other CM chondrites are mineralogically similar to Jodzie, then the order of phyllosilicate abundance is consistent with that of Murray. The Fe/Mg ratio of 1.4 for Jodzie mixed phyllosilicates requires an abundant phyllosilicate of either the brown or gray variety and lesser amounts of the more iron-rich phyllosilicates. We caution against the future use of matrix bulk analyses for defining matrix mineralogy because the results shed little light on the complex nature of mixed, extremely fine-grained components. These analyses can be used for comparison with other meteorite matrices as shown in Table 1. Jodzie bulk phyllosilicates show good compositional agreement with Orgueil phyllosilicates (Kerridge, 1976) with the exception that Jodzie has higher Fe content.

Poorly characterized phases (PCP)

Fuchs *et al.* (1973) described and analyzed amorphous 'Fe-S-O' phases in Murchison that they termed 'poorly characterized phases.' Ramdohr (1973) also described a similar phase 'Fe-C-O' in three CM meteorites. McSween and Richardson (1977) suggested that an Fe-Ni-S-O phase is present in all CM meteorites and, with various

Table 2. Electron microprobe analyses of poorly characterized phases

POORLY CHARACTERIZED PHASES (PCP)					
	Jodzie	Murray			Nogoya
		Type I	Type II	Type III	
SiO_2	0.26	0.86	8.1	11.5	22.0
Al_2O_3	0.10	0.52	1.1	2.2	1.5
"FeO"	54.0	51.5	62.0	54.4	42.0
Cr_2O_3	---	3.3	---	---	---
MgO	0.56	0.70	2.5	4.6	10.2
CaO	0.04	0.05	0.10	0.10	0.04
K_2O	0.36	0.63	0.20	0.10	0.20
P_2O_5	<0.02	0.84	<0.02	<0.02	0.05
Ni	5.8	7.7	6.4	1.2	0.03
S	20.4	17.0	8.8	10.4	12.0
C	2.4	2.2	5.0	8.4	0.2
Sum	83.92	82.0	94.20	92.90	88.22
	Fibrous to platey	Massive	Fibrous and Needles	Granular to formless	Small grains <3 μ

proportions of phyllosilicates, is a low-temperature condensation product in the solar nebula. We find that Jodzie, Nogoya, Murchison and Murray contain amorphous or poorly crystallized phases high in Fe, Ni, O, C, S and in some morphological forms, Si (Table 2). This material is massive, fibrous, needle-like, granular, or formless and is consistent with the description given by Fuchs *et al.* (1973). Composition of the massive form in Murray is similar to that described in Murchison by Fuchs *et al.* (1973), although they did not analyze other forms. In the three forms, we find a correlation between morphological form and composition (Table 2). A high S content appears to be associated with low Si, Al and Mg. Several varieties are shown in Figs 4 and 5. Of the three forms, Type III (granular and formless) is the most abundant. Because these phases lack the virtues of *bona fide* minerals, we collectively refer to them by the acronym PCP.

All forms of PCP are black and opaque in standard thin sections and show a range of color from gray to beige in reflected light. The degree of anisotropy varies with form; massive forms show the least and fibrous the most. Compositions of PCP are puzzling; analytical sums are always less than 100%; some barely exceed 80%. Oxygen measured by the microprobe exceeds amounts necessary for oxide assignments even if we assume that all iron is Fe^{3+} so that a portion of the missing components could be water or S that is oxidized to SO_3. This appears to be a plausible explanation if we convert S in the analyses of Table 2 to SO_3 which bring the sums to nearly 100%. In either case, PCP does not appear to have stoichometry.

The coexistence and inverse relationship of S and Si in PCP were also puzzling until we observed PCP in ultrathin sections. Apparently, Si-bearing PCP consists of two phases: (1) an Fe-rich phyllosilicate phase and (2) a phase rich in Fe, Ni, S and C (Fig. 6). The two-phase mixture was confirmed by TEM and SEM observations. Measurements of accurate compositions for these phases is hampered by their fine-grain size. Figure 7 shows an SEM micrograph of PCP and X-ray images of Fe, S and Mg. Iron and S are clearly enriched in the outer margins; Mg is concentrated in the core areas (phyllosilicate), despite the rather low abundance (<3 wt%) of Mg and lower sensitivity of SEM analysis.

We believe that PCP is important in understanding the formation and evolution of CM mineral matrices and the chemical evolution of C and S in meteorites. The PCP textures and association with other phases strongly suggest an *in situ* alteration origin for most matrix components as opposed to nebular condensation. Because some of the sulfide and PCP veins transcend many phase boundaries, it seems improbable that alteration occurred anywhere other than in the matrix. Many pseudomorphed clasts and chondrules consist entirely of PCP and phyllosilicates. Moreover, many matrix mineral grains, especially calcite, show PCP needles and fibers penetrating into or encrusting on the grain margins (Figs 4 and 5). PCP contains rather high amounts of carbon. Carbon in the most abundant Type III PCP averages about 8 wt% in Murray or about 4 times greater than bulk contents. After phyllosilicates, PCP is the second most abundant phase in the matrix and may account for up to 30 vol% of matrix components. The amount of C associated with PCP and phyllosilicate phases can account for most of the carbon in Murray and, presumably other CM meteorites. The content of C as well as the abundances and types of organic matter and noble gases in CM meteorites seem to be interrelated, a point more fully developed in the discussion section.

Sulfides

Pyrrhotite, pentlandite, 'Q' sulfide and unusual alteration sulfides were found in Murray, in addition to sulfur-bearing PCP. Of these sulfides, pyrrhotite is the most abundant (61%) compared with pentlandite; both show no unusual compositions (Table 3) although those grains in advanced stages of alteration may contain large amounts of oxygen (up to 14 wt%) in their outer margins. An unusual primary sulfide was found in all CM meteorites and is similar to the predicted 'Q' phase composition of a putative noble gas-rich phase in meteorites (Lewis *et al.*, 1975). 'Q' sulfide refers to observed sulfides compositionally similar to a putative gas-rich trace phase in meteorites that is insoluble in HF and HCl but dissolves in oxidizing acids releasing its gas contents (the 'Q'-effect: Lewis *et al.*, 1975; cf. Reynolds *et al.*, 1978 and Ott *et al.*, 1979); its existence and Fe, Ni, Cr-sulfide composition have been inferred from analyses of HNO_3 etchants of meteoritic HF–HCl insoluble residues. Identification of sulfide grains with Q-like compositions neither confirms nor denies the gas-retention and acid solubility properties attributed to the inferred Q-sulfide. Microprobe analyses of 11 small grains ($<10\,\mu$) within the matrix and 4 in forsteritic aggregates show variable composition among the grains (Table 3) with respect to Cr, which appears to have an inverse relationship with Ni. Major elements are homogeneously distributed (Fig. 8) and all observed grains show little evidence of alteration. We are not able to obtain complete analyses for this phase

Table 3. Electron microprobe analyses of primary sulfides in Murray and Nogoya

	Murray			Nogoya
	Pyrrhotite (45)*	Pentlandite (30)*	"Q" sulfide (13)*	"Pentlandite" (17)*
Fe	61.4 - 63.0	31.1 - 45.1	29.1 - 30.4	31.1 - 36.6
S	35.6 - 36.9	32.8 - 33.3	23.9 - 25.9	30.8 - 33.1
Ni	0.04 - 1.24	20.7 - 34.0	18.0 - 26.9	23.7 - 33.3
Cr	<0.02 - 0.21	<0.02 - 0.6	4.0 - 12.5	<0.02 - 0.14
P	<0.02	<0.02	3.1 - 4.0	<0.02 - 1.2
K	<0.02	<0.02	0.35 - 1.0	<0.02 - 0.9
Zn	<0.02	<0.02	<0.02	<0.02 - 22.3
O	<0.05 - 0.6	<0.05	<0.05 - 3.0	<0.05 - 1.2
C	<0.05 - 0.53	<0.05 - 0.03	<0.05	0.08 - 0.6

* Number of grains analyzed.

because the analytical sums of all detectable elements never exceed 92 wt%. Microprobe analyses show little or no oxygen or C. Unfortunately, the fine-grain size and low abundances preclude X-ray diffraction and isotopic analysis.

Calcite and magnetite

Calcite occurs in the matrix as small (few to 95 μ), mostly equant grains that have needles of PCP protruding into the grains (Fig. 4) or are commonly rimmed by PCP (Fig. 5). A few partly altered chondrules and aggregates have small amounts of calcite. Electron microprobe and SEM analyses show calcite to be free of any cations other than Ca.

Calcite–PCP textural relationships indicate paragenetic overlap with calcite formation preceding, although overlapping PCP crystallization. Calcite in Murray and Nogoya chondrules and aggregates is clearly an alteration–replacement product. Matrix calcite is also an alteration product as shown by its growth development with PCP. Carbonates are common alteration products in hydrothermally altered terrestrial ultramafic and mafic rocks. We conclude that calcite in CM meteorites formed in a similar manner and not as low-temperature nebula condensates.

Magnetite occurs as small (< 1–$3\ \mu$) spheres, equant grains (3–7 μ) and massive units. Microprobe analyses show only the presence of Fe and O. Although some magnetite may have formed as a low-temperature condensate, we suggest that most magnetite occurs as an alteration–reaction product, based on its mutual association with other alteration phases. Hematite and γ-Fe_2O_3 were identified in the matrix by X-ray diffraction.

NOGOYA ALTERATION CHARACTERISTICS

Nogoya is a unique meteorite that has received little attention in the past. Our samples of Nogoya, obtained through the courtesy of Professor Carleton Moore, show 95% alteration of precursor materials. The matrix is nearly completely altered to Mg-serpentine and Fe-serpentine (Figs 9 and 10). In addition SEM, microprobe analyses, X-ray diffraction, and TEM diffraction indicate the presence of Fe_3O_4, Fe_2O_3, γ-Fe_2O_3, possibly antigorite and brucite, PCP, gypsum, pentlandite, zincian pentlandite, calcite, epsomite and relic pyrrhotite, forsterite and Ca-poor and Ca-rich pyroxenes.

X-ray diffraction of phyllosilicates in Nogoya show that they are more structurally ordered than phyllosilicates in other CM meteorites. Moreover, they are also more compositionally homogeneous and better physically separated, which together suggest attainment of equilibrium. Phyllosilicate average analyses (Table 1) and example analyses in Fig. 10 show more MgO and less 'FeO' in both Mg-serpentine and Fe-serpentine compared with other CM analyses. Many small ($< 10\ \mu$) grains of pentlandite are dispersed in and around both phyllosilicates (Fig. 10); they have apparently formed from PCP during prolonged alteration with decreasing oxygen fugacity. Recrystallized margins around larger (30 μ) relic pentlandite contain up to 23 wt% Zn that has replaced Ni. Only relic Fe-free olivine and pyroxenes remain, apparently because they were more resistant to alteration.

The most remarkable textural characteristic of Nogoya is the complete replacement of chondrules and aggregates by phyllosilicates, PCP and spongy mosaic calcite (Figs 11 and 12); only 'spinach' remains. Several chondrules contain relic forsteritic olivine completely surrounded by fibrous Mg-serpentine.

Mesh or island textures are common for chondrules and aggregates that still contain relic primary olivine and pyroxene (Fig. 13a). Analyses of phyllosilicates that surround the olivine 'islands' in Fig. 13a are consistent with Fe-serpentine composition. In addition, they contain Cl, which is enriched over relic materials by a factor of 2–10. Petrographic and SEM observations of the phyllosilicates indicate the possible presence of antigorite, although no conclusive TEM electron diffraction has been obtained. Figure 13b shows a typical mesh or island alteration texture of a terrestrial altered ultramafic rock. Nogoya and, to a lesser degree, Cold Bokkeveld, contain a few cross-cutting veins filled with calcite, phyllosilicates, iron oxides, altered sulfides and possibly Fe-rich epsomite.

Other altered aggregates and chondrules in Nogoya contain granular magnetite surrounded by Fe-rich brucite and Fe-serpentine (Fig. 14a). The granular, wispy nature of magnetite is very similar to secondary magnetite in terrestrial serpentinite rocks (Fig. 14b). In addition, the aggregate in Fig. 14a (and in the enlarged view in Fig. 14c) shows an excellent development of colloform texture which is typical of crystallization in a liquid medium. SEM–EDA spectra (Figs 14d through 14f) show the composition of the various colloform layers. Layer *a* appears to be a Mg-rich phyllosilicate whereas layer *b* is Fe-rich. Note that layer *b* is also enriched in K and Cl, which amount to 0.5 and 1.8 wt%, respectively. Interiors of the colloform structures are poorly crystallized or amorphous (optically) and are Mg, Si-rich. Area *c* is possibly Fe-rich brucite; Si that is present is probably contamination from surrounding serpentine. Particles from this aggregate were excavated and analyzed by TEM electron diffraction. Most particles show a strong spacing at 7.3 Å, consistent with serpentine. Other particles show strong spacing at 4.9 and 2.4 Å, consistent with brucite.

Evidence for alteration in place is further strengthened by the textural relationship between matrix and an olivine clast shown in Fig. 15. This fractured grain is corroded by alteration mostly along fractures, which afforded easy access for alteration fluids or gases. The original grain boundary has been completely destroyed by advancing alteration and the grain has been partly replaced by phyllosilicates, iron oxide and calcite..

Abundances and distributions of C, N and S in Nogoya, and data on C isotopic composition are given in Table 4. Although values for total S and N are consistent with other CM meteorites, values of 5.2% for total C are higher than previously observed in any meteorite. Moreover, the bulk carbon isotopic composition (−21.8‰) shows the lowest measured $^{13}C/^{12}C$ ratio of any carbonaceous chondrite except for Karoonda (−25.2‰; BELSKY and KAPLAN, 1970), a C4 meteorite. Carbon in Nogoya shows isotopic affinities with C in ordinary chondrites (BOATO, 1954; BELSKY and KAPLAN, 1970). The amount and isotopic composition of C in the acid insoluble residues point to an indigenous origin for the isotopically light carbon rather than terrestrial contamination. Although these residues from carbonaceous meteorites usually exhibit isotopic compositions close to −16‰, several analyses of CI and CM meteorites yielded values ranging from −18 to −30‰ (KROUSE and MODZELESKI, 1970; NAGY, 1975). The abundance and isotopic composition of carbonate-CO_2 released by acid are not unusual and resemble those of Murchison (CHANG *et al.*, 1978) and other CM meteorites (SMITH and KAPLAN, 1970). The origin of the isotopically light CO, however, is not clear. Acid treatment of Murchison yields a similar CO component. The CO may be gas originally trapped within the meteorite matrix or a product resulting from a partial decomposition of organic matter during acid treatment, or both. In Table 4, the difference between carbon abundances in the whole rock and in the sum of carbonaceous components obtained by acid treatment indicates that water and acid soluble organic matter amounts to roughly 30% of the total carbon, in agreement with analyses of other CM and CI meteorites (HAYES, 1967; SMITH and KAPLAN, 1970). Substantial amounts (60–90%) of sulfur were also extracted by acid as might be expected if water-soluble sulfates were a major component. A small amount of SO_2 was also released, but no H_2S derived from a hydrolyzable sulfide was detectable at the 100-ppm level in either Nogoya or Murchison. KROUSE and MODZELESKI (1970) consistently observed the evolution of SO_2 during acid treatment of carbonaceous meteorites. The occurrence of the oxidized form of sulfur readily accounts for the 'excess oxygen' seen in PCP phases.

A few problems are evident regarding classification, formation and evolution of Nogoya. Relic textures and relic mineralogy are consistent with CM meteorites; however, high bulk carbon (5.2 wt%), relatively high ^{12}C enrichment in the carbon isotopes, and depleted amino acid contents are unusual. Although the 19th century measurements (MASON, 1963) of C (1.62%) and S (3.27%) relegated Nogoya to the CM classification, the data in Table 4 and our mineralogical–petrographic observations place it in a unique category.

