

COMPARISONS OF TEXTURAL AND CHEMICAL VARIATIONS OF
MINERALS IN SOME PRIMITIVE ACHONDRITES AND
AN H7 CHONDRITE, WITH REFERENCE TO THEIR
FORMATION AND COOLING HISTORIES

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Abstract: The textures and trends of chemical variations of minerals in five acapulcoites (Acapulco, Allan Hills (ALH)-77081, ALH-78230, ALH-81187 and ALH-81261), two winonaites (Tierra Blanca and Yamato (Y)-74025) and an H7 chondrite (Y-75008) were studied for comparison to deduce their formation conditions and subsolidus cooling histories. The mineral distribution maps of polished thin sections (PTSs) of ALH-78230, Tierra Blanca, Y-74025, Y-75008, were made from elemental distribution maps of the electron probe microanalyzer to study variations in the distributions of minerals. Textures of these meteorites are fine-grained recrystallized chondrite-like materials including minerals known in chondrites.

Opaque mineral grains have complex shapes, often with large aggregates of Fe-Ni metals, as was observed in Y-74025. Elongation parameters defined as $L/(4\pi S)^{1/2}$ ($=A$), range from 1.4 to 1.6 for acapulcoites, where L is length and S is area of a grain. Despite their formation in different parent bodies, the variety of textures of primitive achondrites suggests that heterogeneity produced by local segregation of partially melted materials on their parent bodies can explain the origin of variation of primitive achondrites. CaO zoning of orthopyroxene can be attributed to the segregation and migration of Ca-rich partial melts. Mg/Fe variations of olivine can be explained by subsolidus reduction, when their parent body was disrupted or fractured by an impact while its interior was still hot.

1. Introduction

This paper constitutes the second part of extensive studies of the primitive achondrites carried out by YUGAMI (1996) and her coworkers. Although winonaites/IAB iron meteorites (*WI*-group) and acapulcoites/lodranites (*AL*-group) are from two different parent bodies on the basis of their oxygen isotope compositions (CLAYTON and MAYEDA, 1996), both sub-groups of primitive achondrites were formed by similar processes (YUGAMI *et al.*, 1998), involving orthopyroxene, olivine, plagioclase, augite, troilite, chromite, Ca-phosphates and FeNi-metal. Comprehensive studies have also

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been conducted of these meteorite groups by many investigators (e.g., KIMURA *et al.*, 1992; NAGAHARA, 1992; TAKEDA *et al.*, 1994; BENEDIX *et al.*, 1996, 1998; MITTELFEHLDT *et al.*, 1996; MCCOY *et al.*; 1997a,b), but their formation models differ in some respects. They all accept the partial melting model.

Somewhat different classification methods were employed among the previous investigators (e.g., PRINZ *et al.*, 1980, 1983; KIMURA *et al.*, 1992; MCCOY *et al.*, 1993, 1996). In our previous paper, we tentatively used criteria based on grain size. According to this definition of MCCOY *et al.* (1993, 1997a), EET84302 and Gibson were classified as lodranites. Clues from trace element distributions of individual minerals (FLOSS, 1998), indicate that EET84302 has more affinities to acapulcoites. YUGAMI *et al.* (1998) reported that the ratios of volume percentage of olivine-orthopyroxene (plagioclase + augite + phosphate) of the primitive achondrites show an enrichment trend with the plagioclase-augite-component. They considered that the plagioclase-augite-rich region of Caddo County B3A (TAKEDA *et al.*, 1997) could be taken as an end-member of the variations. Some parts of EET84302 are enriched in such a component and metal (TAKEDA *et al.*, 1994). The orthopyroxene-rich region of EET 84302,19 and olivine-rich Y-74357 are residues of segregated materials. MCCOY and CARLSON (1998) observed metal segregation in GRA95209 and LEW86220. Sub-group names have been assigned for all these end members, except for plagioclase-augite-rich materials. All meteorite samples in this study can be classified as recrystallized chondritic groups (winonaite and acapulcoite) without extensive segregation, except for a part of Y-74025.

In our previous study, quantitative modal abundances of minerals have been obtained using mineral distribution maps of polished thin sections (PTSs) of six winonaite, two silicate inclusions in IAB iron meteorites, seven acapulcoites, and six lodranites. However, because of the limited space for publication of color maps, we could cite only four such maps (YUGAMI *et al.*, 1998). In this paper, we utilize four mineral distribution maps including that of an H7 chondrite (Y-75008) made from elemental distribution maps of the electron probe microanalyzer, to study variations in the distributions of minerals. The trends of variations of chemical compositions of minerals were also studied from these data, in order to deduce formation processes and subsolidus cooling histories.

2. Samples and Techniques

Polished thin sections (PTSs) of four winonaite (Tierra Blanca, Winona, Y-74025,52-4, Y-75305,52-2) and five acapulcoites (Acapulco, ALH-77081,4, ALH-78230,51-2, ALH-81187,16, ALH-81261,14) were studied by the electron probe microanalyzer (EPMA). The Y-75008 H7 chondrite was also studied for comparison. These samples were supplied by the National Institute of Polar Research, the Meteorite Working Group of the U.S. and the Planetary Materials Database Collections of the University of Tokyo.

