

HIGH RESOLUTION ELECTRON MICROSCOPIC CHARACTERIZATION OF PHYLLOSILICATES AND FINDING OF A NEW TYPE WITH 11Å STRUCTURE IN YAMATO-74662

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Abstract: Matrix materials in Yamato-74662 have been examined by HREM and AEM. Under microscopic observations, phyllosilicates in Yamato-74662 were found; i) as matrix materials, ii) in irregular cavities, and iii) replacing pyroxene and olivine grains. Five types of phyllosilicates in the matrix were observed by HREM; a) 7Å platy phyllosilicate, b) 7Å poorly organized tubular phyllosilicate, c) 17Å platy phyllosilicate, d) poorly organized 7Å layer structures with interlayering of 5Å layer structure and e) 11Å platy phyllosilicate. Parallel intergrowth of the 11Å mineral and 7Å berthierine, and characteristic HREM suggest that the 11Å mineral is a new phyllosilicate with an interstratified structure of serpentine-like and brucite-like layers. Mineralogy and texture of the matrix suggest that the phyllosilicates were derived by alteration from such precursor materials as CV chondrites, and experienced some mixing or brecciation process after (or during) alteration.

1. Introduction

Carbonaceous chondrites are, generally, considered to be the most primitive materials from the early solar system. Their origin is variously inferred (NAGY *et al.*, 1963; LARIMER, 1967; CAMERON, 1973, etc.). Although several classification schemes have been proposed (VAN SCHMUS and WOOD, 1967; WASSON, 1974; MCSWEEN, 1979), Yamato-74662 (Y-74662) can be designated as CM (or CM2) based on MCSWEEN's scheme (CI, CM, CO, CV, CR). Carbonaceous chondrites show various textures in petrographic observations. The petrographic feature that all carbonaceous chondrites share is the presence of significant quantities of dark, fine-grained matrix material containing carbon. Within the matrix materials are considerable amounts of chondrules, inclusions, isolated or aggregated olivine and pyroxene grains and opaque minerals. One of the most remarkable features in CI and CM is that the dark matrix is composed of significant amounts of phyllosilicates with lesser amounts of organic polymers, magnetite, and Fe- and/or Ni-bearing sulfides. Such minerals are very difficult to identify and characterize, and their characterization is one of the most urgent and significant challenges in the study of carbonaceous chondrites to elucidate their origin. Recent high resolution electron microscopic (HREM) studies of the phyllosilicate in the carbonaceous chondrites shed light on the characterization of these minerals. MACKINNON and BUSECK (1979), and AKAI (1979) found a new phyllosilicate in the Murchison meteorite which was not found on the earth yet; furthermore, AKAI

(1980a, b) observed new morphological type of this phyllosilicate. BARBER (1981) also examined four CM meteorites by HREM and characterized the matrix phyllosilicates and associated minerals.

The principal objectives in the present study are to examine the phyllosilicates in Y-74662 mainly by HREM and characterize them mineralogically, and to comment on the process of the formation of the phyllosilicates.

2. Experimental

2.1. Analysis by electron probe microanalyzer (EPMA)

A JXA-5A electron probe microanalyzer was used to determine the chemical composition of the specimens, operating at 15 kV and *ca.* 0.02 μ A specimen current. The electron beam used was about 2~3 μ m in diameter.

2.2. Analysis by analytical electron microscope (AEM)

The analytical electron microscope used was a combined type of H-500 equipped with an energy dispersive X-ray detector. Qualitative analysis was carried out. Accelerating voltage was 100 kV. Counting time for X-rays was 100~60 s.

2.3. High resolution electron microscopy (HREM)

Specimens for EM were prepared by cutting and ion-thinning an epoxy-impregnated petrographic polished thin section. For high resolution electron microscopy, a top entry type JEM 100 C electron microscope with Cs=1.4 mm was operated at 100 kV. An objective aperture of 60 μ m in diameter was used. The defocus value of -1000~0~1500Å (positive value; under focus, negative value; over focus) was usually used, and through-focus images were also taken.

The details of the simulation of electron microscopic images have been studied based on kinematical approximation (ISHIZUKA and UYEDA, 1975).

3. Specimen Description

3.1. General description (textural features)

A very small chondrite chip of Y-74662 was supplied and studied by HREM. Y-74662 is composed mostly of black brown matrix. Dispersed throughout the matrix are isolated or aggregated grains of olivine and pyroxene and other minerals. Many of the aggregated grains may be fragments of chondrules. The ratio of isolated or aggregated grains+chondrules+inclusions to the matrix is approximately equal to that of Murchison. Small cavities with irregular forms are scattered in the matrix and are filled with relatively pure yellowish brown phyllosilicates (Fig. 1).

Preferred orientation of the matrix phyllosilicates in Y-74662 was investigated by FUJIMURA *et al.* (1982).

3.2. Olivine

Olivine is an abundant phase, occurring as isolated crystal fragments or as aggregated grains (or chondrules). Many olivine grains have composition near Fo₀. Small micron-size grains of olivine were found in the matrix by EM.

3.3. *Ca-poor pyroxene*

Compositions of Ca-poor pyroxene occurring as isolated grains and aggregated grains (or chondrules) were usually near Fs_0 . Observations by EM indicated that small micron-size grains of enstatite are contained in the matrix. Disordered features will be discussed later.

3.4. *Ca-rich pyroxene*

Ca-rich pyroxenes occur as aggregated grains or as small grains along the inner wall (rim) of the irregular cavities. The composition of the rim diopside is shown in Table 1. Small grains of diopside were found in the matrix by EM. Exsolution lamellae in diopside will be mentioned later.

Table 1. Electron microprobe analysis of phyllosilicates in Yamato-74662.