DESCRIPTION AND SIGNIFICANCE OF HALOES

'Haloes' are fine-grained spherical to elliptical zones of matrix material that rim most aggregates and a few chondrules. This feature was noticed by FUCHS

Table 4. Abundances of C, N and S, and isotopic composition of C in Nogoya*

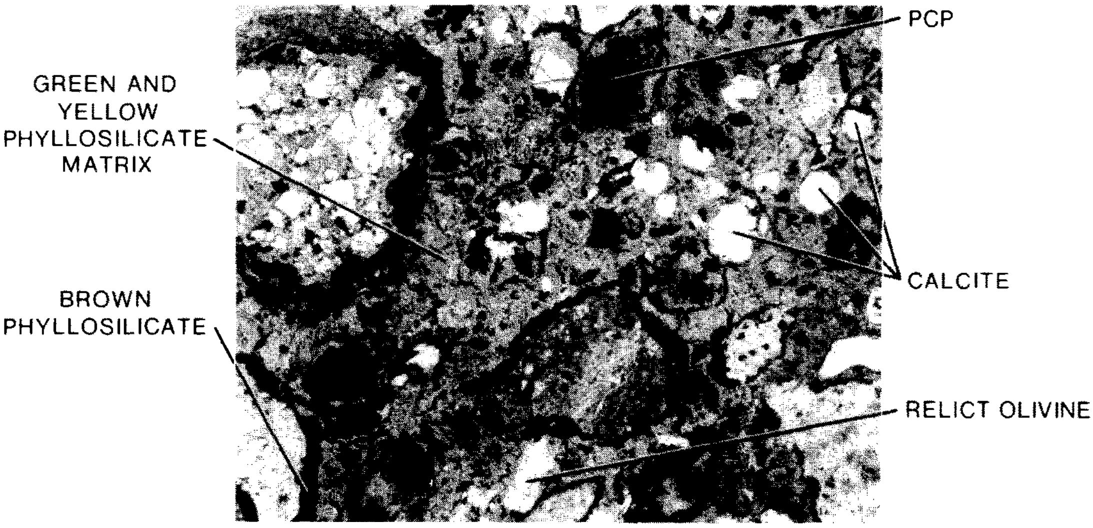
Source	Sample wt (mg)	C (‰)*	$\delta^{13}C_{PDB}$ (‰)	S (‰)*	N (ppm)*
Whole rock	19.4	5.18	-21.7	†	1037
Whole rock	18.9	5.25	-21.9	2.35	758
Whole rock	34.1	(4.27)‡	(-20.6)‡	2.61	1039
Carbonate - CO_2§		0.246	+37.2		
Carbon monoxide§		0.057	-29.9		
Sulfur dioxide§				0.023	
HCl-Insoluble Residue	6.14	3.41	-25.6	0.288	458
HCl-Insoluble Residue	3.25	3.76	-24.6	0.410	970
HCl-Insoluble Residue	11.8	3.85	-25.0	0.973	1036

* Abundances calculated for whole meteorite.

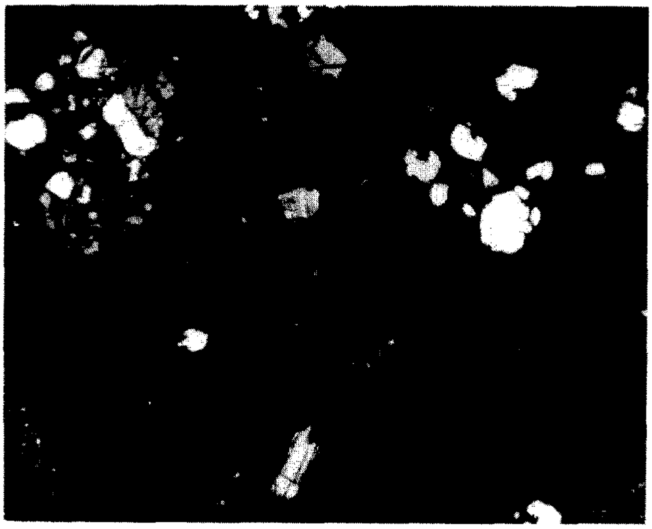
† Sample lost.

‡ Incomplete recovery of carbon.

§ Freed by heating a 19.3-mg sample in 6 N HCl at 120°C.



PHOTOMICROGRAPH
(PLANE LIGHT)



PHOTOMICROGRAPH
(CROSSED POLARIZERS)

Fig. 1. Example of altered matrix in Murray.

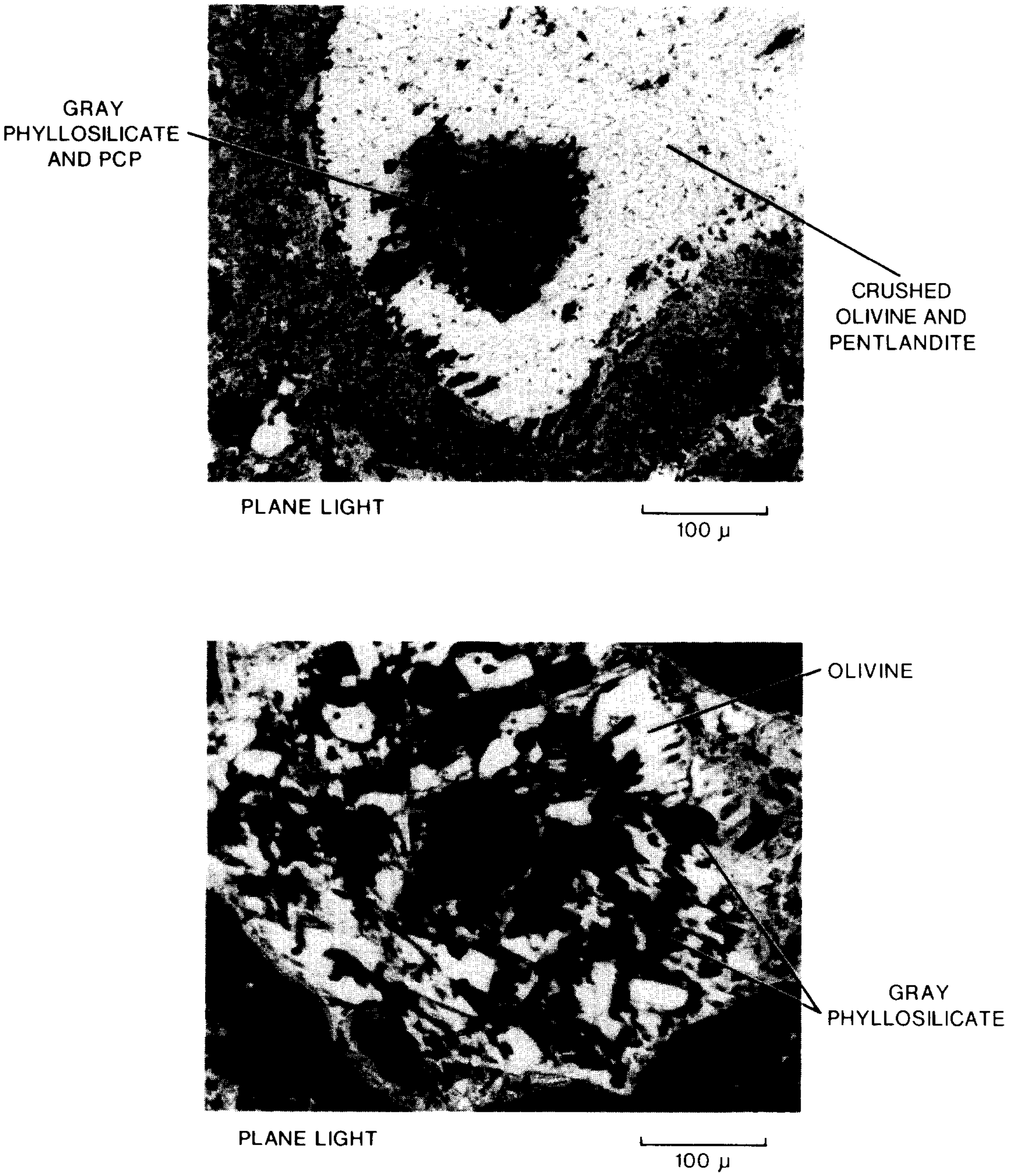
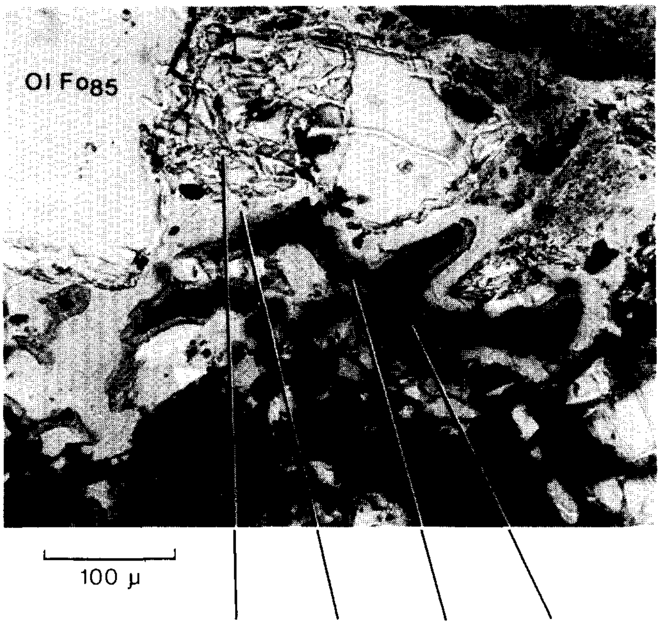
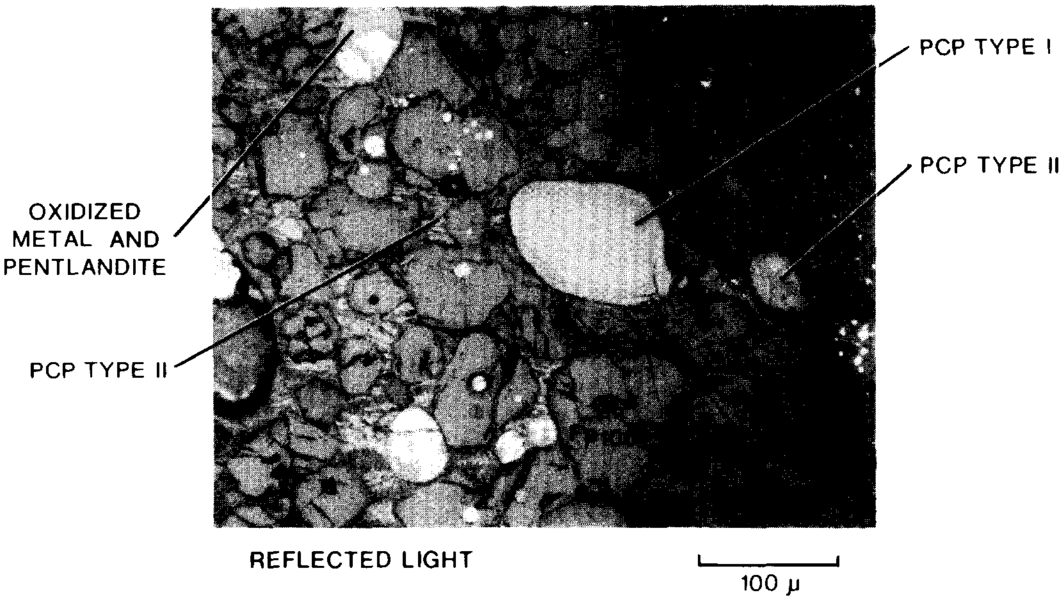


Fig. 2. Examples of incipient alteration in Murray high-temperature aggregates.

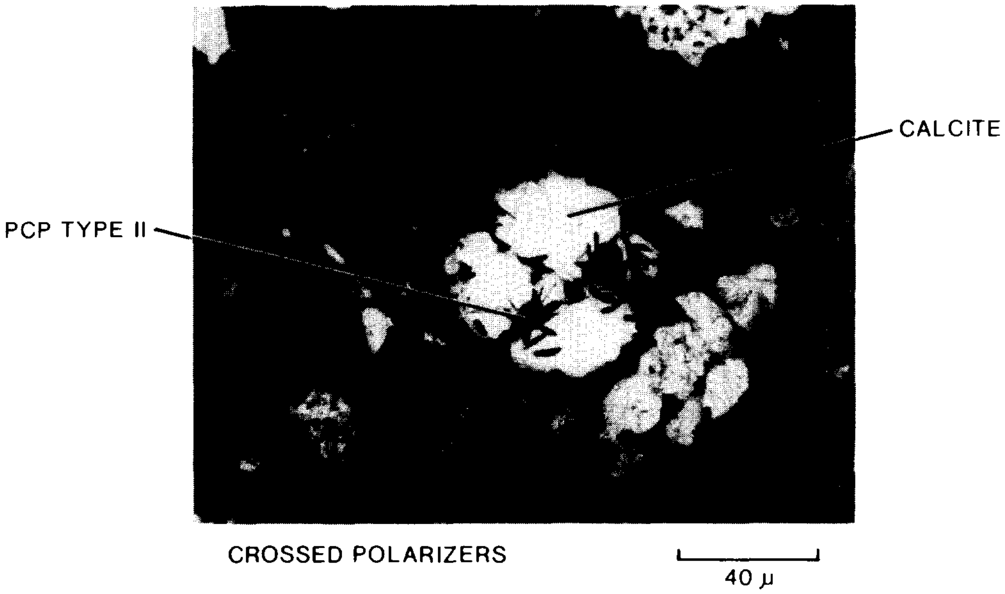


		GRAY	YELLOW	GREEN	PCP
SiO_2	=	35.6	20.8	21.1	11.5
Al_2O_3	=	3.0	2.2	5.2	2.2
Cr_2O_3	=	0.32	0.25	0.08	0.8
'FeO'	=	19.4	46.3	56.8	54.4
MgO	=	17.4	7.6	2.1	4.6
CaO	=	0.24	0.35	0.17	0.10
MnO	=	0.10	0.27	0.56	—
Na_2O	=	0.20	0.09	0.05	—
K_2O	=	0.24	0.10	0.06	0.10
P_2O_5	=	0.16	0.13	<0.04	<0.04
NiO	=	0.91	1.23	0.05	1.68
S	=	0.8	1.2	0.62	10.4
C	=	<1.0	6.3	4.2	8.4

Fig. 3. Colloidal alteration sequence (colloform texture) in Murray.