These PTSs were investigated by EPMA, JEOL JXA-733 at the Ocean Research Institute and the Geological Institute, and JEOL JXA-8900L at the Geological Institute, University of Tokyo. Two-dimensional CMA techniques of EPMA have been applied

to four PTSs to obtain elemental distribution maps of nine elements (Si, Mg, Ca, Fe, Al, Cr, P, S, Ni). Mineral distribution maps of four primitive achondrites and a chondrite (Y-75008) were made from the CMA data processed by using the public domain program NIH Image version 1.57. Distribution maps of eleven minerals (olivine, orthopyroxene, augite, plagioclase, Fe-Ni metal, troilite, chromite, Ca-phosphate, schreibersite, and daubreelite, and weathering products (Fe-hydroxides)), of each PTSs were made and combined (YUGAMI, 1996). Modal abundances of minerals of these samples were obtained by counting the number of pixels for each mineral in the mineral distribution maps. The errors of the modes of minerals are discussed in YUGAMI *et al.* (1998).

Variations in chemical compositions of orthopyroxene and olivine were analyzed along a line by EPMA and their concentrations *versus* distances were plotted along a line from core to rim. The chemical compositions of major minerals have been published by YUGAMI *et al.* (1998).

3. Results

3.1. Descriptions of thin sections of H7 and WI group Y-75008

This sample is a chondrite, but shows an extensively recrystallized texture (H7) (Fig. 1a) comparable to those of primitive achondrites (YANAI and KOJIMA, 1995). The grain sizes of silicate minerals are about 0.1–0.4 mm in diameter. The mineral distribution map (Fig. 2a) shows an uneven distribution of fine plagioclase grains and moderately sized mafic silicates.

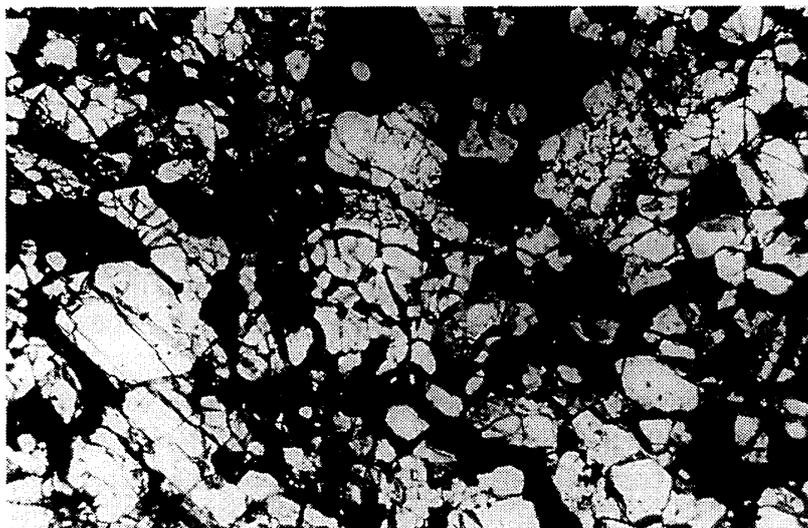
The average *fe*# ($100 \times \text{Fe}/(\text{Fe} + \text{Mg})$, atomic ratio) of olivine is 18.6 and that of orthopyroxene ($\text{En}_{79.9}\text{Fs}_{16.1}\text{Wo}_{4.0}$) is 16.8. These *fe*#s are typical of H chondrites and larger than those of all types of primitive achondrites. Y-75008 has orthopyroxene with cores that have lower CaO contents than rims. Chromite grains show Al_2O_3 zoning, with Al_2O_3 increasing at first and then decreasing from the cores to the rims. The modal abundances of minerals were given by YUGAMI *et al.* (1998).

Y-74025

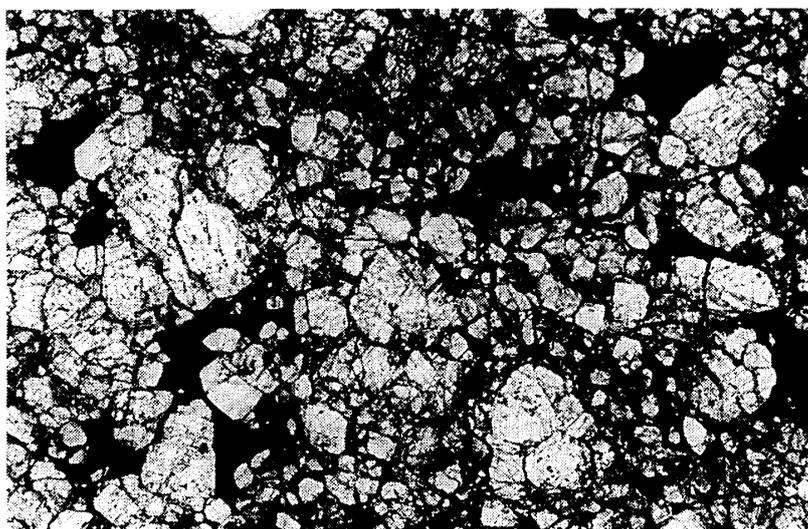
The PTS (Y-74025,52-2) was investigated by KIMURA *et al.* (1992), but we obtained a mineral distribution map (Fig. 2b) of Y-74025,52-4, which contains a large opaque grain. The entire area of this PTS is 38.7 mm². Y-74025,52-2 includes no large opaque grains. Silicate grains are fine (0.1–0.25 mm) and rather dusty. The modal abundances of minerals were given by YUGAMI *et al.* (1998). The largest opaque grain is mainly troilite (Cr 0.35 wt%), with minor FeNi metal and daubreelite in one opaque grain, and its area is 1.3 mm² (3 volume (vol)%, $A = L/(4\pi S)^{1/2} = 5.4$). Ni in kamacite is 6.49 wt%.