	Black matrix				Yellowish brown		Phyllos. in cavity		Rim Di
SiO ₂	25.1	25.1	35.6	32.7	29.1	31.9	17.5	12.7	51.7
TiO ₂	0.0	0.0	0.2	0.1	0.2	0.2	0.3	0.2	0.1
Al ₂ O ₃	4.5	4.1	3.0	2.4	6.2	5.4	26.4	40.1	3.7
Cr ₂ O ₃	0.3	0.2	0.6	0.5	0.1	0.1	0.1	0.0	0.2
FeO	45.1	44.8	33.8	31.1	47.0	33.4	25.6	17.7	3.0
MnO	0.2	0.2	0.1	0.2	0.2	0.1	0.1	0.0	0.1
MgO	11.4	11.8	17.2	19.4	10.4	12.1	16.3	17.9	17.6
CaO	0.4	0.3	0.6	0.3	2.2	6.3	1.7	2.0	23.6
Na ₂ O	0.3	0.1	0.1	0.2	0.3	0.2	0.1	0.0	0.0
K ₂ O	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Total	87.3	86.6	91.2	86.9	95.7	89.7	88.1	90.6	100.0

Phyllos. in cavity: Phyllosilicate in irregular cavities.

Rim Di: Diopside grain in the inner wall (rim) of the irregular cavities.

3.5. *Other minerals in the matrix*

Other minerals found in the matrix are carbonate (CaCO₃), Fe/Ni sulfides.

4. Occurrences and Compositions

Phyllosilicates in Y-74662 are found in three different occurrences.

1) Phyllosilicates as matrix materials. Most of phyllosilicates are present as the matrix phase. Microscopic observations of the thin section indicate that only the matrix of a very thin portion is transparent and heterogeneous with yellowish brown~brown spotty textures. Observation by EM reveals that many small grains of anhydrous minerals and large quantities of amorphous material are contained in the matrix. The results of detailed examination of the matrix phyllosilicates by HREM will be shown later.

2) Phyllosilicates filling irregular cavities. They are relatively pure phyllosilicates, and are greenish brown~yellowish brown without pleochroism. The inner walls of the irregular cavities are sometimes rimmed with small grains of diopside. The phyllosilicates found in the irregular cavities in Y-74662 are very similar to those

found in Murchison (AKAI, 1980b). These textural features suggest that the cavities originated from inclusions.

3) Phyllosilicates replacing olivine and pyroxene. Figs. 2–5 show the phyllosilicates which partially replace olivine and pyroxene grains.

The major-element compositions of the phyllosilicates were obtained from EPMA analysis. The composition analyzed does not always imply a single phase, because the phyllosilicates contain many other submicroscopic mineral grains and amorphous materials. The compositions of the phyllosilicates occurred in three different modes are shown in Table 1 and Figs. 2–5. The compositions of relatively pure phyllosilicates in the irregular cavities can be interpreted as intermediate between berthierine and brucite. These compositions are rich in Al and are very similar to those of phyllosilicates in irregular cavities in Murchison (AKAI, 1980b). Compositions of CM matrix have been reported by FUCHS *et al.* (1973), MCSWEEN and RICHARDSON (1977) and BUNCH and CHANG (1980). Matrix composition in Y-74662 is similar to that in other CM chondrites.

5. HREM and AEM investigations

5.1. Amorphous materials and pyroxenes

Amorphous materials which show no distinct ED patterns are widely distributed in the matrix. Figure 6 shows the amorphous materials. They are composed mainly of Fe, S, Si, Ca, Cr, etc.

Micron-size pyroxene grains in the matrix phyllosilicates were found by EM observations. Stacking disorder in enstatite was characteristic (Fig. 7). Exsolution lamellae in diopside were found. Both thick (001) lamellae and thin (100) lamellae were observed (Fig. 8). This may be a clue to the cooling history (cooling rate) of the diopside grain.

5.2. Phyllosilicates

Because of small sizes of the phyllosilicates in irregular cavities and the phyllosilicates replacing olivine and pyroxene, attempts to characterize these phyllosilicates were unsuccessful. Phyllosilicates in the matrix were closely examined by HREM. Phyllosilicates found in Y-74662 matrix can be divided into the following five types in structure and morphology mainly by HREM.

a) 7Å platy phyllosilicate. Phyllosilicate of this type is considered to be berthierine. It is characterized by stacking disorder. Figure 9 shows an ED pattern of the b^*c^* reciprocal lattice plane. It is composed of regularly arranged spots in $k=3n$ levels and streaks in $k \neq 3n$ levels. These features indicate a disordered structure with $(n/3)b$ displacements. This feature is similar to that in Murchison (AKAI, 1980b). Qualitative investigation on the compositions of the phyllosilicates by AEM suggests considerable compositional (especially, Fe/(Mg+Fe) ratio) fluctuation; $I_{Fe}/(I_{Mg}+I_{Fe})$ ratio changes from 0.61 to 0.86. An example of the AEM analysis is shown in Fig. 10.

b) 7Å-poorly organized tubular phyllosilicates. These may correspond to a chrysotile structure of low crystallinity (Fig. 11).

c) 17Å platy phyllosilicates. These may be the same as found in the Murchison

Fig. 1. Photomicrograph of matrix in Y-74662 (opened polars). Scale bar is 0.1 mm. The irregular cavity is filled with relatively pure phyllosilicates. The inner wall of the cavity is rimmed by small grains of diopside.