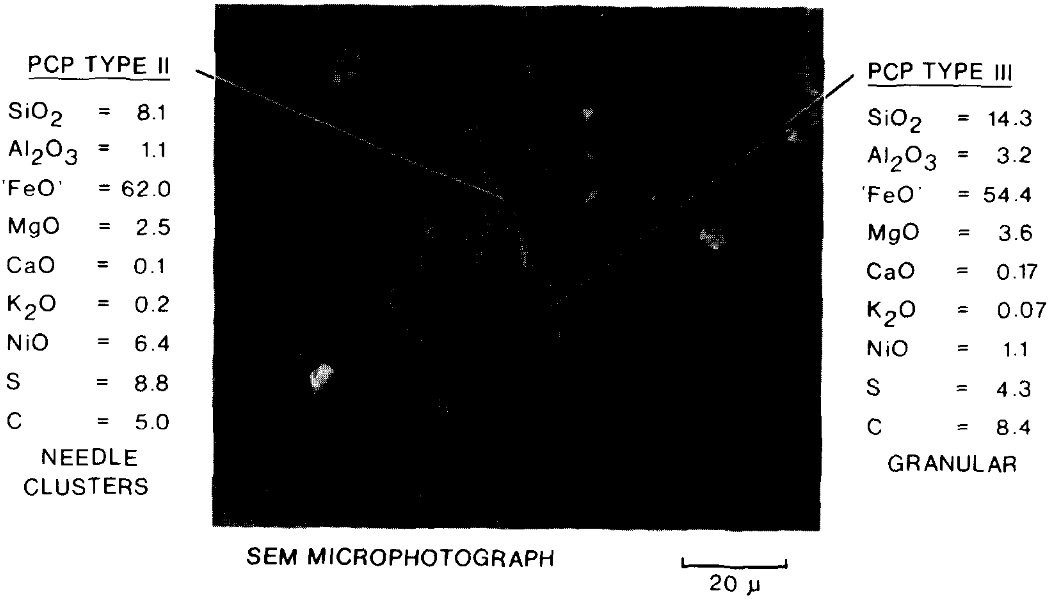


REFLECTED LIGHT

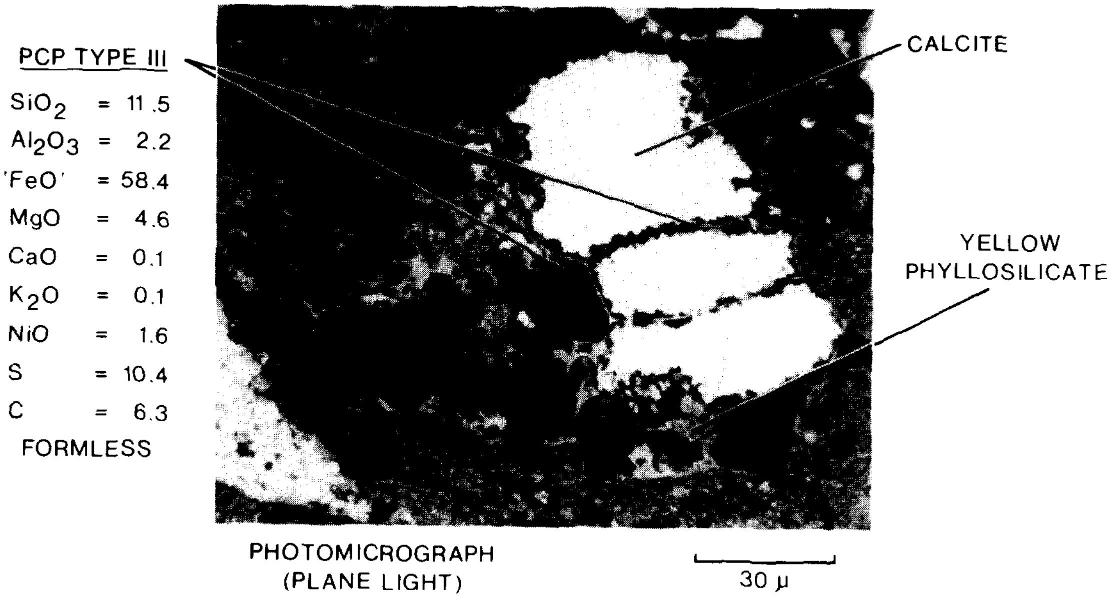


CROSSED POLARIZERS

Fig. 4. Examples of PCP textures and compositions in Murray.



SEM MICROPHOTOGRAPH



PHOTOMICROGRAPH
(PLANE LIGHT)

Fig. 5. Examples of PCP textures and associated phases in Murray.

Fig. 6. PCP (dark) intergrown with phyllosilicate (center portions) as seen in an ultrathin section (8 μ) of Murray (diameter of 3-grain cluster is 10 μ).

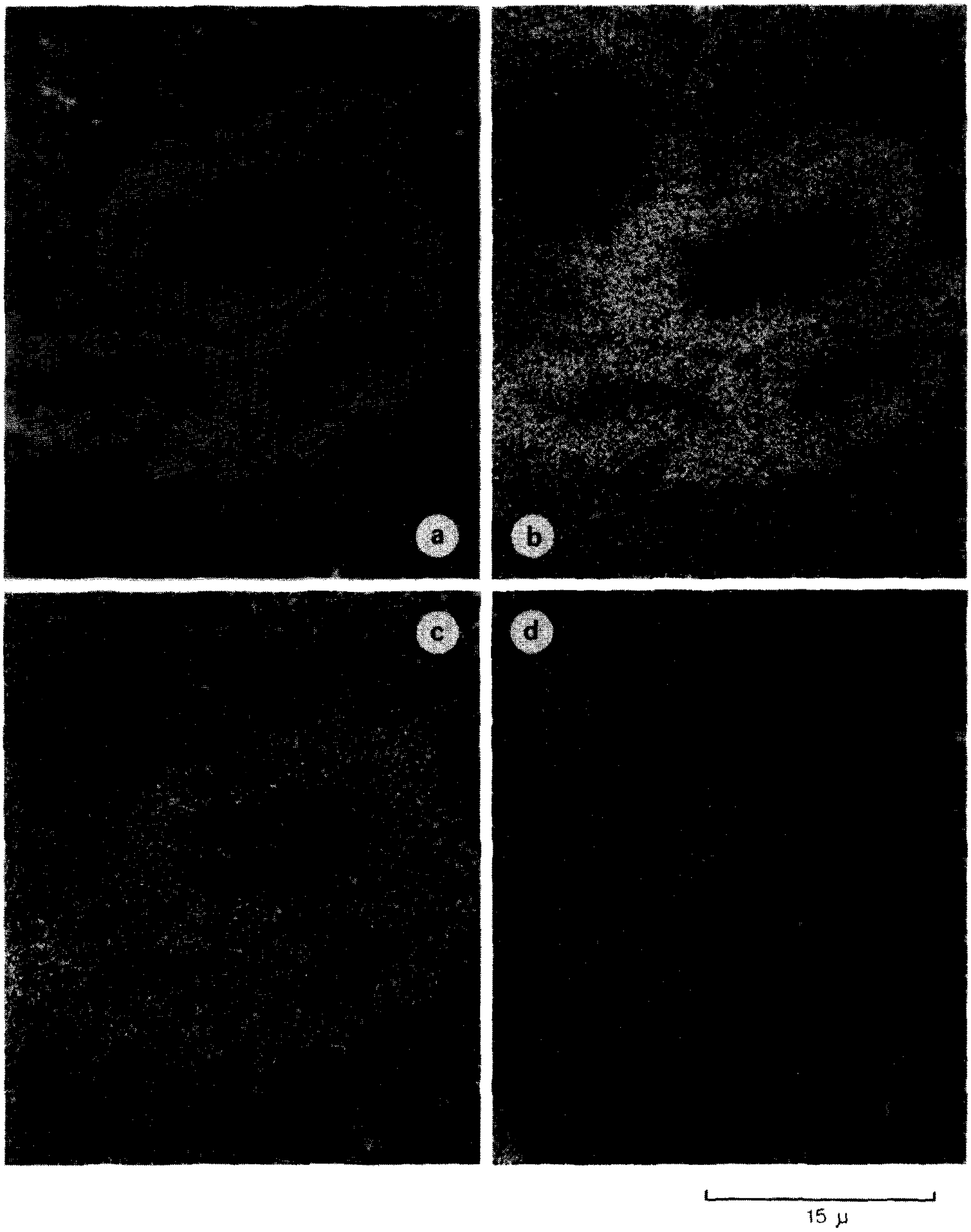


Fig. 7. (a) SEM photograph of PCP-phyllosilicate clusters shown in Fig. 6. (b) Fe X-radiation image of *a*. (c) S X-radiation image of *a*. (d) Mg X-radiation image of *a*.

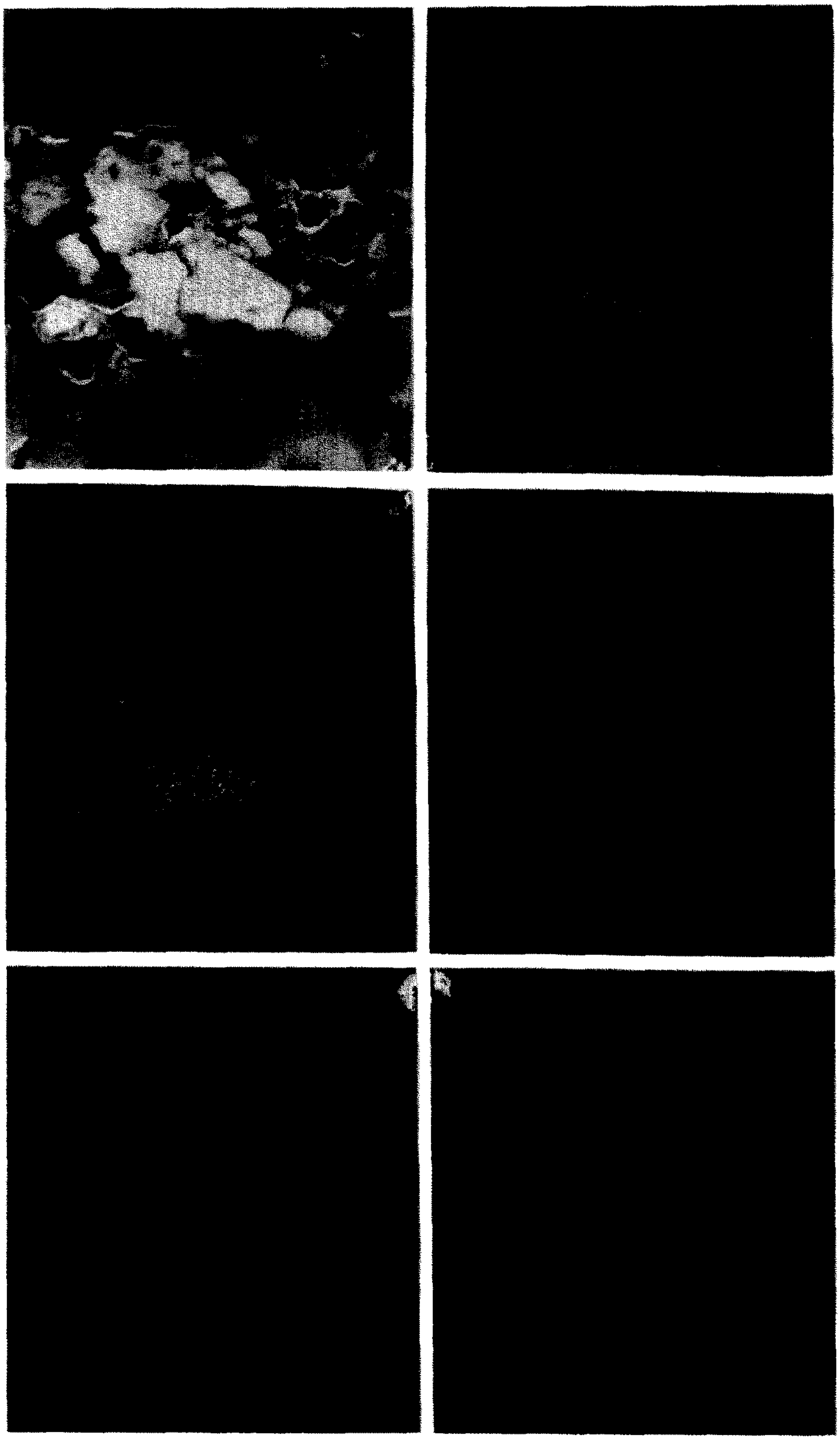


Fig. 8. 'Q' sulfide. (a) SEM photograph of 'Q' sulfide in Murray. Field width = 15 μ. (b) Fe X-radiation. (c) S X-radiation. (d) Cr X-radiation.

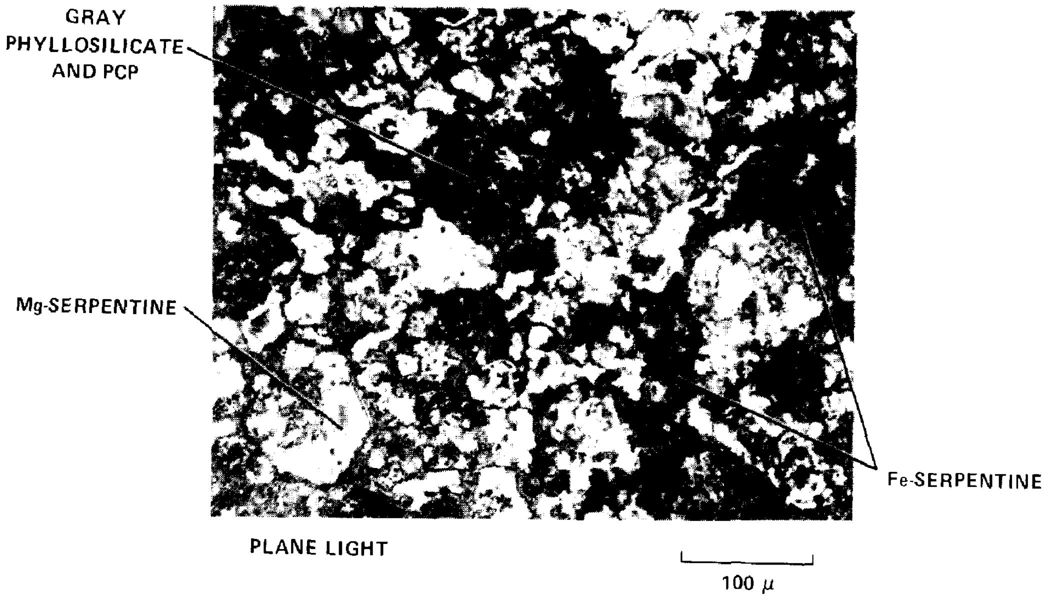
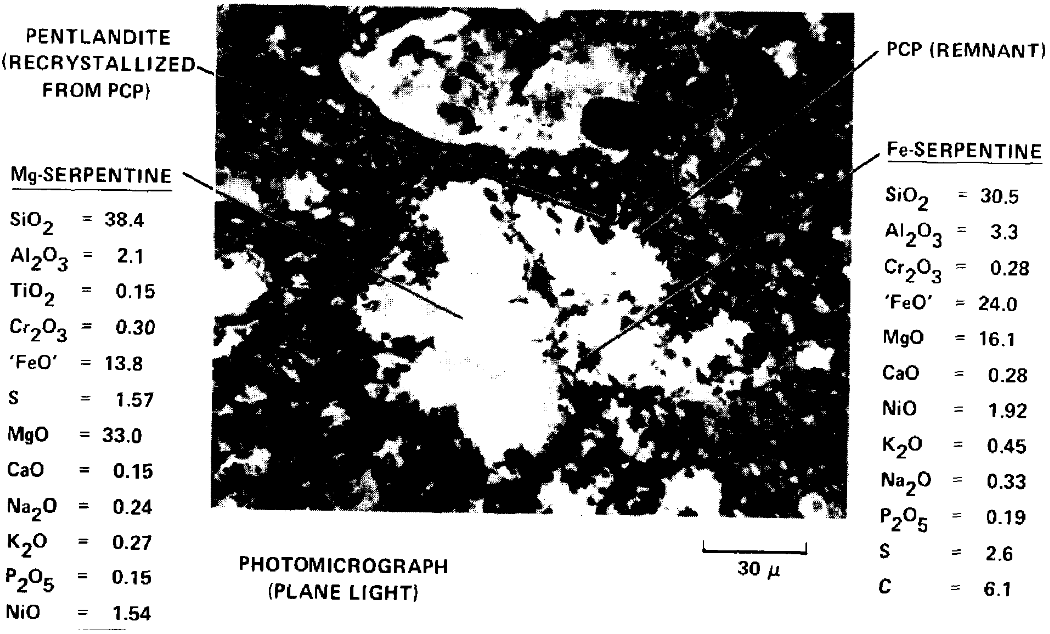


Fig. 9. Typical matrix texture of Nogoya.



Mg-SERPENTINE

SiO_2 = 38.4
Al_2O_3 = 2.1
TiO_2 = 0.15
Cr_2O_3 = 0.30
'FeO' = 13.8
S = 1.57
MgO = 33.0
CaO = 0.15
Na_2O = 0.24
K_2O = 0.27
P_2O_5 = 0.15
NiO = 1.54

Fe-SERPENTINE

SiO_2 = 30.5
Al_2O_3 = 3.3
Cr_2O_3 = 0.28
'FeO' = 24.0
MgO = 16.1
CaO = 0.28
NiO = 1.92
K_2O = 0.45
Na_2O = 0.33
P_2O_5 = 0.19
S = 2.6
C = 6.1

Fig. 10. Nogoya phase relationships and compositions.