Y-75305

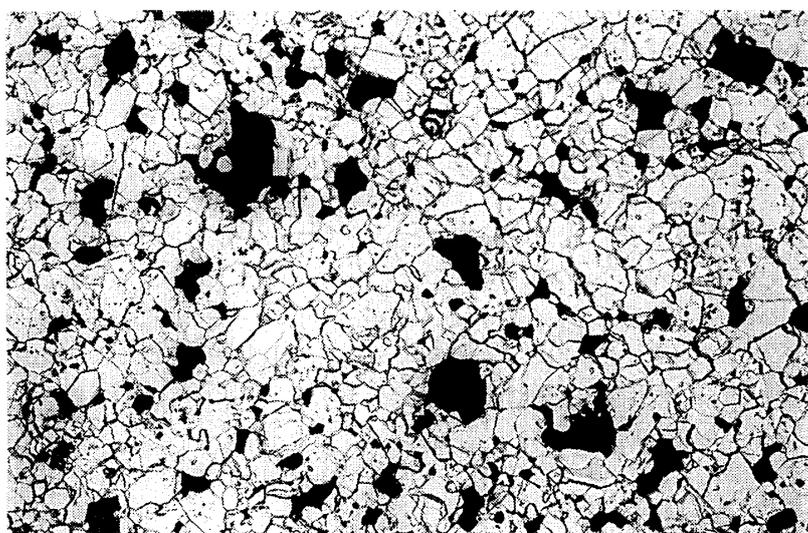
The same PTS (Y-75305,52-2) was investigated previously by KIMURA *et al.* (1992). The entire area of PTS Y-75305,52-2 is 20.4 mm². The area of the largest opaque grain is 2.2 mm² (11 vol%, $A = 3.9$). The modal abundances of minerals were



a. Y-75008.
Width is 2.5 mm.



b. ALH-81187.
Width is 3.3 mm.



c. ALH-81261.
Width is 3.3 mm.

Fig. 1. Photomicrographs of primitive achondrites. Transmitted light.

given by YUGAMI *et al.* (1998). The original grain seems to have been larger because it is cut by the edge of the PTS. The large metallic grains in PTS,52-2 nearly enclose a cluster of silicate grains. PTS,52-1 roughly looks like PTS,52-2. Ni in kamacite is 7.25 wt% and Cr in troilite is 0.37 wt%.

Winona

Winonaites were named after this meteorite (PRINZ *et al.*, 1980), but it is so seriously weathered that it may not be appropriate to be claimed as a type specimen. Silicate and Ca-phosphate grains are buried in considerable amounts of weathering products. Silicate grains are about 0.05–0.12 mm in diameter. This PTS includes a rather large Ca-phosphate grain (about 0.45 mm across). Modal abundances of silicates and Ca-phosphates were obtained by the same technique as those of the other samples in this study: olivine 23 vol%, orthopyroxene 45.4%, augite 0.46%, plagioclase 22.8% and Ca-phosphate 8.35%. The *fe*# of orthopyroxene is 6.7 and that of olivine is 3.7. CaO distribution of orthopyroxene is constant (Fig. 3).

Tierra Blanca

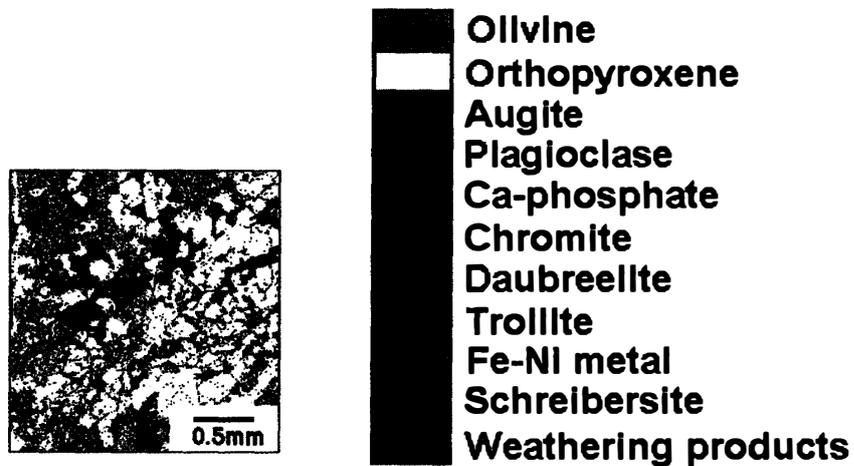
Tierra Blanca is fine-grained (0.1–0.2 mm in diameter) and most of the opaque materials that fill the interstices of silicate grains are weathering products. The mineral distribution map (Fig. 2c) shows fairly even distribution of minerals. The composition of the Tierra Blanca orthopyroxene is almost constant, $\text{Ca}_{2.2}\text{Mg}_{90.7}\text{Fe}_{7.1}$ and *fe*# = 7.4, but the *fe*#s in olivine grains show reverse zoning (Fig. 4). The cores of olivine grains have *fe*# = 5.4 and the rims have *fe*# = about 3.5. These values are larger than those of KING *et al.* (1981) and some other winonaites (KIMURA *et al.*, 1992). The modal abundance of minerals was given by YUGAMI *et al.* (1998).