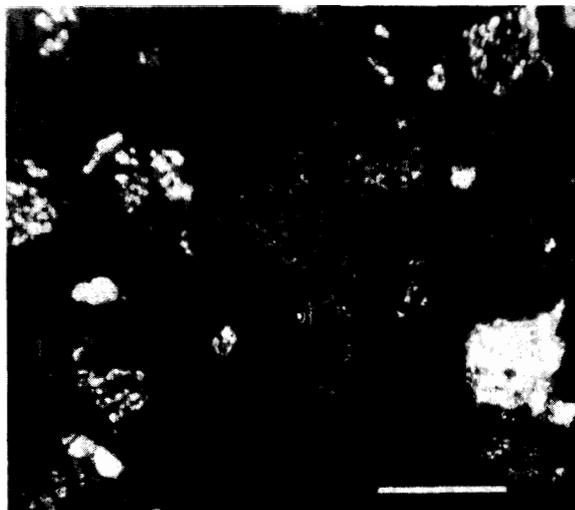


Fig. 2. Photomicrograph of Y-74662 (opened polars). A part of olivine crystal aggregate is replaced by phyllosilicates. Scale bar is 0.1 mm.

	Phyllosilicate (O)	Olivine
SiO ₂	36.2	42.7
TiO ₂	1.0	0.0
Al ₂ O ₃	4.6	0.0
Cr ₂ O ₃	0.0	0.3
FeO	14.0	0.7
MnO	0.1	0.0
MgO	15.6	56.4
CaO	12.7	0.3
Na ₂ O	0.1	0.0
K ₂ O	0.0	0.0
Total	84.3	100.4

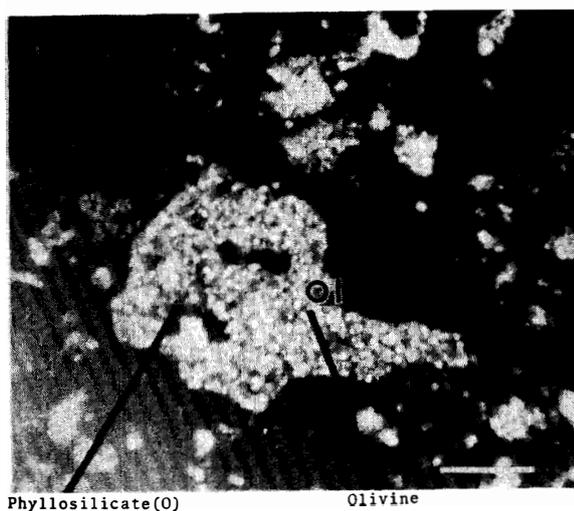
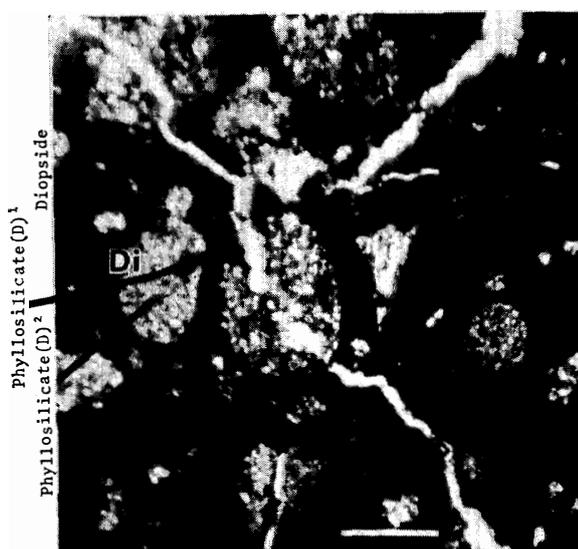


Fig. 3. Photomicrograph of Y-74662 (opened polars). A part of diopside crystal aggregate is replaced by phyllosilicates. Scale bar is 0.1 mm.

	Diopside	Phyllo- silicate(D) ¹	Phyllo- silicate(D) ²
SiO ₂	50.7	23.8	23.9
TiO ₂	0.4	0.1	0.1
Al ₂ O ₃	7.1	5.7	11.2
Cr ₂ O ₃	1.1	0.2	0.4
FeO	1.2	46.3	35.2
MnO	0.4	0.1	0.2
MgO	21.1	9.2	11.1
CaO	18.8	0.3	0.3
Na ₂ O	0.0	0.3	0.0
K ₂ O	0.0	0.0	0.0
Total	100.8	86.0	82.4



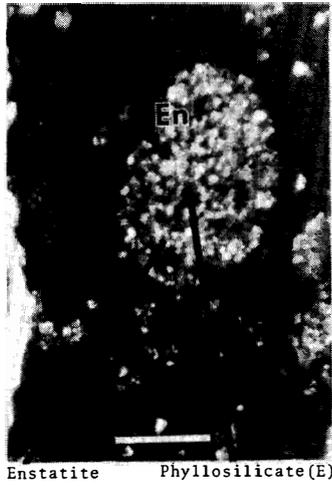


Fig. 4. Photomicrograph of Y-74662 (opened polars). A part of enstatite aggregate is replaced by phyllosilicates. Scale bar is 0.1 mm.

	Enstatite	Phyllo-silicate (E)
SiO ₂	57.4	39.1
TiO ₂	0.2	0.2
Al ₂ O ₃	1.9	6.9
Cr ₂ O ₃	0.7	0.2
FeO	1.2	34.2
MnO	0.0	0.2
MgO	37.8	12.1
CaO	1.0	2.4
Na ₂ O	0.0	0.2
K ₂ O	0.0	0.0
Total	100.2	95.5