Mg-SERPENTINE

SiO_2 = 34.1
Al_2O_3 = 2.6
Cr_2O_3 = 0.05
'FeO' = 18.6
MgO = 24.5
CaO = 0.16
NiO = 0.06
K_2O = 0.32
Na_2O = 0.05
P_2O_5 = <0.03
S = 0.07
C = 2.1

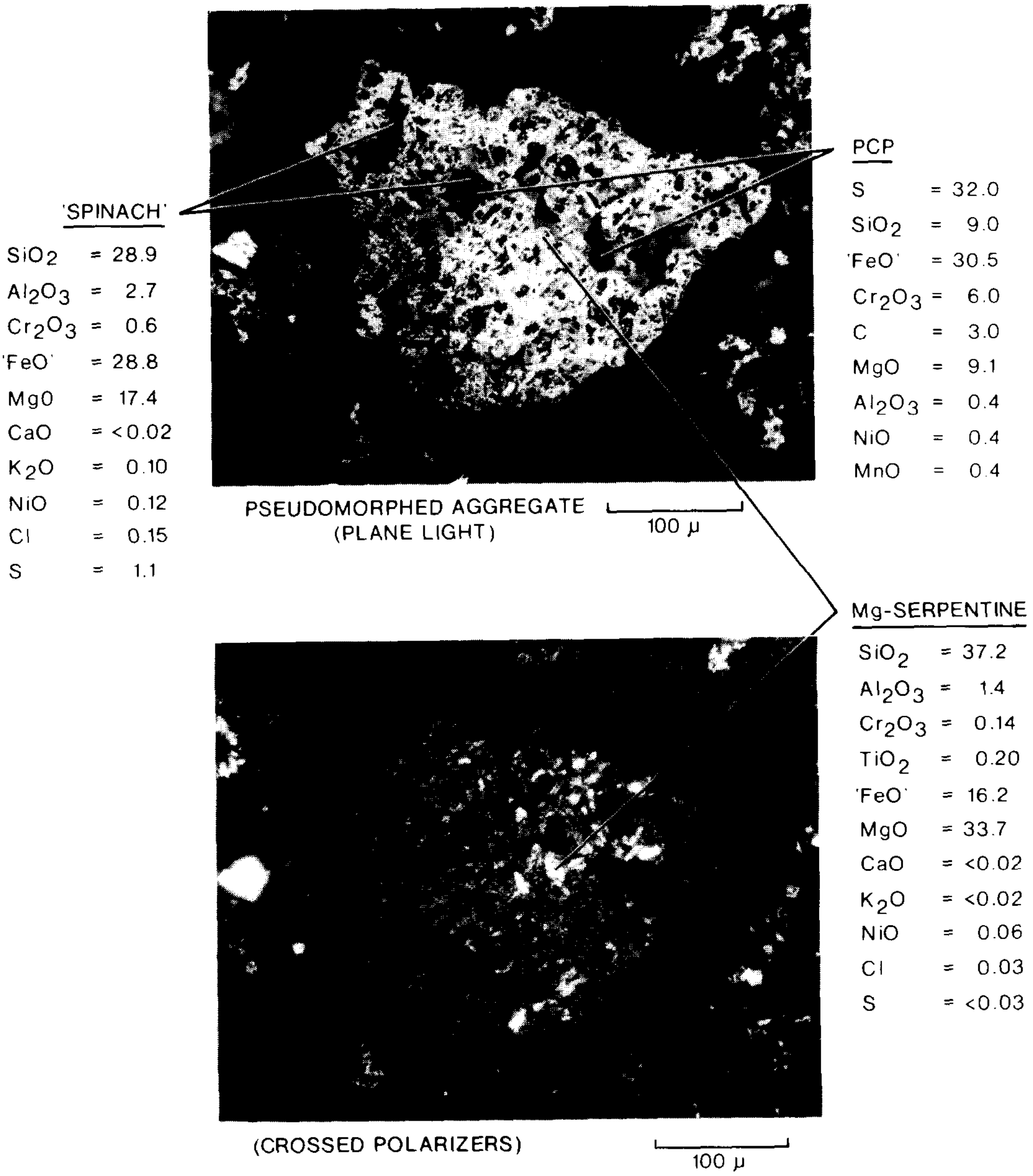
'SPINACH'

SiO_2 = 32.0
Al_2O_3 = 3.4
Cr_2O_3 = 0.19
'FeO' = 34.2
MgO = 15.5
CaO = 0.28
NiO = 0.15
K_2O = 0.41
Na_2O = 0.16
P_2O_5 = 0.08
S = 0.23
C = 1.2

PSEUDOMORPHED CHONDRULE
PHOTOMICROGRAPH
(PLANE LIGHT)

PHOTOMICROGRAPH
(CROSSED POLARIZED LIGHT)

Fig. 11. Nogoya chondrule pseudomorphed by calcite and phyllosilicates.



'SPINACH'

SiO_2 = 28.9
Al_2O_3 = 2.7
Cr_2O_3 = 0.6
'FeO' = 28.8
MgO = 17.4
CaO = <0.02
K_2O = 0.10
NiO = 0.12
Cl = 0.15
S = 1.1

PCP

S = 32.0
SiO_2 = 9.0
'FeO' = 30.5
Cr_2O_3 = 6.0
C = 3.0
MgO = 9.1
Al_2O_3 = 0.4
NiO = 0.4
MnO = 0.4

Mg-SERPENTINE

SiO_2 = 37.2
Al_2O_3 = 1.4
Cr_2O_3 = 0.14
TiO_2 = 0.20
'FeO' = 16.2
MgO = 33.7
CaO = <0.02
K_2O = <0.02
NiO = 0.06
Cl = 0.03
S = <0.03

Fig. 12. Nogoya aggregate pseudomorphed by PCP and phyllosilicates.

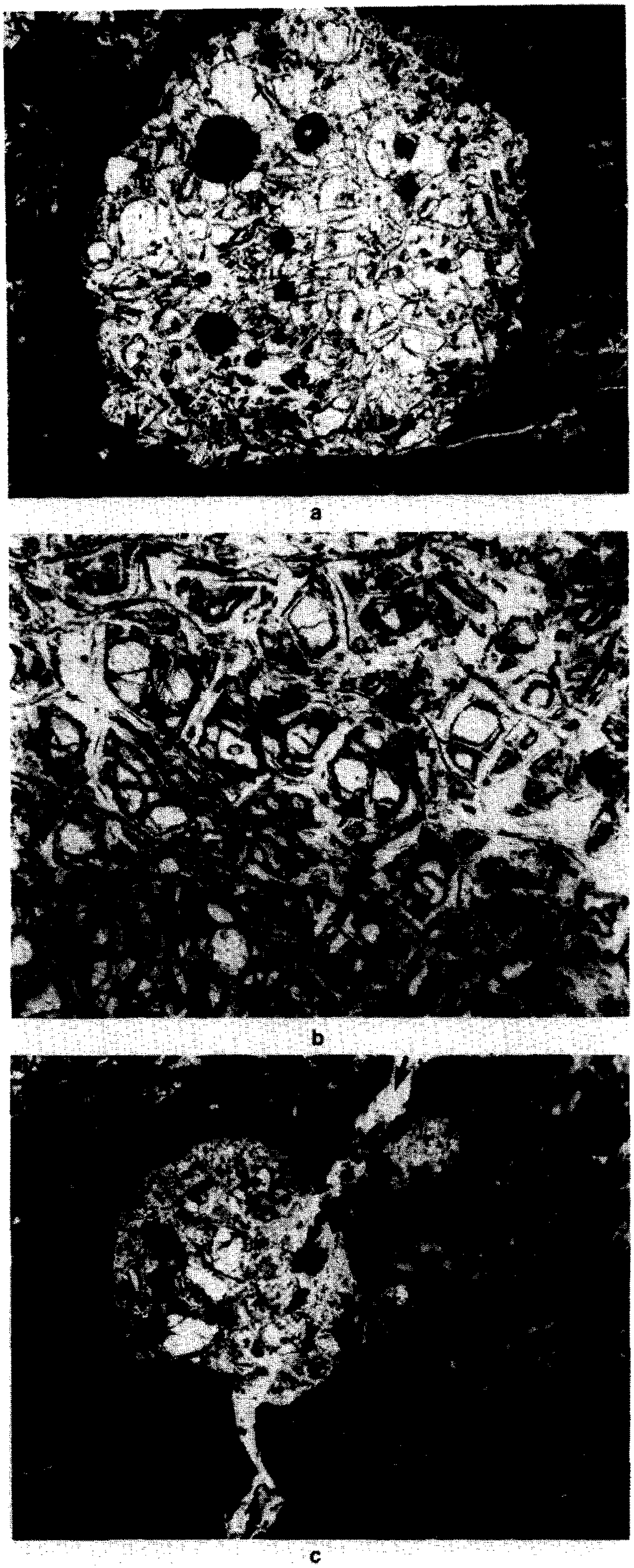


Fig. 13. (a) Nogoya chondrule consisting originally of three olivine grains now partially altered to fibrous and massive phyllosilicates in a manner similar to terrestrial serpentinite mesh or island textures. Dark rounded areas within the chondrule are brownish iron oxides and altered sulfides. Small dark grains are PCP. Plane light, field width = 1.8 mm. (b) Example of a terrestrial serpentine mesh texture showing island of relic olivine surrounded by antigorite and lizardite serpentines. Del Norte County, California. Plane light, field width = 1.8 mm. (c) Nogoya. Cross-cutting vein (small arrows) consisting of calcite, phyllosilicates, brownish iron oxides and Fe-epsomite that transects an aggregate of relic olivine (large arrow) surrounded by phyllosilicates. Plane light, field width = 1.8 mm.

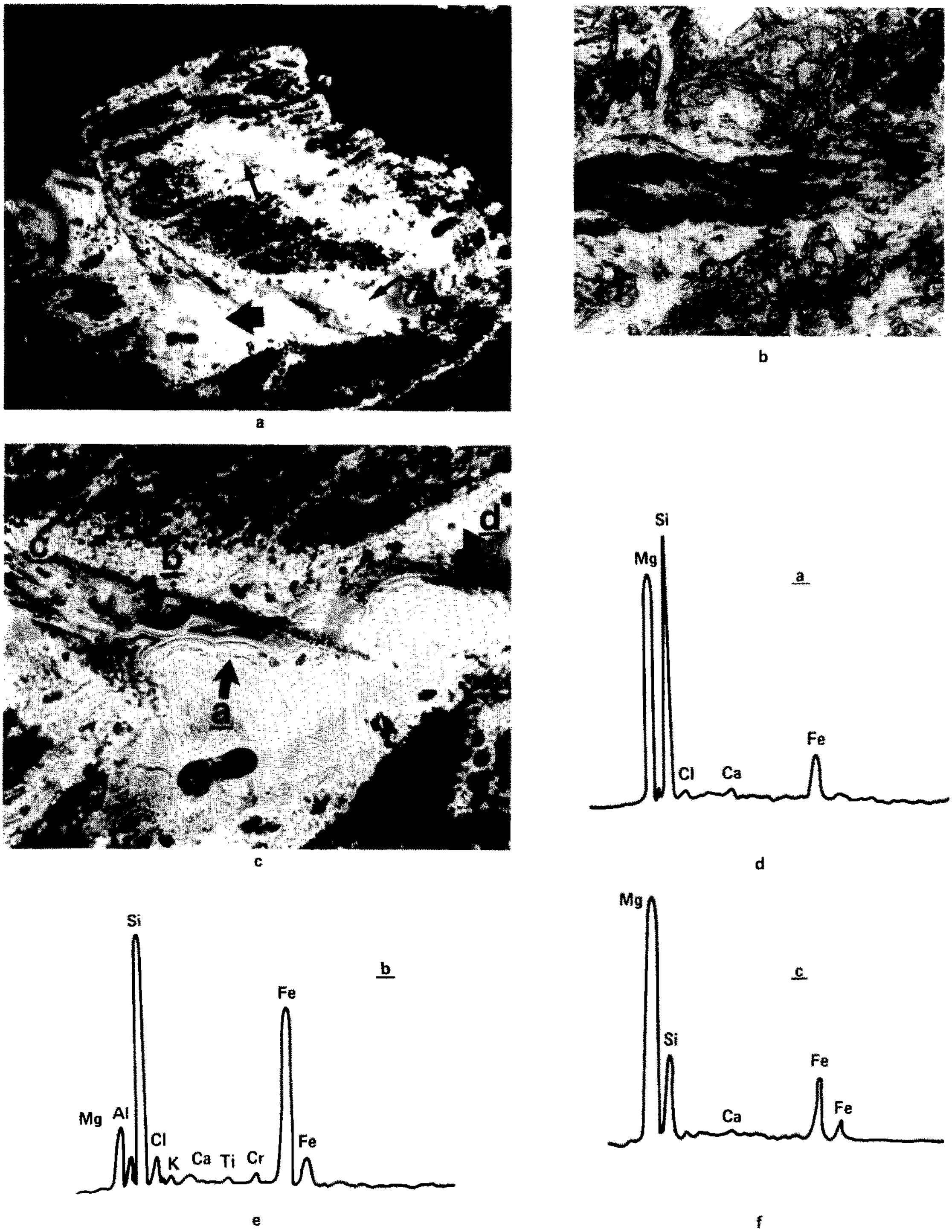


Fig. 14. (a) Nogoya. Aggregate altered to Mg-phyllosilicates and amorphous colloidal material (light color), magnetite (dark granular patches) with interstitial Fe-brucite, and Fe-serpentine (intermediate shades). Arrows point to colloform areas, large arrow points to enlarged area (c). Plane light, field width = 1.8 mm. (b) Terrestrial serpentinite that shows similar structured and granular development of secondary magnetite, which is surrounded by serpentine. Plane light, field width = 1.8 mm. (c) Enlarged area from (a) that shows colloform textural, plane light, field width = 0.45 mm. Area a is Mg-phyllosilicate (SEM–EDA X-ray emission spectrograph *a*, Fig. 14d). Area *b* is Fe-serpentine (SEM–EDA X-ray emission spectrograph *b*, Fig. 14e). Area c is probably Fe-brucite (SEM–EDA X-ray spectrograph *c*, Fig. 14c). Area *d* relic olivine.

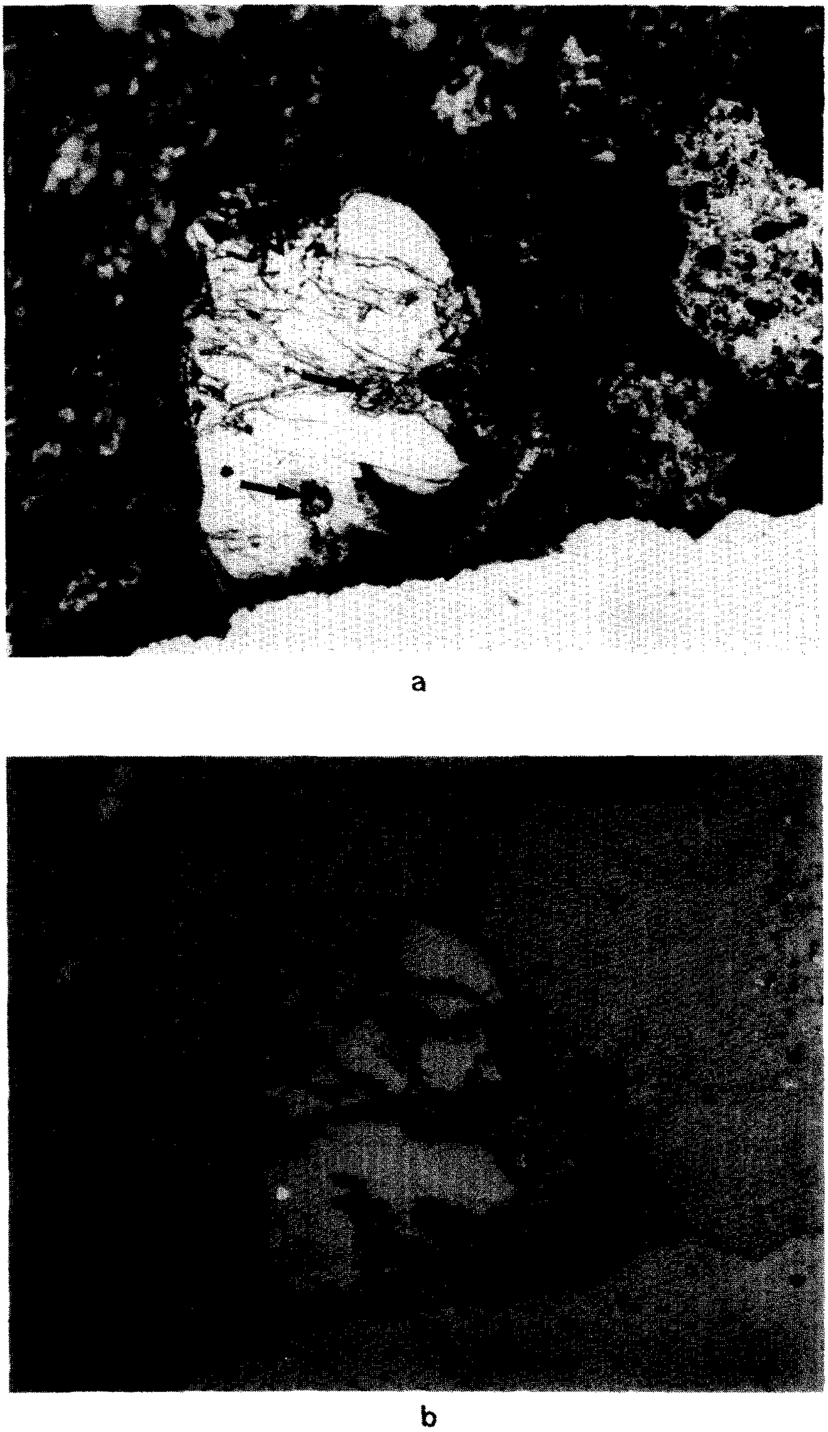


Fig. 15. (a) Olivine grain in Nogoya matrix, which is partially replaced on margins and in fractures by phyllosilicates and iron oxides (large filled arrow) and calcite (small open arrows). Small filled arrows point to iron oxide replacement of metal and sulfide. Plane light, field width = 0.45 mm. (b) Same as (a) in reflected light. Dashed line shows prealteration grain outline. Filled arrow shows direction of enhanced alteration-replacement along a system of subparallel fractures.