The equilibration temperature of 900°C–1100°C was estimated from the orthopyroxene composition of Tierra Blanca by pyroxene geothermometry (LINDSLEY, 1983). A temperature of 1200°C by two pyroxene geothermometry has been reported (BENEDIX *et al.*, 1998).

3.2. Descriptions of thin sections of AL-group

Acapulco

Acapulcoites (*e.g.*, PALME *et al.*, 1981; ZIFFEL *et al.*, 1995) are named after this meteorite. Acapulco is slightly coarser-grained (up to about 0.4 mm across) than two acapulcoites described below (Fig. 1b, c). The modal abundances of minerals obtained from our mineral distribution map were reported by YUGAMI *et al.* (1998). The present description emphasizes the presence of dusty inclusions in orthopyroxene and its relationship to the chemical zoning of the CaO contents. The dusty inclusions are oriented along the *c*-direction of the orthopyroxene grains. The CaO contents show zoning (Fig. 3) from the dusty cores to the rims of orthopyroxene as was observed for some lodranites by MIYAMOTO and TAKEDA (1994). The CaO content at the dusty core is low (about 0.6 wt%) and it increases away from the core (about 1.1 wt%) at first and then decreases toward the rim (1.0 wt%) facing to mostly olivine (TAKEDA and MIYAMOTO, 1994). The trends do not change significantly depending on the minerals facing the orthopyroxene. The zigzag distributions of the dots in Fig. 3 express the



a. Y-75008 H7 chondrite.



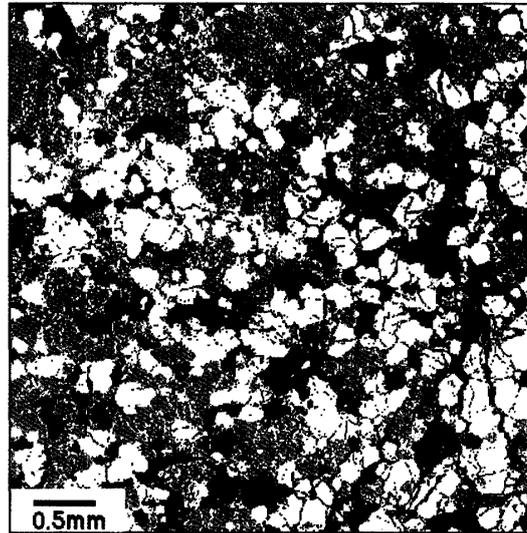
b. Y-74025,52-4 winonaite.

Figs. 2. Mineral distribution maps of primitive achondrites.

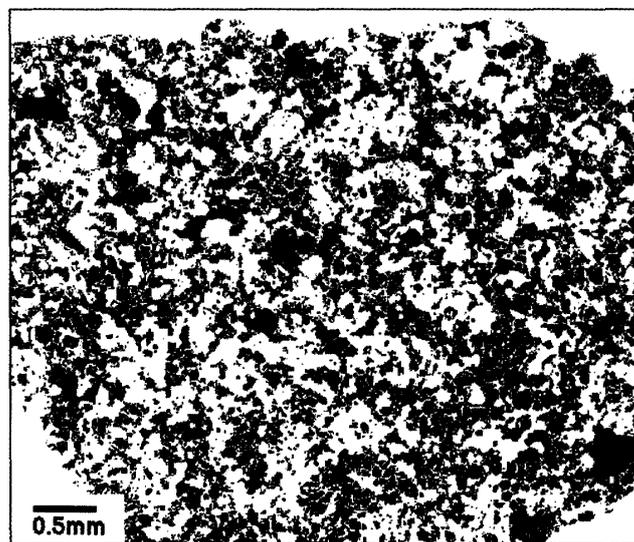
range of errors of the measurements. The $fe\#$ s of olivine (12.0) and orthopyroxene (10.7) are slightly higher than those of other acapulcoites (Fig. 4). The PTS of Acapulco we measured contained higher modal abundances of plagioclase than chondrites (e.g. Y-75008). EET84302,28 showed a similar trend. This observation suggests that some parts of acapulcoite parent body are enriched in the plagioclase-rich gabbro lithology.