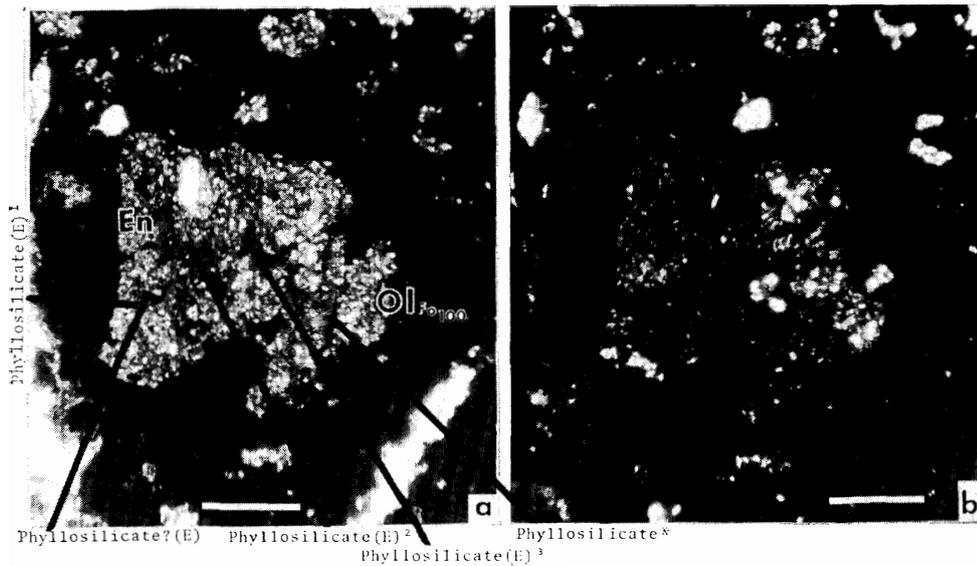


Fig. 5. Photomicrographs of Y-74662 (a: opened polars, b: crossed polars). Scale bar is 0.1 mm.

	Phyllo-silicate (E) ¹	Phyllo-silicate? (E)	Phyllo-silicate (E) ²	Phyllo-silicate (E) ³	Phyllo-silicate*
SiO ₂	26.2	45.1	30.6	28.5	22.6
TiO ₂	0.1	0.5	0.1	0.2	0.1
Al ₂ O ₃	3.4	3.3	4.6	4.3	4.0
Cr ₂ O ₃	0.4	2.1	0.5	0.7	0.2
FeO	39.5	6.7	40.0	26.6	45.7
MnO	0.1	0.4	0.2	0.1	0.1
MgO	9.9	19.3	10.7	13.8	7.9
CaO	1.1	14.1	0.5	3.4	0.2
Na ₂ O	0.4	0.1	0.4	0.1	0.3
K ₂ O	0.0	0.0	0.0	0.0	0.0
Total	81.1	91.6	87.6	77.7	81.1

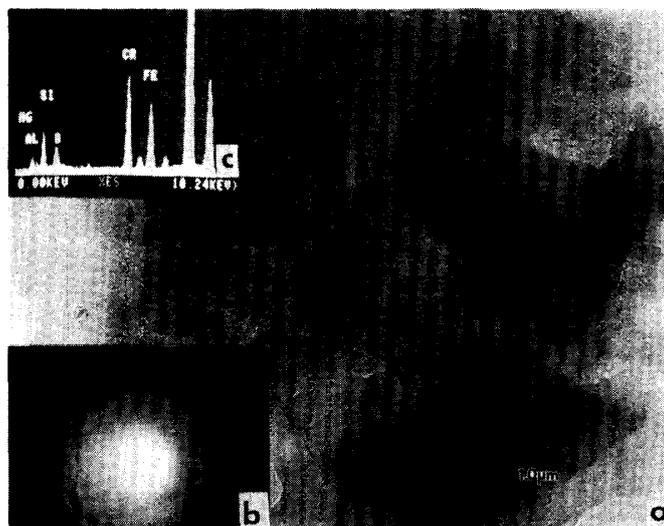


Fig. 6. *a*: Electron micrograph showing amorphous materials in the matrix. *b*: ED pattern. It shows no distinct ED pattern. *c*: Analysis by AEM.

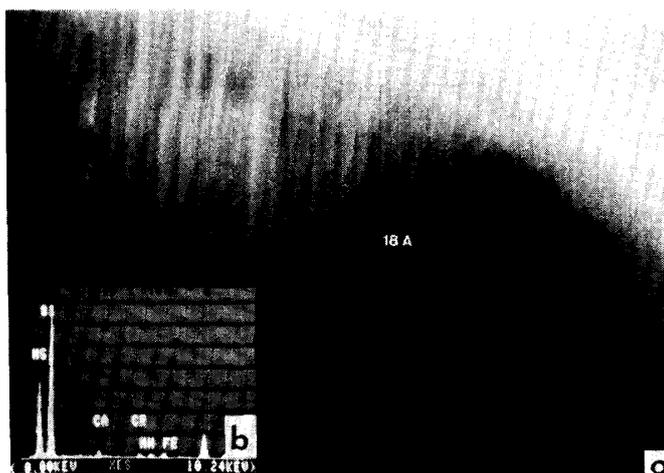


Fig. 7. *a*: Electron micrograph of disordered structure found in micron-size enstatite grain in the matrix. *b*: Analysis by AEM.

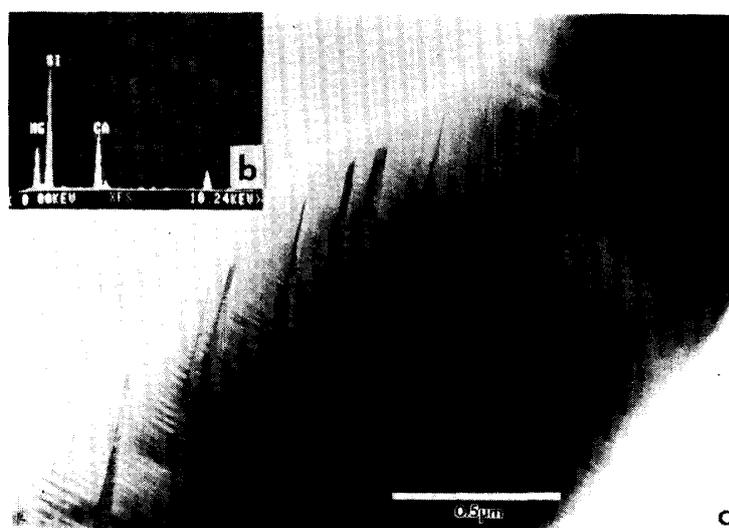


Fig. 8. *a*: Electron micrograph of small diopside grain in the matrix. A tweed-like texture composed of (100) and (001) exsolution lamellae is found. *b*: Analysis by AEM.