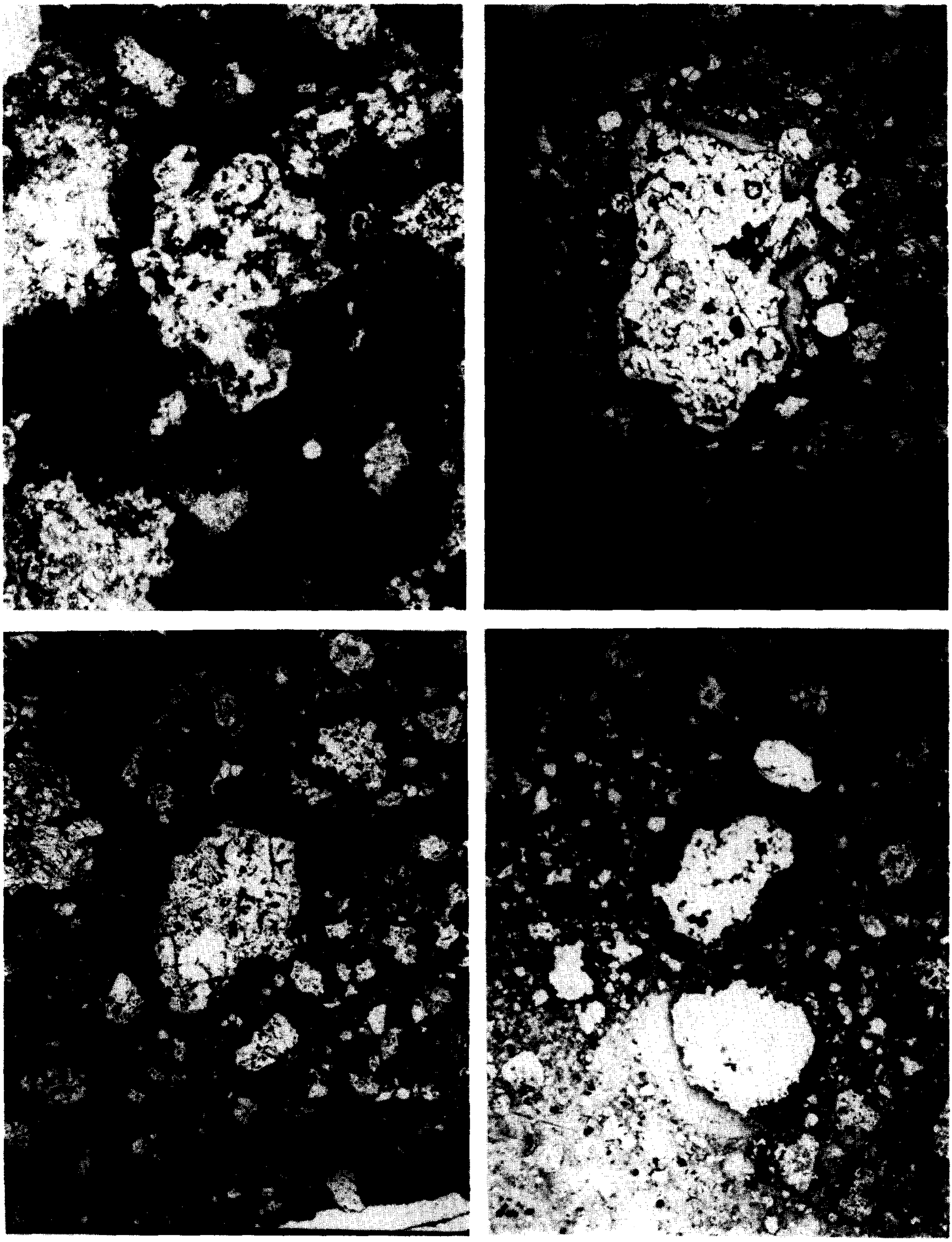


Fig. 16. Examples of C-rich haloes in CM and C3 meteorites. (a) Halo surrounding aggregate in Allende. (b) Altered halo surrounding aggregate in Vigarano. (c) Altered halo surrounding aggregate in Murchison. (d) Altered haloes surrounding aggregate in Murray.

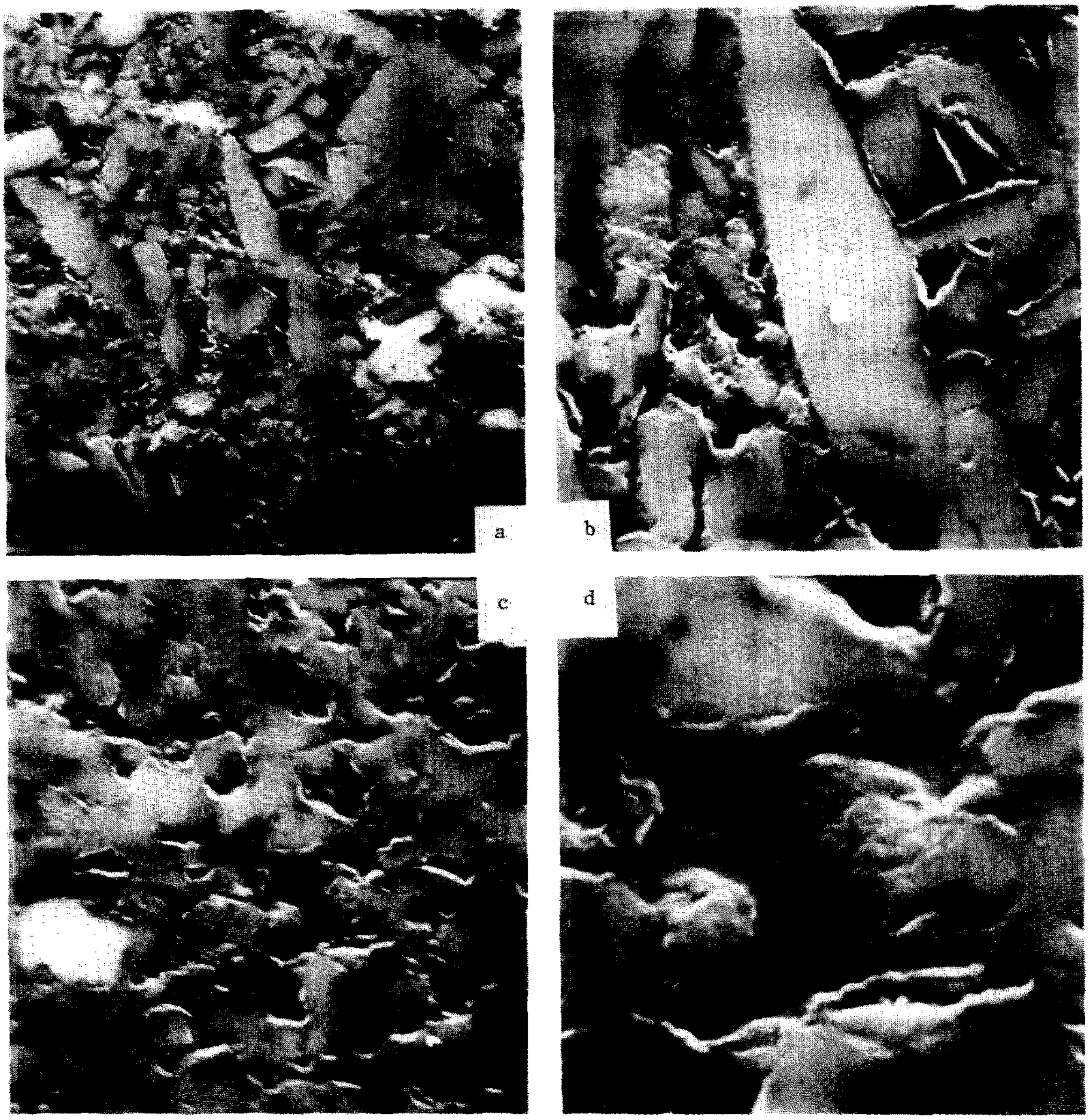


Fig. 18. Carbon and olivine relationships in Allende haloes and matrix. (a) SEM photograph of Allende matrix (polished thin section)—elongated grains are olivine, bright grains are sulfides (field width = 12 μ). (b) Enlarged area in (a) showing long olivine grain and carbonaceous coating of grains (field width = 3 μ). (c) SEM photograph of interlocking olivine grains in Allende halo (field width = 12 μ). (d) Enlarged central portion of (c) showing carbonaceous micromounds on olivine surfaces (field width = 3 μ).

et al. (1973) in Murchison. Examples of haloes in CM meteorites are shown in Fig. 16 with examples of haloes in two C3(V) meteorites, Vigarano and Allende. Haloes in CM meteorites are commonly less than 1/3 of the aggregate thickness and completely surround the aggregate. The haloes consist of phyllosilicates, PCP, carbonaceous matter and fine-grained sulfides with rare mineral fragments no larger than 5 μ. In standard thin sections, haloes appear dark, due primarily to large amounts of opaque phases. In ultrathin sections the opacity is diminished and fine-grained textures can be observed. Elongate grains in haloes are commonly oriented parallel or tangentially to the aggregate surface. Microprobe analyses of haloes indicate enrichment of carbon relative to the bulk meteorite (Fig. 17). Carbon shows a weak positive correlation with S and no correlation with Fe. In addition, C is concentrated at the aggregate halo interface and diminishes toward the meteorite matrix (Fig. 17).

Haloes are less abundant in C3 meteorites, although there is a correlation of haloed aggregates with bulk C content. Less than 15% of the olivine aggregates in Allende (total rock C = 0.3%, CHANG *et al.*, 1978) have haloes (8 thin sections), whereas about 40% have haloes (2 thin sections) in Vigarano (C = 1.15%, GIBSON *et al.*, 1971); all remnant aggregates in Nogoya (C = 5%, Table 4) have haloes. Moreover, haloes in Allende are irregular in shape and thickness and may not completely surround the aggregate. On the other hand, haloes in Allende are the most C-enriched, ranging 4–8 times the bulk C abundance (Fig. 17). The C3(O) chondrite Isna has no haloes and contains 0.19% C (METHOT *et al.*, 1975). Vigarano haloes are similar in bulk phase content to those in CM meteorites as phyllosilicates are common in Vigarano (Table 5). SEM observations and EDA and microprobe analyses show that Allende haloes consist of an interlocking network of mostly small (1–3 μ) olivine (Fo_{48-52}) with minor amounts of platelet metal and sulfides and high void space (Fig. 18). We observed a micromound texture on olivine surfaces in both haloes and matrix, in agreement with earlier findings of GREEN *et al.* (1971) and BAUMAN *et al.* (1973); the latter group interpreted the texture as vapor-deposited carbon. In contrast, matrix

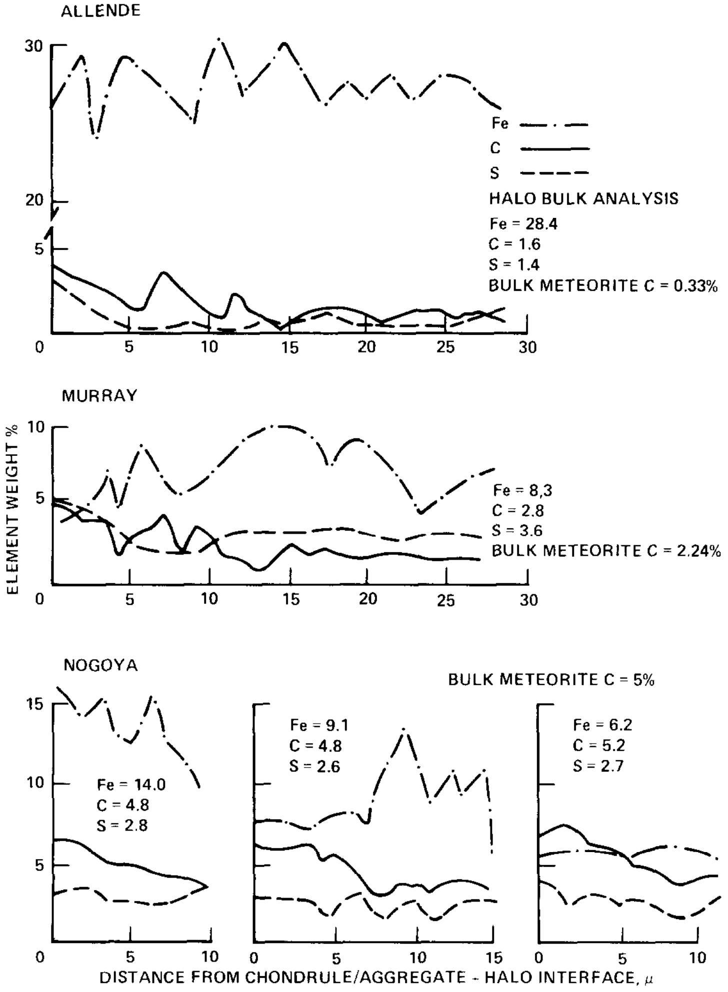


Fig. 17. Microprobe step scans for Fe, C, and S in Allende, Murray and Nogoya haloes showing enrichment of C near halo-chondrite or aggregate interface.

Table 5. Electron microprobe analyses of phyllosilicate and haloes in Vigarano

	Phyllosilicate	Haloes
SiO_2	30.9	28.8 - 41.9
Al_2O_3	5.0	2.1 - 4.0
TiO_2	0.08	0.11 - 0.16
Cr_2O_3	0.55	0.3 - 0.4
"FeO"	28.9	23.1 - 45.6
MgO	26.7	11.7 - 22.6
CaO	0.67	0.13 - 3.7
Na_2O	0.98	0.06 - 0.20
K_2O	0.64	0.04 - 0.18
P_2O_5	0.09	0.17 - 0.78
S	0.32	1.10 - 3.75
NiO	2.33	0.98 - 3.32
C*	0.80	0.8 - 2.1
H_2O†	10.00	
Sum	97.96	

* Carbon not included in totals.
† H_2O assumed to be 10%.

olivine (Fo_{48-52}) is larger (5 μ), has poorly developed micromounds, lower void space and is mixed with other phases (pyroxenes and high-temperature phases). Carbon appears to be associated entirely with olivine grains (CHANG *et al.*, 1978). Halo and matrix olivine contain low amounts of P, Al and S (Table 6). These elements and C do not coordinate well within the olivine structure and their presence may be attributed to substances adsorbed on olivine surfaces either during or after grain growth or both. No mineral inclusions or negative crystals were observed with the SEM to a maximum size of 500 Å. Haloes could represent the initial growth period of low-temperature condensates in contact with earlier formed high-temperature phases.