ALH-77081

This meteorite is the first primitive achondrite recovered from Antarctica (e.g.,



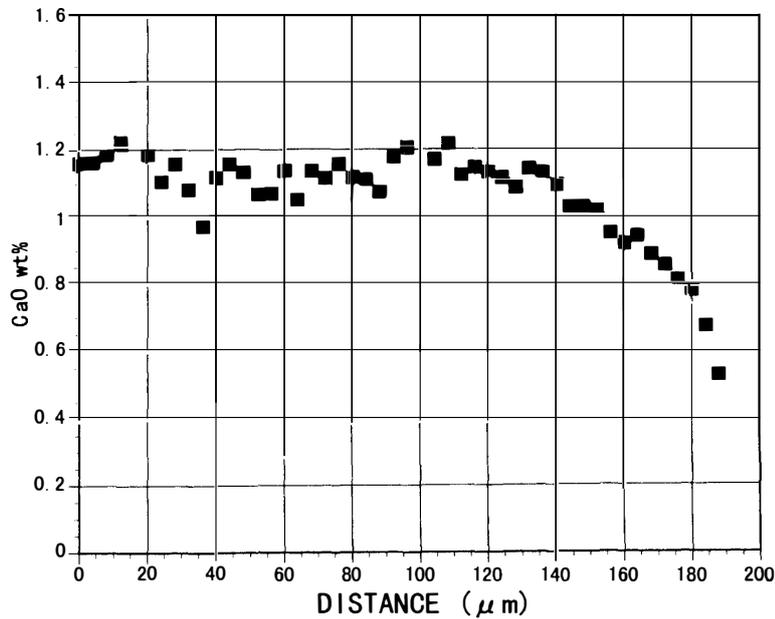
c. Tierra Blanca winonaite.



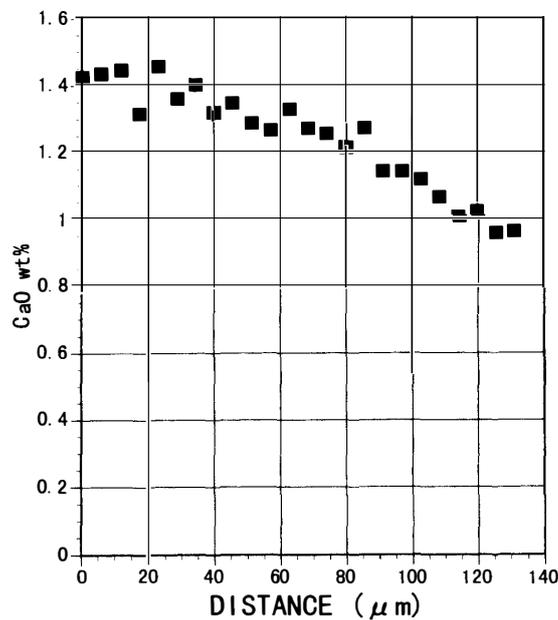
d. ALH-78230 acapulcoite.

Fig. 2 (continued).

TAKEDA *et al.*, 1980; SCHULTZ *et al.*, 1982). ALH-77081 has a fine-grained (up to about 0.25 mm across) recrystallized texture which is typical of acapulcoites. The PTS size is small (about 3.5 mm \times 1.8 mm). The modal abundances of minerals obtained from the mineral distribution map were given by YUGAMI *et al.* (1998). The *fe* # of olivine (10.3)



a. Gibson.



b. Y-791491.

Fig. 3. CaO zoning of orthopyroxenes in primitive achondrites (from core to rim).

and orthopyroxene (10.2) are similar to many acapulcoites and $fe\#$ s of olivine and orthopyroxene are not zoned. The CaO content shows zoning, decreasing toward the rims of the orthopyroxene grains. Opaque grain shapes are the least complicated in the AL group (The average A of the largest five opaque grains is 1.6).