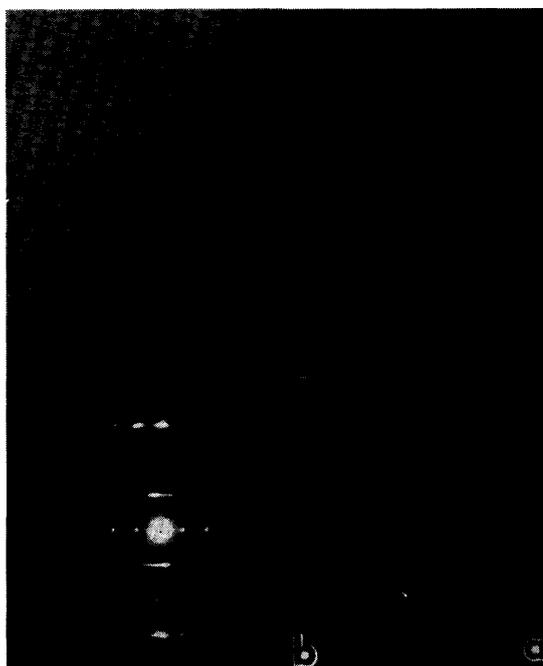


Fig. 9. a: High resolution electron micrograph of 7 Å phyllosilicate (serpentine structure). b: ED pattern.

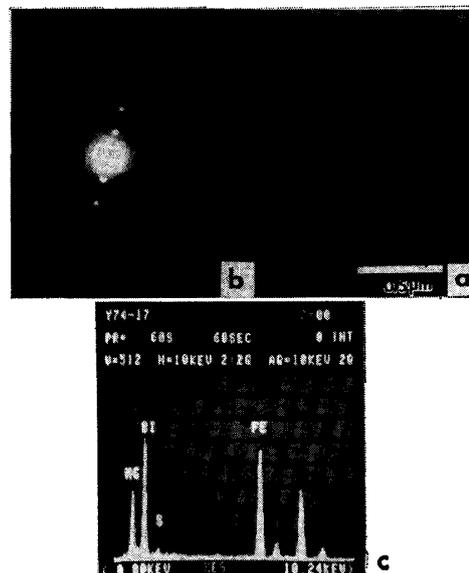


Fig. 10. a: Electron micrograph of 7 Å phyllosilicate. b: ED pattern. c: Qualitative analysis by AEM.

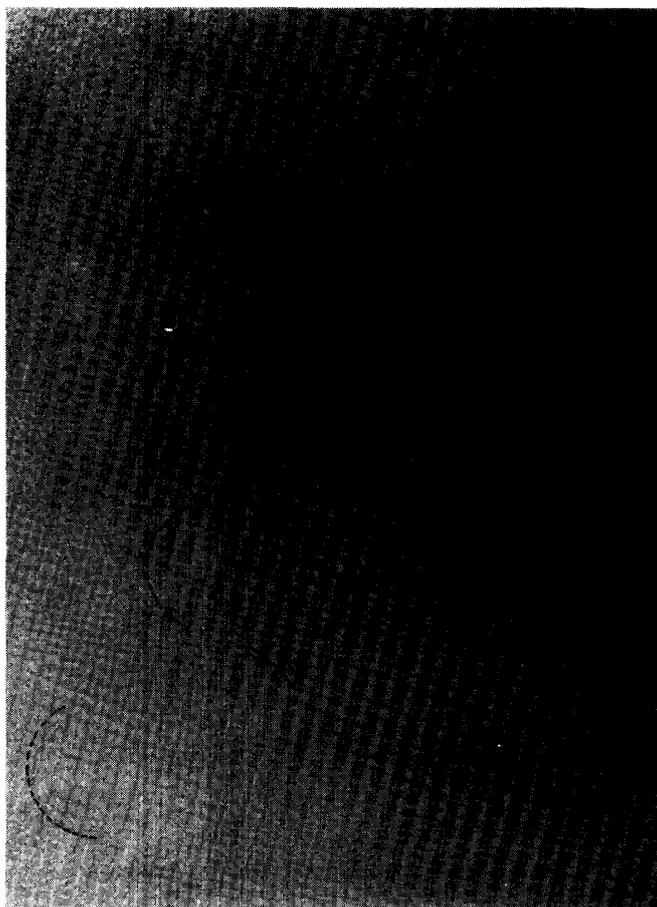


Fig. 11. High resolution electron micrograph of a section of the poorly organized tubular phyllosilicate with 7 Å structure.