DISCUSSION

CM matrices: condensation or alteration origin?

The CM meteorite matrices contain phyllosilicates and other secondary minerals in a contextured manner that clearly indicate low-temperature alteration of precursive material on or near the surface region of a parent body. This conclusion is consistent with those of DUFRESNE and ANDERS (1962), NAGY *et al.* (1963), BOSTRÖM and FREDRIKSSON (1966), KERRIDGE (1977) and RICHARDSON (1978) for Orgueil matrix but in marked contrast with those of MCSWEEN and RICHARDSON (1977) and ARRHENIUS and ALFVEN (1971) who suggested a condensation origin from, respectively, the solar nebula and a low density partially excited gas. ARRHENIUS and ALFVEN (1971) based their suggestion on a conclusion drawn by JEFFREY and ANDERS (1970) that phyllosilicates in Orgueil were host phases for noble gases. FRICK and MONIOT (1976, 1977) and SRINAVASAN *et al.* (1977), however, have demonstrated that an acid-insoluble carbonaceous residue contains the main noble gas carrier(s) in Orgueil and CM meteorites, rather than phyllosilicate. MCSWEEN and RICHARDSON (1977), on the basis of matrix bulk analyses, suggested that compositional variations among CM meteorites resulted from mixing of two condensation components, phyllosilicates and an amorphous Fe–Ni–S–O phase (PCP), which is inconsistent with our observations.

To conclude that phyllosilicates formed as alteration products in a parent body, two major considerations must be met: (1) it must be demonstrated convincingly that phyllosilicates were indeed alteration derived and (2) pertinent observations must clearly show that alteration occurred within host rock and not prior to parent body accretion. To satisfy the first condition we cite from our petrographic observations, the presence of assemblages typical of terrestrial hydrous alteration, mainly combinations of serpentines, magnetite, calcite, hydrated iron oxides, awaruite and pentlandite. Textural relationships of these assemblages are highly suggestive of secondary alteration processes. Nogoya and Cold Bokkeveld contain a very high percentage (>95% for Nogoya) of alteration minerals, with remaining olivine and pyroxenes showing evidence of partial alteration in place. The most striking evidence for aqueous (liquid and/or gaseous) alteration is the presence of pseudomorphed chondrules, aggregates and clasts replaced by calcite, phyllosilicates and iron oxides (Figs 10–12). Colloform textures (Figs 3 and 14) are known to form only through aqueous activity. The redistribution of iron is

Table 6. Electron microprobe analyses of matrix and halo olivine in Allende

	Halo olivine (14)*		Matrix olivine (10)*	
SiO_2	34.5	(34 - 35)	35.6	(34.1 - 35.3)
Al_2O_3	0.67	(0.5 - 0.75)	0.38	(0.25 - 0.45)
Cr_2O_3	0.28	(0.2 - 0.34)	0.13	(0.05 - 0.18)
FeO	40.6	(39.8 - 41)	39.4	(39.4 - 40.2)
MgO	23.1	(22.7 - 23.5)	22.8	(21.6 - 23)
CaO	0.45	(0.4 - 0.52)	0.5	(0.02 - 0.09)
NiO	0.48	(0.4 - 0.56)	0.37	(0.2 - 0.44)
P	0.18	(0.11 - 0.25)	0.06	(0.02 - 0.1)
S	0.64	(0.51 - 0.7)	0.06	(0.03 - 0.13)
Sum	100.90		98.85	

* Number of grains analyzed.

highly suggestive of aqueous alteration. During alteration, iron may enter serpentine minerals and brucite or form secondary opaque minerals, magnetite, awaruite, PCP and pentlandite (MOODY, 1976), all of which are present in Nogoya and, with the exception of brucite, in all other CM meteorites that were studied. Formation of magnetite and pentlandite in CI meteorites (KERRIDGE *et al.*, 1979) and in CM meteorites (KERRIDGE and BUNCH, 1979) probably occurred through aqueous alteration processes. MCKEE and MOORE (1979) reported important observations from their high-resolution, SEM study of Murray and Nogoya phyllosilicates that give additional support to the alteration theory: (1) the range of laboratory-separated particle morphologies and structures are very similar to those of serpentines hydrothermally synthesized from olivine; (2) cylindrical axis thinness in some particles is similar to terrestrial serpentine minerals formed from pyroxenes. In addition, they found that some of the particles are probably admixtures of serpentine–talc–brucite, a situation that would unlikely arise through primary nebular or vapor condensation.

Our own SEM and petrographic observations, electron diffraction and SEM analyses indicate the probable presence of brucite in conjunction with magnetite in an altered aggregate in Nogoya (Fig. 14). This assemblage, in addition to being supportive of an alteration scenario, is quite useful in estimating f_{O_2} and temperatures of formation. MOODY (1976) reported from experimental work that brucite probably forms prior to magnetite in an alteration sequence. Magnetite and brucite [$Mg(OH)_2$] form at the expense of Fe-rich brucite on increasing temperature (not exceeding 375°C), total pressure of 0.3–2.0 kbar, and low f_{O_2}. At f_{O_2} higher than Fe–Fe_3O_4 or Fe_3O_4–Ni_3Fe field stabilities, iron either forms magnetite and bypasses brucite formation or forms Fe-rich brucite, which breaks down on decreasing temperature to form magnetite. The probable presence of Fe-rich brucite with magnetite in Nogoya established environmental limits of temperature, total pressure and f_{O_2}, which are within the environmental range that permit organic synthesis and preservation of the observed organic compounds in Nogoya (see below).

The main question to be answered is not whether alteration occurred, but where it took place—in the solar nebula or in a parent body. We conclude from the available data that all forms of alteration could have occurred within a parent body. We do not preclude some preaccretion alteration; the evidence seems to justify near-surface alteration of regolith-like material by gaseous or aqueous processes or by both. Pseudomorphic replacement, cross-cutting veins, colloform textures, serpentinite-like mineralogy and matrices composed of interlocking growths (sometimes spherulitic, e.g. Fig. 9) of alteration minerals and mesh textures argue for solid rock (or poorly consolidated regolith) alterations as opposed to nebular gas-grain interaction. Petrofabric analyses of CM meteorites show weak to pronounced lineation of matrix components and inclusions similar to the fabric of Allende. KING and KING (1978) attributed the lineation in Allende to overburden compaction or shock lithification in a regolith. There does not seem to be conclusive evidence in CM meteorites to indicate whether the observed fabrics are the result of simple compaction, flow orientation, or some other process. The important consideration here is that alteration occurred during or after lineation based on the textural continuity of phyllosilicates and other alteration minerals within clasts, through the clast-host boundaries, and into the host matrix.

The presence of from < 150 to 2000 ppm of chlorine in CM phyllosilicates and even larger amounts in amorphous colloidal material (up to 3200 ppm) of Nogoya (Fig. 14) indicates a possible redistribution of chlorine by a medium during alteration. SEM–EDA and electron microprobe analyses of unaltered olivine in CM and C3 meteorites show chlorine to be from below detection limits (< 150 ppm) to 300 ppm. Interstitial glass or poorly crystallized material in chondrules and aggregates in CM chondrites contains chlorine in amounts up to 2.1 wt%, the average being 0.4 wt%. Apparently, chlorine was redistributed during alteration into secondary phases and did not form separate chloride salts, as only poor correlations were found between Cl and Na or K. Beam scanning (SEM–EDA) of cross-cutting veins and pseudomorphs show higher amounts of chlorine on the average than the matrices, which are higher in chlorine than primary silicates. Redistribution of chlorine, especially in veins, pseudomorphic structures and colloidal materials suggest fluid or vapor transport, or both. From premliminary microprobe bulk analyses of CM matrices, there appears to be an inverse relationship between the chlorine content and degree of alteration. Nogoya, the most altered CM, contains only 400 ± 50 ppm (average) compared with 700 ± 100 ppm for the next most altered CM, Cold Bokkeveld, 1000 ± 150 ppm for Murray, and 1400 ± 200 ppm for Murchison, the least altered CM in our study. We find a similar trend among C3 meteorites with unaltered Allende matrix containing 1200 ± 175 ppm, Mokoia 1100 ± 150 ppm; the most altered C3 meteorite, Vigarano, contains 800 ± 100 ppm chlorine. Although the data are few, it seems reasonable to suggest that if Cl contents of these meterorites are not inherent to initial bulk compositions, the chlorine may have been partially leached out during aqueous alteration. Similarly, the rather high incidence of calcite replacement in pseudomorphic structures and in limited vein occurrences in Nogoya also suggests that CO_2 or H_2CO_3 was available in a fluid or gas phase. The amount of carbonate alteration is low compared with terrestrial serpentinites. The presence of large amounts of carbonates in altered terrestrial rocks is commonly the result of an open system in which joints and fractures serve as conduits for CO_2-bearing fluids. CM meteorites are nearly devoid of open channel ways for migrating fluids, although Orgueil is obviously veined. There is little evidence to support an extensive open system exchange in CM meteorites during alteration; the system may have been open with respect to species like H_2O and CO_2 which can remain in the gaseous state. The volume of fluids and gases and rates of exchange between them and solids are important with respect to metasomatic effects, which are minor or nonexistent if a fixed volume of fluid or gas remains in a closed system. The probable high porosity of prealteration material (regolith) greatly aided the alteration processes in CM meteorites, although the lack of fractures restricted or precluded significant metasomatic effects. Veining (fracture filling) shows a positive correlation with the degree of alteration from no visible veining in Murchison and Murray, little veining in Cold Bokkeveld and Nogoya to significant veining and more complete alteration in Orgueil.

KERRIDGE (1977) concluded that sympathetic variations of Ni and S in Orgueil (CI) phyllosilicates was probably the result of aqueous alteration of a Ni-bearing sulfide. We concur that most of the Ni and S in CM (and probably C3) phyllosilicates resulted from decomposition of a Ni-bearing sulfide, although the sympathetic correlation is not as clear in CM phyllosilicates, because of the intimate association of phyllosilicates with PCP. Poor single-phase grain resolution during microprobe analyses does not allow for good statistical confirmation.

Because of the excellent ion-exchange properties of clays, KERRIDGE (1977) reasoned that Na in phyllosilicates should be more depleted than K relative to bulk meteorite compositions. From our analyses of four different phyllosilicates in Murray (53 individual microprobe analyses), Na_2O/K_2O ratios show a narrow range between 0.77 and 0.91 whereas the bulk meteorite ratio is 5.3 (WIIK, 1956). For Nogoya phyllosilicates (two types; 36 analyses) a range of 0.83 to 0.90 is indicated with a bulk ratio of 2.6 (FRIEDHEIM, 1888; AHRENS *et al.*, 1969). Analyses of Na and K in Murchison and Jodzie phyllosilicates concur with this trend and we conclude that the enhancement of K in phyllosilicates is the result of aqueous ion exchange.

Probably the main objection to *in situ* alteration is the occurrence of altered and unaltered materials in proximity to each other. Actually, nearly all components in CM matrices, clasts, chondrules and aggregates, show various degrees of alteration. If the viewpoint is taken that CM meteorites are regolith breccias that have formed in a manner analogous to lunar surface breccias but to a lesser degree of maturation, then this objection loses its plausibility. The constant flux of new material, together with impact excavation and mixing of older deposits could leave a heterogeneous mixture of altered and unaltered material if the time period of hydrothermal activity was short-lived.

C3 phyllosilicates

Phyllosilicates in CI and CM meteorites are well known, but what do we know about C3 phyllosilicates? VAN SCHMUS and HAYES (1974) summarized chemical and petrographic correlations among carbonaceous chondrites. They note that two C3(V) meteorites are known to contain phyllosilicates: Al Rais and Renazzo with H/100 Si (total rock) ratios of 186 and 111, respectively. Other known C3(V) meteorites have olivine matrices with H/100 Si ratios ranging from 84 for Grosnaja to 0.01 for Allende. To date, we have been able to study three C3(V) meteorites, Vigarano, Mokoia and Isna, in addition to Allende. Vigarano is described as having an olivine matrix and a H/100 Si ratio of 58 (VAN SCHMUS and HAYES, 1974). Although olivine is the dominate matrix phase, our ultrathin section observations confirm the presence of a large amount of phyllosilicates and amorphous to poorly crystalline material resulting most probably from alteration of precursive olivine, metal and sulfides. This is also true for Mokoia which contains less phyllosilicate in comparison with Vigarano. Isna does not appear to contain any phyllosilicate.

Phyllosilicates in Vigarano are distinctively different from those in CI and CM in terms of petrographic characteristics and composition. Vigarano phyllosilicates contain greater amounts of alkalis, Cr, Ca and Al (Table 5) compared with CI and CM varieties (Table 1). The 'FeO'/MgO ratios are similar, although total 'FeO' and MgO contents are greater. We find that there are definite correlations between phyllosilicate compositions and host bulk compositions or precursive phases, for example, Vigarano bulk composition is enhanced in Ca, Al, Cr and alkalis compared with CM meteorites (MASON, 1963).

Vigarano (C3V) phyllosilicates are also Na depleted but to a lesser degree. Electron microprobe analyses of the dominate phyllosilicate (Table 5) show a Na_2O/K_2O ratio of 1.54 compared with a bulk ratio of 7.2 (MASON, 1963). The smaller depletion of Na is probably the result of poor ion-exchange efficiency stemming from limited ion transport (low water content).

Because the H/100 Si ratio of Vigarano is typical of the 10 C3(V) meteorites listed by VAN SCHMUS and HAYES (1974), we assume that they must also contain phyllosilicates and that they were subjected to alteration during the residence time in the parent body.

The only sources of H in these meteorites are in indigenous organic matter, hydrated salts (e.g. epsomite) and adsorbed terrestrial water. We note that the H/C ratios calculated from VAN SCHMUS and HAYES (1974) for these seven C3(V) meteorites range from 3.4 (Vigarano) to 13.4 (Coolidge). If we adopt the conservative view that the maximum H/C ratio of meteoritic organic matter is 3 (KAPLAN, 1971) then substantial amounts of H remain to be taken into account. Among the C3(V) meteorites, Mokoia ($H/C = 3.8$) is the only one for which an estimate has been made of the contribution of terrestrial water (released by heating in vacuum to 180°C) to the total water content (released from 180 to 800°C). BOATO (1954) concluded that 38% of the water released from Mokoia could have been terrestrial contamination. Although there are insufficient data to quantitatively assess the relative contributions of terrestrial and meteoritic H in the C3(V) meteorites, the clear presence of phyllosilicates in Vigarano, Mokoia, Al Rais and Renazzo coupled with respective H/C ratios of 3.4, 3.8, 4.5 and 5.2, lend strength to our contention that other C3(V) meteorites with comparable or higher H/C ratios also contain phyllosilicates. We suggest that most carbonaceous chondrites underwent some alteration on their parent body. Moreover, because it is unlikely that all carbonaceous chondrites originated from the same parent body, it appears reasonable to assume that low-temperature hydrothermal alteration was common among many parent bodies. In this context we point out that hydrothermal alternation is not limited to carbonaceous chondrites. BUNCH and REID (1975) and ASHWORTH and HUTCHISON (1975) interpreted aqueous alteration in the nahklite meteorites to be preterrestrial in origin; and the latter authors also reported evidence of similar alteration in the Weston chondrite.