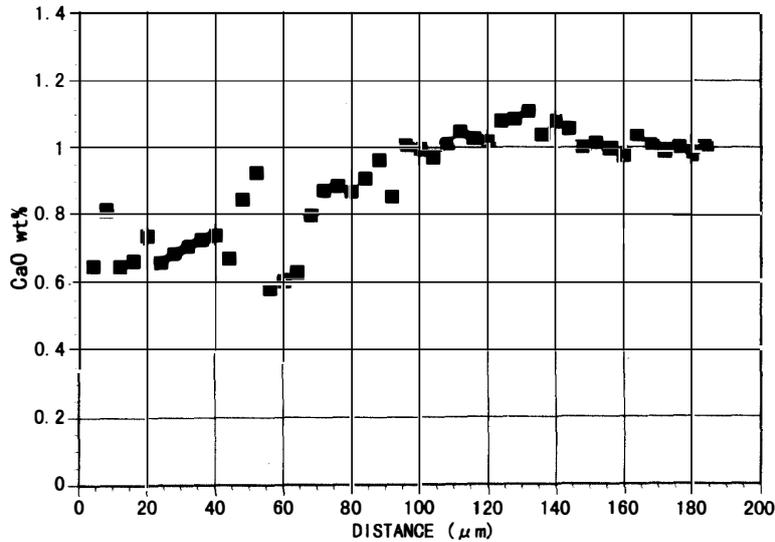
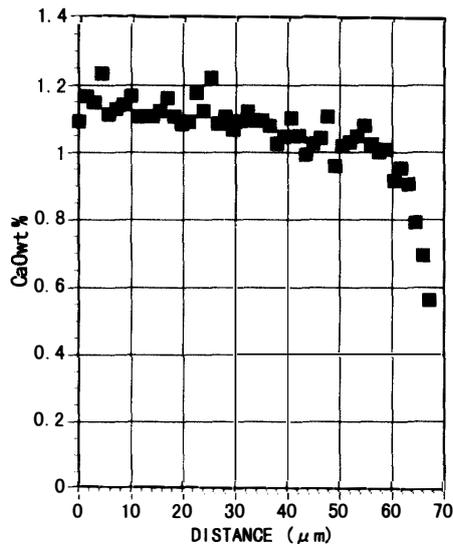
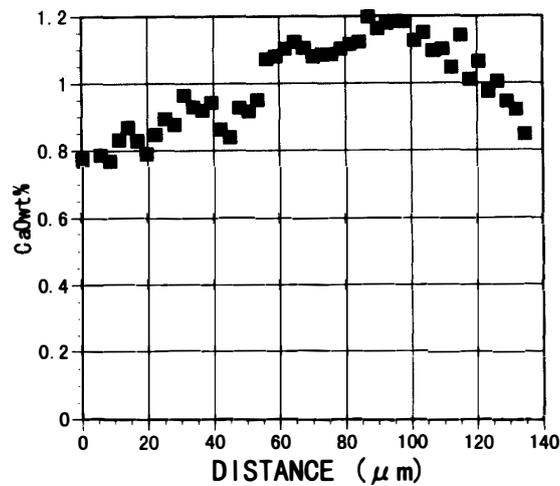
c. *Acapulco (with inclusions).*d. *Acapulco (without inclusions).*

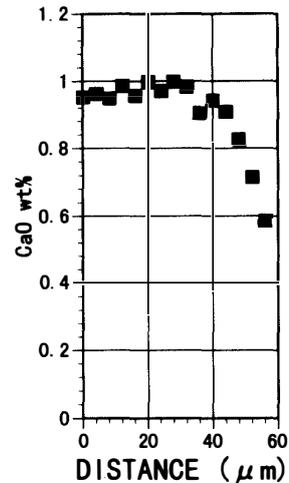
Fig. 3 (continued).

ALH-78230

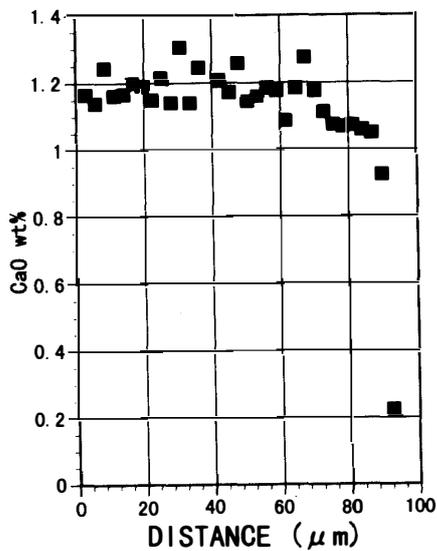
ALH-78230 has a fine-grained (up to about 0.25 mm) texture, mineral abundances and chemical compositions of minerals similar to those of ALH-77081. The PTS size is about 9 mm × 5 mm. A mineral distribution map of ALH-78230 (Fig. 2d) shows the typical recrystallized texture of acapulcoites. The metal grain sizes (up to 0.4 mm) are larger than those of plagioclase grains, which are larger than those of the H7 plagioclase, and distribute more evenly than those of H7. The average $fe\#$ of orthopyroxene is 10.2 and that of olivine is 10.2. The orthopyroxene does not show $fe\#$ zoning but it shows CaO zoning like other typical acapulcoites and lodranites. The average A of the largest five opaque grains is 1.7. It plots at the acapulcoite region in Fig. 6 of YUGAMI *et al.* (1998).



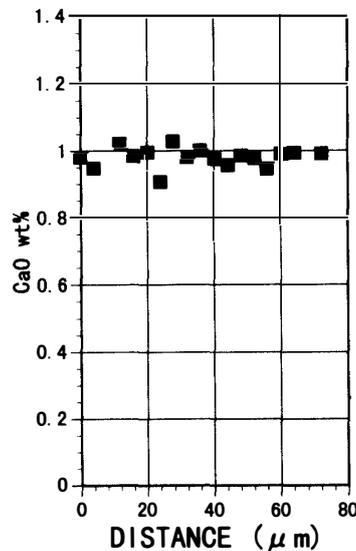
e. EET84302 (with inclusions).