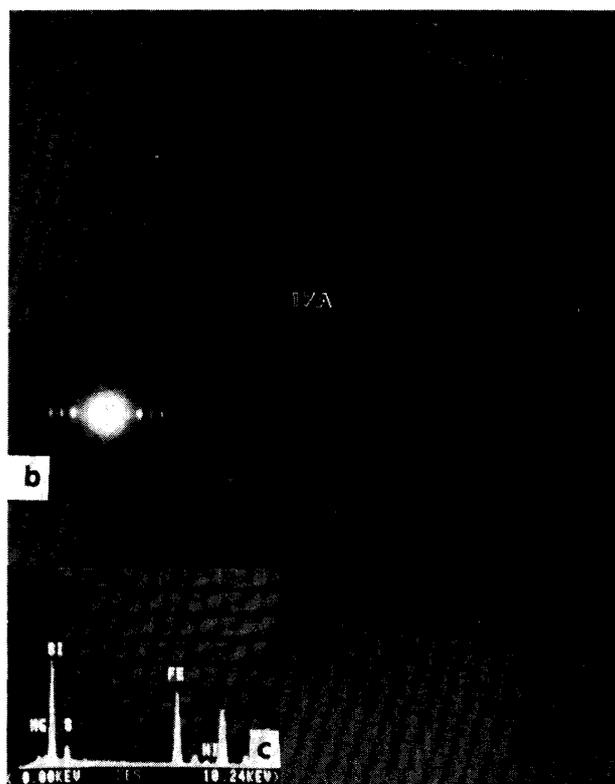


Fig. 12. *a*: High resolution electron micrograph of a "new" phyllosilicate (1:1:1×2 type). *b*: ED pattern. *c*: Analysis by AEM. Small peak of S and Ni may be due to sulfide contamination.

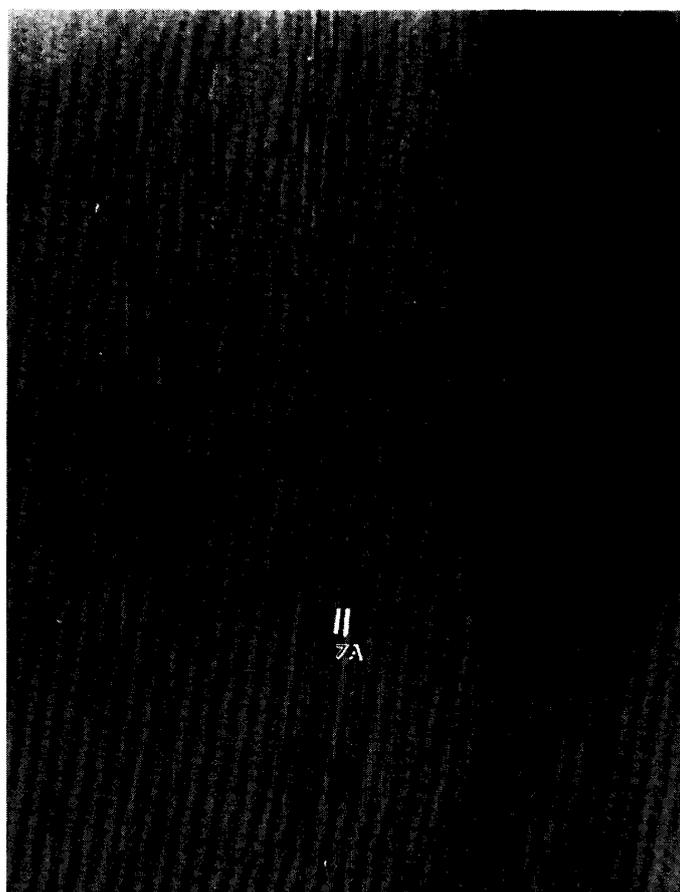


Fig. 13. High resolution electron micrograph of poorly organized interlayering of 7Å and 5Å layer structures. It is composed of only several lattice fringes. S: serpentine layer, B: brucite layer.

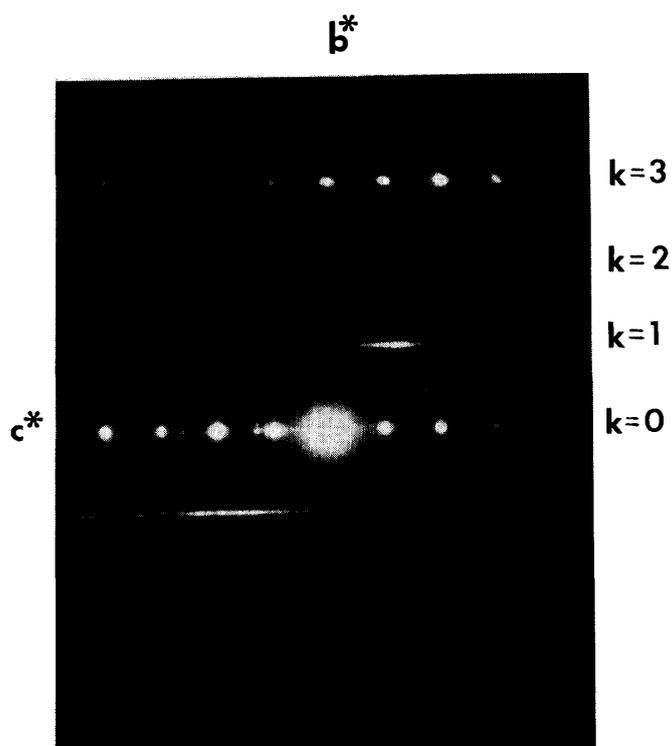


Fig. 14. Electron diffraction pattern of a phyllosilicate representing the b^*c^* reciprocal lattice plane. The 11.0\AA^{-1} spots-array is accompanied by the 7.3\AA^{-1} spots-array. Regularly arranged one spots-array and two streaks along c^* suggest a stacking disorder with $(n/3)$ b displacement.