Stable isotope compositions

The oxygen isotopic compositions of carbonate in CI and phyllosilicates in CI and CM matrices have been interpreted in terms of narrow ranges of formation temperatures for these phases (ONUNA *et al.*, 1972): 360 ± 15 K for Orgueil (CI), 380 ± 15 K for Murray (CM) and Cold Bokkeveld (CM). ONUNA *et al.* (1974) pointed out, however, that the discovery of nonchemical isotope effects in carbonaceous meteorites signified large uncertainties in their previous estimate of the oxygen isotopic composition of nebula gas on which formation temperatures of putative nebula-derived products were based. Despite the appropriateness of a low-temperature origin for carbonates and phyllosilicates in hydrous matrices, ignorance of isotopic composition(s) of the gaseous oxygen resevoir(s) from which these minerals were derived or with which they may have exchanged, either in the solar nebula or on parent bodies, precludes assignment of specific formation temperatures on the basis of available isotopic data.

On a three-isotope plot, oxygen compositions for CI and CM hydrous matrices lie above and below the terrestrial fractionation line, respectively, and each appears to define lines with slope ~0.5, suggestive of mass fractionation trends (CLAYTON and MAYEDA, 1977; CLAYTON, 1978). It has been pointed out (CLAYTON, 1978) that the oxygen in CI matrices could have been derived from the same reservoir as the oxygen in H-chondrites, as these two types of meteorites can be related by a common fractionation line. Except for the achondritic portions of the brecciated meteorites, Bencubbin and Weatherford, and possibly material from the unusual chondrite Kakangari, noncarbonaceous meteorites cannot be related to CM matrices by mass fractionation (CLAYTON *et al.*, 1976; CLAYTON and MAYEDA, 1978; CLAYTON, 1978). With the possible exception of the most iron-rich olivines in C3 meteorites, which lie with CM matrices on a common fractionation line (see the floor discussion that follows CLAYTON, 1978), the bulk oxygen reservoirs measured in CI and CM hydrous matrices are isotopically distinct and cannot be related to each other or to anhydrous phases in CM and other carbonaceous meteorites by fractionation processes alone. A referee has suggested that all CM parental materials were *initially* endowed with the same proportions of anomalous ^{16}O-rich oxygen (from percursive anhydrous phases) and normal oxygen (from water), the various compositions corresponding to different temperatures for formation. Whether it is reasonable that the same proportions of these isotopic reservoirs could have occurred among different CM parent bodies or nebula source regions is unclear. If all CM meteorites originated from the same parent body or nebula source region, then local variations in temperature and chemistry would be required.

In the discussion following the CLAYTON (1978) paper, Clayton acknowledged the possibility that low-temperature oxygen exchange between an ^{16}O-depleted gas and chondritic material could account for the heavy isotope enrichment of CI and CM hydrous matrices, provided that the exchange was sufficiently extensive and involved a sufficiently large reservoir as to obliterate the original composition. These requirements are weakened, however, if highly ^{16}O-depleted isotopic compositions are allowed for the low-temperature oxygen reservoir(s). Thus, oxygen isotope data do not preclude the combining of isotopically distinct precursive anhydrous phases from one (or more) high-temperature source regions with isotopically distinct water from one (or more) low-temperature source regions to form hydrated reaction products. Formation of hydrous matrices could have occurred either in the low-temperature regions of the solar nebula or on a partially formed accreting parent body. Oxygen isotope data alone cannot yet distinguish between the two possibilities. Our mineralogical and petrological observations, however, argue strongly in favor of parent body alteration environments.

Both CI and CM hydrous matrices are depleted in ^{16}O relative to anhydrous phases of carbonaceous meteorites (CLAYTON and MAYEDA, 1977; CLAYTON *et al.*, 1977). Moreover CI matrices, whose mineralogy and petrology exhibit most prominently the effects of aqueous hydrothermal alteration, are more strongly depleted in ^{16}O than CM matrices. The difference in depletion may reflect isotopically different sources of water, or different amounts of isotopically similar water in the matrices, or isotopically different precursive anhydrous phases, or combinations of these factors. McSween and Richardson (1977) have concluded, however, that the hydrous matrices in carbonaceous meteorites could not have been produced by hydration of C3 olivine because oxygen isotopic differences between CI, CM and C3 matrices argue against a common progenitor. Since the isotopic composition(s) of water in the solar nebula or on parent bodies are unknown and since precursive anhydrous phases could have undergone extensive low-temperature alteration and oxygen exchange with water reservoirs, there are few constraints that can be rigorously imposed on the isotopic compositions of precursive phases. For these reasons, McSween and Richardson's (1977) conclusion appears to be premature.

Large variations in the isotopic composition of nitrogen also exist between classes of meteorites and within individual meteorites (Injerd and Kaplan, 1974; Kung and Clayton, 1978). The nitrogen in CI and CM meteorites is strongly enriched in ^{15}N by factors of 20–110‰ relative to that in C3 and enstatite chondrites, but only weakly enriched relative to ordinary chondrites. These variations may be interpreted as manifestations of isotopic inhomogeneity in the source regions, although 'specially contrived fractionation processes cannot be ruled out' (Kung and Clayton, 1978). Interestingly, kinetic isotope effects associated with a Fischer–Tropsch type (FTT) and Miller–Urey syntheses of nitrogenous organic matter cannot account for the N-isotopic variations (Kung and Clayton, 1978).

In chondritic meteorites where the mineral assemblages show no exposure to a hydrous environment, carbon occurs predominantly in a complex, macromolecular, reduced form, most of which is insoluble in acids (Hayes, 1967; Belsky and Kaplan, 1970; Smith and Kaplan, 1970; Chang *et al.*, 1978). In CI and CM meteorites, which contain both anhydrous phases and minerals that reflect exposure to or synthesis in an aqueous environment, carbon, though appearing largely in macromolecular assemblages, also takes the form of carbonate and organic compounds extractable by water and solvents. Available measurements on chondritic meteorites indicate that the carbon isotopic composition of acid-insoluble carbonaceous matter is relatively enriched in ^{12}C, with $\delta^{13}C$ values (relative to Pee Dee belemnite throughout this paper) ranging from −11 to −30‰ (Hayes, 1967; Belsky and Kaplan, 1970; Krouse and Modzeleski, 1970; Kvenvolden *et al.*, 1970; Smith and Kaplan, 1970; Chang *et al.*, 1978; this work, Table 4). In contrast, carbonate carbon is relatively enriched in ^{13}C with $\delta^{13}C$ values ranging from +25 to +70‰ (Clayton, 1963; Krouse and Modzeleski, 1970; Kvenvolden *et al.*, 1970; Smith and Kaplan, 1970; Chang *et al.*, 1978; this work, Table 4) [except for a single value of +4‰ reported by Krouse and Modzeleski (1970) for the CI Ivuna]. Thus, large differences, of the order of 40 to 100‰, exist between the isotopic compositions of macromolecular acid-insoluble carbonaceous matter and carbonate.

The large isotopic differences previously had been interpreted as the result of kinetic isotope effects associated with a nebular FTT synthesis of CO_2 and organic matter from CO and H_2 (Lancet and Anders, 1970; Anders *et al.*, 1973). The carbon isotopic composition of compounds extractable with organic solvents from CI and CM meteorites ranges in $\delta^{13}C$ values from −28 to +6‰ (Hayes, 1967; Krouse and Modzeleski, 1970; Kvenvolden *et al.*, 1970; Smith and Kaplan, 1970). These measurements may have been influenced to various degrees by terrestrial contaminants (Smith and Kaplan, 1970). But if the range of isotopic compositions is real, then the data are consistent with a FTT synthesis for these compounds. Recent analyses by Chang *et al.* (1978), however, revealed that water-soluble organic matter in Murchison (CM) was strongly enriched in ^{13}C, with $\delta^{13}C$ values ranging from +13 to +44‰, similar to that of coexisting carbonate (+44‰) but contrary to expected isotopic compositions based on the FTT model. Conceivably, these organic compounds were made from isotopically heavy CO_2 produced as a by-product of FTT reactions in the nebula (E. Anders, personal communication 1978). By analogy with the Fischer–Tropsch reaction (Anderson, 1961), however, *synthesis from CO_2 in the nebula could have proceeded only after all CO had been consumed* or converted to other species. The plausibility of such a scenario for nebular evolution and organic synthesis remains to be assessed, and the isotopic fractionation associated with FTT synthesis of organic matter from CO_2 awaits elucidation. Possibly, the polar, water-soluble, organic compounds were produced by processes distinct from those which produced the rest of the organic matter. Apparently, however, recent data indicate a need to re-examine the FTT synthesis model and its implications for the chemistry and isotopic composition of C and N in carbonaceous meteorites.

An alternative explanation for the large isotopic fractionations was offered by Clayton (1963) who suggested that the carbon isotopic heterogeneity among carbonaceous components may have resulted from their origins from sources with different nucleosynthetic histories. This interpretation becomes more plausible now that explanation of oxygen and nitrogen isotopic patterns in meteorites appears to require nebular isotopic heterogeneity and nuclear effects in addition to mass-dependent fractionation. In this context we point out that the strongest reported enrichments of the heavy isotopes of O, N and C occur in just those meteorites that show ample evidence of having been exposed to hydrous, low-temperature environments. We suggest that associated with these environments were reservoirs of O, N and C that were isotopically distinct from the reservoirs that contributed these elements to anhydrous phases. In terms of the two component model for carbonaceous meteorites (Wood, 1963; Anders, 1964; Anders *et al.*, 1976) this view implies a sub-division of the low-temperature component into two fractions, one characterized by anhydrous origins and one derived from hydrous origins, the latter providing the water for hydrothermal alteration of anhydrous phases, CO_2 for carbonate formation, water-soluble organic compounds, and possibly other volatile species.

Sources of water for hydrothermal alteration

Presumably, the water in the hydrous matrices of CI and CM meteorites had a primary origin in the gaseous solar nebula. Whether the effects of the water on meteorite components were achieved in the nebula or on a parent body depends on the interpretation of mineralogic, petrologic, chemical and isotopic observations. KERRIDGE and BUNCH (1979) summarized the evidence indicating that CI and CM meteorites were formed in the regoliths of their parent bodies and that their hydrous mineralization marked one of the earliest dated events in the solar system. Although these authors judged asteroids to be the probable parent bodies for CI and CM meteorites, they also considered protocometary bodies and extinct (inactive) cometary cores. They also attributed the water and other volatiles to local sources within the parent bodies. Thus, observations pointing to *in situ* hydrothermal alteration on parent bodies imply that the water was acquired from the nebula initially in condensed or chemically bound forms during accretion of the parent bodies. The internal source concept was derived from the earlier suggestion of DuFRESNE and ANDERS (1962) that hydrothermal alterations of anhydrous silicates in near-surface regions of a parent body could have been carried out by water and other volatiles that were transported from depth where they were released from pre-existing phases by internal heating, presumably as a result of radionuclide decay (possibly of the short-lived radionuclide ^{26}Al [LEE *et al.*, 1977]).

In the absence of data requiring a nebula origin for phyllosilicates, however, it is worthwhile to consider ice rather than an earlier generation of phyllosilicates (as suggested by DuFRESNE and ANDERS, 1962 and LARIMER and ANDERS, 1967), as an internal source of water. In this context we regard the parent bodies as heterogeneous agglomerates that accreted planetesimals containing different amounts of both anhydrous and hydrous—but phyllosilicate-free—volatile-rich material. Although the presence or absence of phyllosilicates in comets remains to be established, current conceptions of comets and, presumably, of protocometary bodies with low dust-to-gas ratios (0.5–2; DELSEMME, 1977) do correspond to what we envision to have been planetesimals dominated by hydrous volatile-rich components. Thus, proto-asteroidal and proto-cometary bodies are viewed as components of a distribution of planetesimals that when formed at increasing heliocentric distances accreted increasing proportions of ice and other volatile-rich phases. Dynamical processes can be envisioned (see WEIDENSCHILLING, 1977, for example) that would foster accretion into parent bodies of anhydrous and hydrous components originally formed in widely separated source regions that could have differed widely in temperature, pressure and chemical and isotopic composition. The record of extensive mixing of meteorites of one (or more) type(s) within a meteorite of another type summarized by WILKENING (1977) is noteworthy in this context.

In addition to an internal source, water and other volatiles could have been supplied at or just below the surfaces of CI and CM parent bodies. Formation of transient atmospheres and release of volatiles in near-surface reservoirs were likely to have accompanied the accretionary growth of parent bodies as a result of low-velocity impacts heating both projectiles and targets. The longevity of an atmosphere formed during accretionary growth of a parent body has not been investigated. But it is reasonable to expect that it would permeate a regolith and persist as long as the production rate of atmospheric species exceeded the escape rate, and this may have been possible during a period of rapid accretionary growth as envisioned for the production of bodies up to 500 km in diameter from 1-km-sized planetesimals (HARTMANN, 1978; GREENBERG *et al.*, 1978). Clearly, trapping of volatiles within thick global ejecta blankets during regolith development on parent bodies (HOUSEN *et al.*, 1979) would have provided a reasonable mechanism for water retention and hydrothermal alteration. In addition it is possible that if internal heating was sufficiently intense to yield a differentiated parent body, volatiles would have been expelled to form a transient atmosphere. In this connection is is noteworthy that the 1018-km-diameter asteroid, Ceres, exhibits spectral reflectance properties which resemble those of the CM Murchison (MATSON *et al.*, 1978).

Implicit in the preceding discussion of ice as the internal water source is the requirement that the nebula temperature fell from about 400 K to below the freezing point of water without equilibration between dust and gas and consequent nebular alteration of anhydrous phases (KERRIDGE and BUNCH, 1979). In contrast, the equilibrium condensation model predicts appearance of magnetite at 405 K and phyllosilicates below 350 K as nebular alteration products (GROSSMAN and LARIMER, 1974). Furthermore, the internal water concept (regardless of whether the source consisted of ice or phyllosilicates, or both) places volatile-rich phases in the interiors of parent bodies as well as at the surfaces. This distribution also represents a departure from a simple heterogeneous accretion of the products from the equilibrium condensation model, which predicts the concentration of volatiles on the exterior and of anhydrous phases in the interiors of carbonaceous chondrite parent bodies. The latter distribution is based on the assumption that as the nebula temperature dropped, mineral condensates continued to take up more volatiles by equilibration with the nebula gas while concomitantly accreting onto parent bodies. The temperature at which equilibration ceased was taken to be the accretion temperature.

Arguments favoring continued nebula equilibration below 400 K are founded on interpretations of measurements derived from meteorite analyses and from model studies (see Table 6, GROSSMAN and LARIMER, 1974): (1) oxygen isotope ratios for CI and CM matrix components—taken to represent accretion temperatures under the assumptions that isotopic exchange occurred in the nebula and that the nebula oxygen isotopic composition could be adequately estimated (ONUMA *et al.*, 1972; 1974); (2) the difference in $^{13}C/^{12}C$ ratio between carbonate and organic C—attributed to kinetically controlled FTT synthesis in the nebula (LANCET and ANDERS, 1970); (3) the presence of magnetite and phyllosilicates—presumed to reflect formation due to equilibration of water in the nebula with suspended anhydrous mineral grains (LARIMER and ANDERS, 1967; GROSSMAN and LARIMER, 1974); (4) the abundance of Ar in magnetite synthesized under simulated nebula conditions—hypothesized to account for ^{36}Ar in meteoritic magnetite on the basis of equilibrium solubility (LANCET and ANDERS, 1973); (5) the Tl abundance in CI meteorites presumed to indicate its low-temperature equilibrium condensation from the nebula gas (LARIMER and ANDERS, 1967).

We have pointed out above that there are grounds for uncertainty about the bases on which the interpretations for items (1) and (2) were made. Item (3) can hardly be used as unambiguous support for nebula equilibration now that our own petrographic observations and those of KERRIDGE *et al.* (1979) point to a parent body origin for phyllosilicate and magnetite. Item (4) carries little weight insofar as magnetite can also be interpreted as a product of parent body processes. In addition, the requisite elemental fractionation of the heavy gases found in host phases of the planetary noble gas component in carbonaceous meteorites (see LEWIS *et al.*, 1975; REYNOLDS *et al.*, 1978; FRICK *et al.*, 1979) does not occur in the synthesis of magnetite (LANCET and ANDERS, 1973). Item (5) remains, but its applicability depends on the assumption that the pressure in the nebula where Tl condensed was less than 10^{-5} atm (LARIMER and ANDERS, 1967; GROSSMAN and LARIMER, 1974).