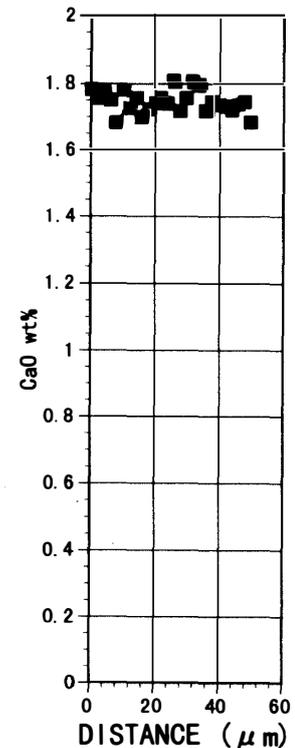
f. ALH-81261.



g. Tierra Blanca.



h. Winona.



i. ALH-81187.

Fig. 3 (continued).

ALH-81187

The recrystallized texture (Fig. 1b) which is typical of the primitive achondrites is disturbed at grain boundaries and minute dusty inclusions are present within these crystals. The silicate minerals have a medium grain size (0.05–0.6 mm across) and brownish small fragments that fill the space between them, and the silicate grains do not

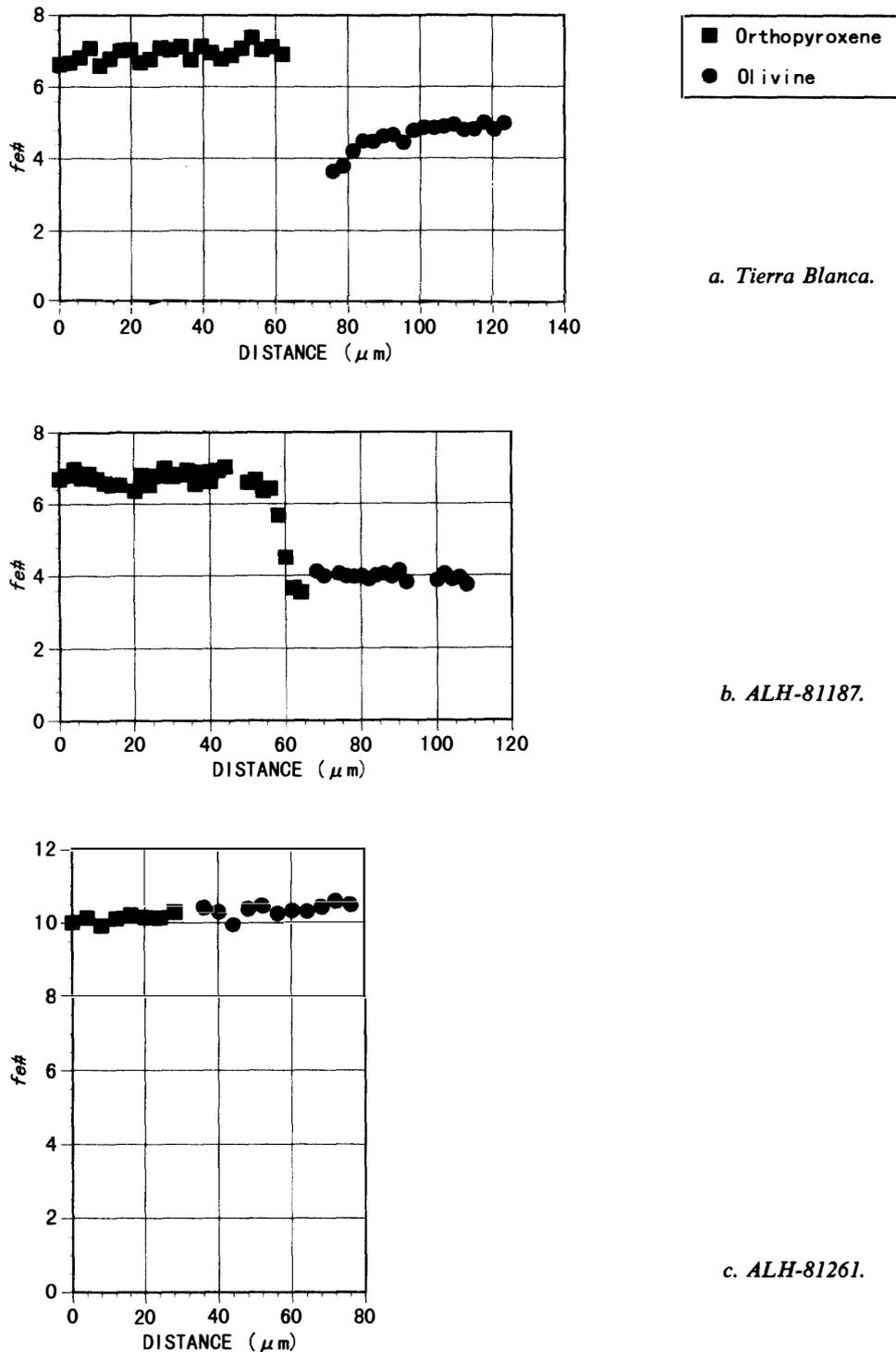


Fig. 4. Zoning profiles of the $fe\#$ from an orthopyroxene core to the adjacent olivine core of Tierra Blanca, ALH-81187, ALH-81261.

show curved grain boundaries common in acapulcoites such as ALH-77081. The modal abundances of minerals are approximately: olivine 21.0% orthopyroxene 51.5%, augite 2.0%, plagioclase 5.9%, FeNi-metal, troilite, etc. 19.6% (YUGAMI *et al.*, 1994).