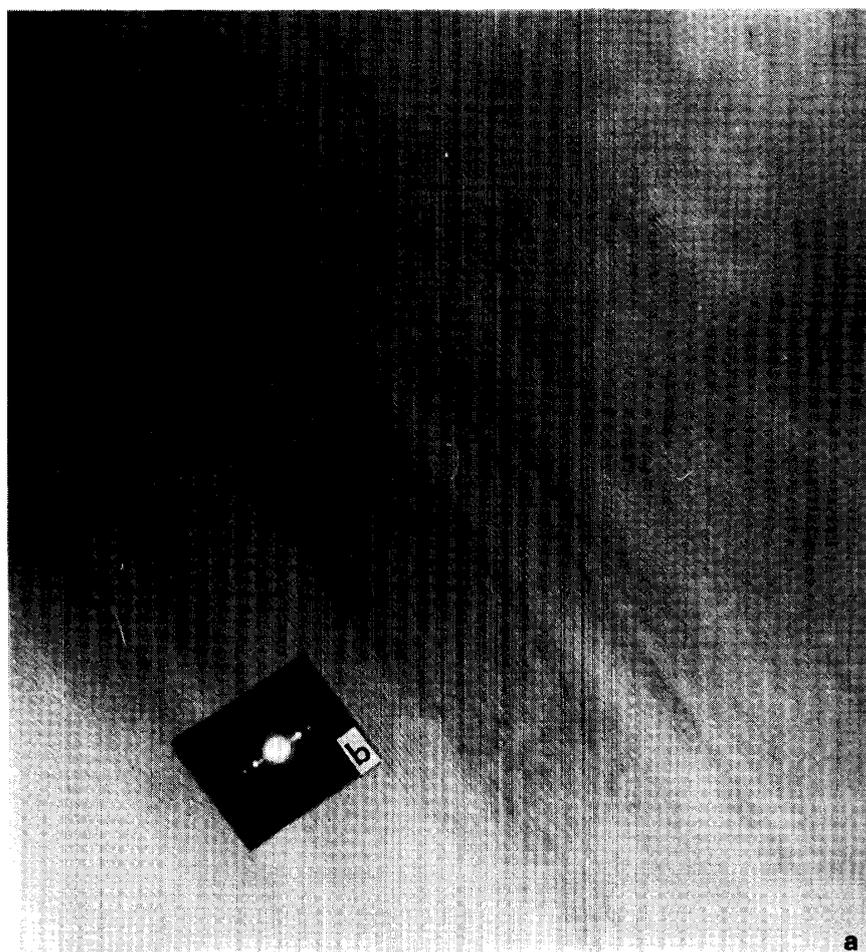


Fig. 15. a: Electron micrograph indicating parallel intergrowth of 7\AA and 11\AA layer lattice phyllosilicates. b: ED pattern.

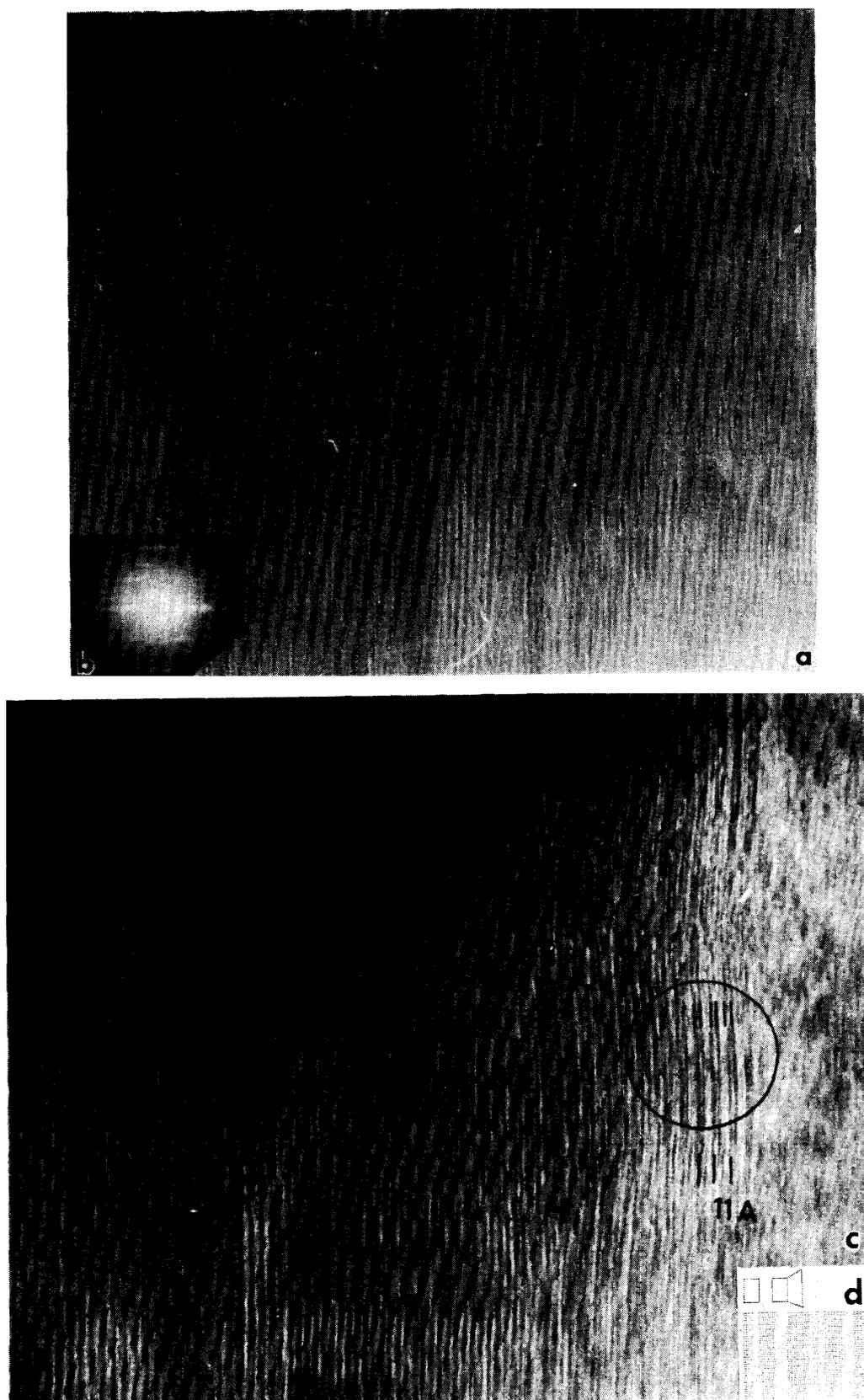


Fig. 16. *a, c*: High resolution electron micrographs of an 11 Å layer lattice structure. *b*: SAED pattern. *d*: Simulated EM image and the corresponding structure model.