On the whole, the available evidence for dust-gas equilibration below 400 K in the nebula appears not to be compelling. Indeed, parent body origins are quite reasonable for some of the processes previously assumed to have occurred in the nebula. Thus, the possibility remains that in the nebular regions where components of carbonaceous meteorites were formed prior to their assembly into parent bodies, the transition from 400 K to below the freezing point of water occurred without hydrous alteration of anhydrous phases. Although knowledge of the composition of comets is limited, it does suggest that their chemistry is difficult to accommodate without imposing exotic constraints on the equilibrium condensation model (DELSEMME and RUD, 1977). If comets and CI, CM and some C3 parent bodies were members of a population, that acquired different initial contents of ice and volatiles by similar processes, then a common difficulty in formulating a nebula–equilibrium origin for their low-temperature hydrous phases and volatiles is expected. In any case, evidence for a nebula–equilibrium origin for the hydrous matrices of carbonaceous meteorites appears to be weaker than previously thought, and reexamination of these objects from the perspective of parent body alteration processes may yield fresh insights into their origin and evolution.

Organic chemistry

As a consequence of the internal heating model, organic synthesis could have occurred within the parent body as hot gases or hot solutions or both, containing the elements H, C, N, O (and S) in reactive forms were transported from the interior to near-surface regions, provided that the thermally labile products could be deposited and stored at relatively cool (<350 K) sites. Synthesis could also have taken place in ejecta blankets as they cooled and in transient atmospheres. Syntheses in the interior and in ejecta blankets might have occurred with grain surfaces acting as catalysts, analogous to the FTT reactions (which involve CO, H_2 and NH_3) advocated by ANDERS *et al.* (1973) for organic synthesis in the nebula, but with H_2O as the dominant constituent rather than H_2. Relevant to this context are reports that organic compounds are synthesized when hot gas mixtures of H_2O, CO and NH_3 (PONCELET *et al.*, 1975) and H_2O, CO_2, NH_3 (HARVEY *et al.*, 1971) were passed over mineral inorganic substrates. Syntheses in a transient atmosphere could have proceeded by Miller–Urey reactions and by shock mechanisms associated with infalling material (BAR-NUN *et al.*, 1970).

If the planetesimals that made up CM and CI parent bodies contained material similar in chemical composition to models of cometary and protocometary nuclei (abundant volatiles and organic matter, low gas-to-dust ratio) then organic matter may have been present *prior* to the onset of internal heating and hydrothermal alteration processes. In the presence of water, hydrolytic reactions of pre-existing organic compounds may have been important (for instance the hydrolysis of nitriles, amides, or esters to carboxylic acids). Although recently DELSEMME and RUD (1977) contrived a quenched-equilibrium model to explain their presence in cometary (and presumably protocometary) bodies, the nature and origin of cometary organic compounds remains poorly understood.

Contributions from some or all of the sources mentioned above, plus others not readily apparent, may be represented in the organic matter that occurs in hydrous carbonaceous meteorites but is absent anhydrous meteorites. Further detailed consideration of the origin(s) of the organic matter lies outside the scope of this paper (see ANDERS *et al.*, 1973, and MILLER *et al.*, 1976, however, for earlier discussions).

The amount and molecular composition of abiotically synthesized organic matter and the degree of hydrothermal alteration may be related and may provide insight into the temperature, timing and duration of the two processes. Amino acids are known to be thermally labile in meteorite matrices; heating of Murchison under vacuum for 1 week at 458 K reduced the abundances of individual amino acids by factors of 2 to 17 (CRONIN and MOORE, 1974). If hydrothermal synthesis of phyllosilicates occurred at temperatures lower than 350 K over periods of months or years, then heat-sensitive organic compounds could have been formed prior to or concurrently with the alteration process and some of them would have survived. In this case we might expect that the abundances of these compounds in meteorites would correlate inversely with the degree of alteration. In this context, the sixfold depletion of amino acids in Nogoya relative to their abundances in Murray and Murchison (CRONIN and MOORE, 1976) is consistent with the substantially higher degree of alteration in Nogoya. If phyllosilicate formation occurred at temperatures substantially higher than 350 K, then the presence of amino acids and other heat-sensitive compounds, if synthesized previously or concurrently, implies alteration over periods of weeks to months. Longer periods of alteration at elevated temperatures are not precluded if heat-sensitive compounds were made elsewhere and then transported to their final sites after the alteration environment cooled.

It is noteworthy that LAWLESS *et al.* (1974) identified malonic acid [$H_2C(CO_2H)_2$] in hot water extracts of Murchison. Since thermal decomposition of this dicarboxylic acid is expected to occur very rapidly on a geological time scale at 300 K (S. L. MILLER, private communication, 1979), its presence indicates an upper limit for the temperature of the parent body environment in which it was either synthesized or preserved. Quantitative measurements of the abundances of diagnostic heat-sensitive organic compounds in carbonaceous meteorites are scarce; additional data would be useful in establishing constraints on their low-temperature thermal histories.

Precursive material and alteration mechanics

Although the physical–chemical characteristics of the alteration environment remain to be clearly defined, it is instructive to consider a hypothetical alteration scenario and the experimental observations that bear on its credibility. For purposes of illustration we assume that unaltered material resembles anhydrous chondritic meteorites and use Allende as a model only to focus the discussion, rather than to assign it a specific precursive role.

Carbon in Allende appears to be associated predominantly with olivine grains and is concentrated on their surfaces in the form of carbonaceous micromounds. It is uncertain whether the C-rich micromounds were vapor-deposited directly on olivine surfaces while the grains were suspended in a gas phase (in the nebula or on a parent body, or both) or whether they were produced separately then mixed with olivine grains in a parent body regolith, or both. Acid insoluble carbonaceous matter has been implicated as the carrier phase of at least two noble gas components in Allende (LEWIS *et al.*, 1975; LEWIS *et al.*, 1977; REYNOLDS *et al.*, 1978; FRICK and CHANG, 1978; OTT *et al.*, 1979). No definitive evidence links the micromounds directly to the acid insoluble carbonaceous residues; however, the former comprise the only directly observed C-rich substances in Allende. Although the natural mechanisms of host phase synthesis and gas trapping are unknown, FRICK *et al.* (1979) and KOTHARI *et al.* (1979) have shown that laboratory syntheses of carbonaceous host materials result in high concentrations and strong elemental fractionation of the trapped noble gases characteristic of the planetary component in meteorites. Thus, the available data are compatible with the idea that noble gas incorporation accompanied formation of the carbonaceous micromounds.

After its accretion into the parent body regolith, alteration of precursive anhydrous material required the presence of at least H_2O and CO_2. Methane, CO, N_2 and other gases may also have been present. The CO_2 is required to supply the carbonate found in hydrous matrices. In the presence of H_2O and CO_2, olivines and other anhydrous silicates were altered to phyllosilicates and sulfides were converted to more oxidized forms of S (see below). The consequent disruption of olivine structure caused colloidal separation of the associated carbonaceous micromounds, which tended to aggregate and associate with alteration products derived from sulfide and metal to yield various forms of PCP. Thus, the original association of S and C with olivine is replaced by the association of PCP with phyllosilicates. (In this connection, it is noteworthy that MCKEE and MOORE (1979) and MACKINNON and BUSECK (1979) have observed ubiquitous coatings of amorphous material on phyllosilicate particles obtained from CM matrices and suggested that they might be carbonaceous.) According to this view, the noble-gas-rich acid insoluble carbonaceous residues from hydrous meteorites are derived predominantly from PCP and include micromound material. Although noble gases in large part must be retained in host phases during alteration, some gas loss cannot be excluded nor can incorporation of additional noble gases into phases *newly formed* during the process. In any case, evidence pointing to carbonaceous carriers for several trapped noble gas components in CM and CI meteorites (SRINIVASAN *et al.*, 1977; REYNOLDS *et al.*, 1978; EBERHARDT *et al.*, 1979; ALAERTS *et al.*, 1979) suggests that material in their parent bodies sampled a multiplicity of environments characterized by different noble gas compositions.

Abiotic organic synthesis in hydrous regions of the parent body (see above) could have supplied some of the organic compounds that occur in CM and CI meteorites but are absent in anhydrous meteorites. The presence of organic matter in association with precursive anhydrous phases or in the water reservoir (or both) may have facilitated the hydrothermal alteration process if the suggestion by SIFFERT (1979) that organic matter lowers the activation energy for clay synthesis in terrestrial systems proves correct and is applicable by analogy to the meteorite parent body milieux.

In nebula condensation models (GROSSMAN and LARIMER, 1974; LEWIS, 1972), troilite is the only sulfur species predicted to form at temperatures higher than the condensation point of NH_4SH (<250 K); its formation temperature range is 680–600 K. ANDERS *et al.* (1976) suggested the possible formation of sulfides of the trace elements Tl, Cd, Bi and Pb at lower temperatures (≤ 400 K), but their abundances are expected to be much lower than that of troilite; little direct evidence exists for their presence in meteorites. For the present discussion, we take the view that the chemical evolution of sulfur in carbonaceous meteorites involves essentially one reservoir of sulfur, that contained in unaltered (by H_2O) anhydrous chondritic material and represented by troilite. (We note, however, that in Allende the sulfides occur as pentlandite and troilite (CLARKE *et al.*, 1970).

The concept of a common source of S for meteorites was articulated by KAPLAN and HULSTON (1966) and was based on the uniformity of isotopic compositions for total S in 20 iron and stony meteorites: $\delta^{34}S_{CD}$ values encompassed a range of less than ±1‰. Because the bulk and weighted average isotopic composition of S in the meteorites they studied were essentially constant, despite variations among different S phases, these workers suggested that *in situ* fractionation by chemical processes occurred in a system that was closed to significant losses of component sulfur phases.

If all the sulfur in anhydrous precursive materials existed initially in the sulfide form, then progressive alteration oxidation reactions analogous to those first proposed by LEWIS (1967) naturally yield as products the elemental sulfur and sulfate observed in hydrous meteorites (see also STEGER and DESJARDINS, 1978). In this context we regard PCP as heterogeneous matrix components containing much of the oxidized and partially oxidized intermediates and products of sulfide alteration mixed with other poorly crystalline materials. The redox properties of sulfide permit facile oxidation under circumstances where oxidation of reduced carbonaceous material is slow. DUFRESNE and ANDERS (1962) have pointed out that magnetite, sulfate, reduced organic matter, carbonate and troilite can coexist at or near equilibrium with water. Thus, if H_2O were added to an anhydrous carbonaceous chondritic mineral assemblage containing troilite, partial conversion of the troilite to sulfate would have been thermodynamically spontaneous and, therefore, would not have required additional oxidants. The presence of elemental sulfur probably reflects kinetic barriers to its conversion to the thermodynamically stable sulfate, as first suggested by DUFRESNE and ANDERS (1962). Terrestrial geochemical cycles involving sulfur, sulfide, sulfite, and sulfate species are very poorly understood (MEYER *et al.*, 1979).

Variations in isotopic composition ($\Delta\delta^{34}S$) between sulfide and elemental S and sulfate within individual carbonaceous meteorites ranged from 0.2 to 5.6‰, with enrichment of ^{32}S in sulfate and elemental S relative to sulfide (KAPLAN and HULSTON, 1966; MONSTER *et al.*, 1965). From their isotopic study of troilite oxidation, LEWIS and KROUSE (1969) concluded that kinetic isotope effects associated with a low-temperature (<373 K) alteration oxidation process could account for the common occurrence of isotopically light sulfate and isotopically heavy sulfide. Sulfate may be derived from the most rapidly altered sulfide, whereas the intact sulfide may represent the slowest to alter. Our failure to detect H_2S above the 100-ppm level during acid treatment of Murchison and Nogoya is consistent with the preferential alteration of hydrolyzable sulfide and preservation of hydrolysis-resistant sulfide. Hydrolyzable sulfide occurred as a low proportion of the total S in the CI and CM carbonaceous chondrites examined by KAPLAN and HULSTON (1966).

The isotopic fractionation scheme proposed by MONSTER *et al.* (1965) to explain the sulfur isotope distribution among phases involved the hydrolysis-disproportionation of pre-existing elemental S according to the following general equation:

$$4S + 3H_2O \rightleftharpoons 2H_2S + S_2O_3^{2-} + 2H^+$$

Liquid water is required, and the H_2S and $S_2O_3^{2-}$ were postulated to be precursors for the sulfides and sulfates in carbonaceous meteorites. The experimental basis for the scheme has been criticized by LEWIS and KROUSE (1969). Moreover, elemental S is not predicted to appear in meteorites by any condensation theories; it has not been found in any anhydrous meteorites, and its presence in C3 meteorites (DUFRESNE and ANDERS, 1962) coincides with the occurrence of sulfate, carbonate and phyllosilicate, themselves the products of alteration processes. We believe that the available evidence, though not precluding the scheme of MONSTER *et al.* (1965), is most compatible with the view that sulfate and elemental S in CM and C3 meteorites result from alteration-oxidation of pre-existing sulfide. The latter alternative provides a model for the chemical evolution of sulfur-bearing phases that is consistent with that of other constituents of the hydrous matrices. It is possible that in meteorites that have undergone complete or nearly complete alteration—for example, Nogoya and Orgueil—second-generation sulfide and sulfate may have been formed from elemental sulfur that was already an alteration product derived originally from sulfide.

GOODING (1978) concluded from a thermodynamic study of probable gas–solid weathering reactions of precursive Martian surface rocks, that alteration products, derived from reactions in the 240–298 K temperature range, consist essentially of carbonates, iron oxides, clays (kaolinite) and $FeSO_4$ or $FeSO_4 \cdot H_2O$. Thus, the theoretical alteration assemblage on the Martian surface is very similar to the alteration products in CM meteorites, exclusive of organic compounds. Moreover, both are similar to terrestrial evaporite deposits formed in conjunction with volcanic or mafic rock detritus. Another interesting parallel may be made from preliminary near-infrared observations of Io's surface (POLLACK *et al.*, 1978, and POLLACK and BUNCH, unpublished data). Significant amounts of ferrous salts (possibly iron sulfates) appear to be present, which suggests an evaporite model for the development of Io's surface.

These somewhat loosely formed analogies seem to suggest a common development of iron sulfates on the surface of different planetary bodies, either through gas–solid or liquid–solid alteration reactions.

CONCLUSIONS

From evidence we have presented, coupled with existing data on CM meteorites, we believe the following conclusions are firmly based:

(1) CM matrix phases, primarily phyllosilicates, PCP, carbonate and some magnetite, were produced by aqueous alteration in a parent body at temperatures <400 K. Precursive matrix minerals were likely to have been Fe-rich ferromagnesian minerals (mostly olivine), metal, and Ni-bearing sulfides.

(2) Carbonaceous matter is predominantly associated with PCP in altered matrices. In mostly unaltered carbonaceous meteorites (C3's) and probably in precursive parents of CM's, carbonaceous matter is concentrated on olivine in the matrix as well as in dark haloes that surround some chondrules and aggregates, with higher abundances of C occurring in the halo olivines.

(3) Synthesis of the extractable organic compounds in carbonaceous meteorites may have occurred prior to or in the same environment as did aqueous alteration of anhydrous minerals. Whole or partial preservation of this organic matter may reflect the degree of overlap in episodes of synthesis and alteration. The occurrence of thermally labile organic compounds indicates that their synthesis did not take place prior to or simultaneously with hydrothermal alteration, if alteration occurred over months at temperatures much above 350 K. After their synthesis the environmental temperatures never exceeded the stability of the organic compounds.

(4) Parent body alteration processes are not incompatible with the known isotopic compositions of C, N, S and O in CM meteorites.

(5) CM meteorites were part of a parent body regolith in which the alteration process possibly took place in a partial open system: addition of at least H_2O, CO_2, and extractable organic compounds and partial removal of leached Cl and possibly Na.

Acknowledgements—We thank KEN LENNON for combustion analyses and J. ERLICHMAN for assistance in microprobe analyses. We also thank Professor C. B. MOORE (Arizona State University) for samples of Murray and Nogoya; Professor ANDERS, Dr MCSWEEN, Dr VAN SCHMUS and Dr KERRIDGE for critical reviews; and Dr H. HARTMAN for valuable discussions.

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