meteorite (MACKINNON and BUSECK, 1979; AKAI, 1979, 1980b) (Fig. 12). It was determined to be an extremely Fe-rich silicate by AEM (Fig. 12c). However, the tubular form of this 17Å interstratified mineral as found in Murchison (AKAI, 1980a, b) was not observed in Y-74662.

d) Poorly organized 7Å layer structures with interlayering of a 5Å layer structure (Fig. 13). It can be interpreted as interlayering of serpentine- and brucite-structures. Only a few 5Å and 7Å lattice fringes form interlayered structure.

e) 11Å platy phyllosilicates. Parallel intergrowth of this 11Å phyllosilicate and 7Å platy phyllosilicate (type a) was observed (Figs. 14 and 15). This 11Å platy phyllosilicate may be the same one as found in Murchison (AKAI, 1980b). The structural scheme was preliminarily suggested by AKAI (1980b); that is, an interstratified structure consisting of a 1:1 layer plus a brucite layer (1:1:1 type). The 11Å spacing seems to be a little small for the regular stacking of a serpentine layer and a brucite layer. Thus the possibility of another and unknown type of layer structure cannot be denied completely. Regardless such ambiguities and lack of AEM data, the parallel intergrowth of the 11Å structure and 7Å serpentine structures, and characteristic lattice images (Figs. 16a and 16c), strongly suggest that the 11Å structure is a silicate with an interstratified structure consisting of a serpentine-like structure and a brucite-like structure. It is observed that the 11Å structure is composed of one wide lattice fringe (7Å) and one narrow fringe (in the circle in Fig. 16c). At this point, the result of simulation of the 11Å phyllosilicate (Fig. 16d) is consistent with the observation (Fig. 16c).

6. Summary and Discussion

The genesis of the matrix phyllosilicates in CI and CM has variously been interpreted; as an aqueous alteration product of anhydrous minerals on the parent body or planetesimal (FISH *et al.*, 1960; DUFRESNE and ANDERS, 1962; NAGY *et al.*, 1963; BOSTRÖM and FREDRIKSSON, 1966; BUNCH and CHANG, 1978, 1980); as a product of the final stage of nebular condensation (LORD, 1965; LARIMAER, 1967; GROSSMAN, 1972); as residual interstellar grains (CAMERON, 1973); as a residue derived from comets (HEYMANN, 1978); and derived from interplanetary dust and planet rings (WILKENING, 1978).

The results of the present investigation and the previous data on CM meteorites suggest the following possible genesis of Y-74662.

6.1. Possible genesis of Y-74662

The overall textural relationships of the phyllosilicates, other matrix phases, chondrule fragments, and irregular cavities in Y-74662 strongly suggest that the phyllosilicates were derived by alteration from such precursor materials as CV meteorites (*e.g.* Allende). This conclusion is essentially consistent with those of DUFRESNE and ANDERS (1962), NAGY *et al.* (1963) and BUNCH and CHANG (1980).

The irregular cavities filled with phyllosilicates indicate a similarity to those found in hydrothermally altered tuff breccias on the earth. Such textures are characteristic of a process related to aqueous activity. The presence of diopside grains often found along the inner wall of the cavities seems to be evidence that they were originally in-

clusions such as often found in CV chondrites (*e.g.* Allende). The inclusions might be altered and then the cavities might be filled with phyllosilicates deposited from Fe-rich aqueous solutions (and/or gaseous phases). That is, this texture can be interpreted as 'pseudomorphs' of inclusions.

The compositions of the phyllosilicates formed in olivine or pyroxene (aggregate) differ greatly from those of the host minerals and are very Fe-rich. This suggests that the aqueous solutions (or gaseous phases) were very Fe-rich. The origin of the Fe might be from iron metal of Fe/Ni sulfides in the precursor materials by reaction with water or a gaseous phase.

Phyllosilicates in the matrix phase are not unique in structure, morphology, and composition, but are composed of various types (at least five types). This suggests that the phyllosilicates were formed in nonequilibrium conditions or that they experienced some brecciation process or some other mixing process by impact or another event after (or during) alteration. The fact that the unaltered anhydrous minerals in CM (*e.g.* the extreme case is euhedral olivine crystals in Murchison: FUCHS *et al.*, 1973; AKAI, 1980b) coexist with matrix phyllosilicates may also suggest this multistage process; that is, predominant alteration and accompanied mixing process.

6.2. Condition of the phyllosilicates formation

The presence of the 17Å and 11Å new phyllosilicate (1:1:1×2 and 1:1:1 type respectively) in Y-74662 suggests an environmental condition of phyllosilicate formation which has not yet been found on the earth. The condition of the phyllosilicate formation is estimated to be near the condition of coexisting serpentine and brucite, but more detailed experimental work is necessary.

The same phyllosilicates paragenesis and mineralogical features containing the 17Å and 11Å phyllosilicates found both in Y-74662 and Murchison strongly suggest a similarity in the environmental condition of phyllosilicate formation and their genetical relations.

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