Brown veinlets and small stained areas are limonite. The average $fe\#$ of orthopyroxene is 5.7 and that of olivine is 4.2, not much different from those of winonaites (KIMURA *et al.*, 1992). Orthopyroxene does not show CaO zoning (Fig. 3). The profile of $fe\#$ of olivine from core to rim of a crystal is flat, but $fe\#$ of some pyroxene grains varies from 6.4–6.9 (core) to 2.0–4.3 (rim) (the core to rim distance is about 0.035 mm) (Fig. 4).

ALH-81261

The texture of ALH-81261 (Fig. 1c) is similar to that of ALH-77081. The modal abundance of minerals is approximately: olivine 31.0%, orthopyroxene 36.2%, augite 5.3%, plagioclase 15.1%, FeNi-metal, troilite, etc. 11.8%, chromite, 0.5% and phosphate 0.3% (YUGAMI *et al.*, 1994). The average of $fe\#$ of pyroxene is 9.9 and that of olivine is 10.1. The silicate grains have complex, rounded boundaries. The opaque-silicate boundaries also show similar shapes. Some opaque grains consist of FeNi metal, troilite and chromite within the same curved boundaries but those between them are rather straight. The silicate grains have inclusions generally in regions near cores, and some fine dusty inclusions are aligned along surfaces that look like previously existed boundaries. However, the density of the overall inclusions is smaller than that of ALH-81187. The $fe\#$ s are smaller at the rims (10.2) than at the cores (10.7) in some olivine grains (Fig. 4). The analyses of chromite give an average $Fe/(Mg+Fe)$ atomic ratio = 0.63 and $Cr/(Al+Cr) = 0.62$, ZnO in chromite is 1.2 wt%.

4. Discussion

4.1. Grain sizes

The silicate grain sizes of lodranites/acapulcoites have been used as a measure to set a boundary between lodranites and acapulcoites by previous investigators (McCoy *et al.*, 1993, 1996). Those of "lodranites" with depletion of partial melt (about 300–500 μ m) are in general larger than those of recrystallized "acapulcoites" (about 100–200 μ m) without significant loss or enrichment of partial melts. Grain size distributions of minerals in acapulcoites (Fig. 2d) and winonaites (Fig. 2b, c) are similar and are not much different from recrystallized H chondrites (Fig. 2a). The grain size distribution of Y-75008 (H7) is even more heterogeneous than that of the primitive achondrites; plagioclase grains are finer than those of mafic silicates (Fig. 2a). The plagioclase grains are distributed presumably in a former matrix region before recrystallization. The coarse mafic silicates may have been inherited from chondrules. More or less even distribution of minerals in a large scale in the primitive achondrites implies that brecciation and mixing were extensive before recrystallization, of small amounts of partial melt.

The silicates in EET84302 are comparable to those in lodranites in grain size, but that meteorite contains a remarkably high amount (15.2 vol%) of large plagioclase grains with some augite and FeNi metal. The same coarse-grained plagioclase-rich areas with some augite have been found in silicate inclusions (SI) in Caddo County. These materials can be grouped as a hypothetical gabbroic material, which is a segregated partial melt. Because the trace element concentrations of EET84302 are as

high as those of Acapulco (FLOSS, 1998), it is difficult to classify this meteorite as a lodranite by our definition.

The distinction of winonaites and SI in IAB irons is not clear, and winonaites often include very large opaque mineral grains or veins such as seen in Y-74025 (Fig. 2b). The grain sizes of silicates of the Y-74025 winonaite are smaller than those of average winonaites, and it includes much smaller (0.02–0.2 mm) plagioclase and augite grains. A noteworthy texture of Y-74025 is that it also includes one extremely large opaque grain with complicated shapes, which were reported previously for Y-8005 (YUGAMI *et al.*, 1998). The observation of the grain sizes of many primitive achondrites, revealed that the grain sizes are not good indicators of classification of the sub-class.

4.2. Distribution of opaque minerals

The opaque grains of primitive achondrites are mainly FeNi-metal and troilite, and accessory chromite. The low-Fe (presumably reduced) members of primitive achondrites also include schreibersite and daubreelite, which are subsolidus products (KIMURA *et al.*, 1992). The FeNi-metal and troilite are proposed to be low-temperature-melting materials (TAKEDA *et al.*, 1994), on the basis of the presence of eutectic melt in the Fe-Ni-S system. Most opaque grains in primitive achondrites have shapes that fill the interstice of silicate grains (YUGAMI *et al.*, 1998).

Texture variations range from fine-grained recrystallized chondrite-like materials including all kinds of minerals known in chondrites, to coarse-grained materials with elongated, curved or joined opaque mineral grains (FeNi metal, troilite and rare chromite), often with large aggregates of FeNi metal (5.5 × 2 mm in size). Concentration or depletion of plagioclase-augite-rich materials and FeNi-metal and troilite characterizes the mineralogical heterogeneity of the coarser-grained samples.

The amounts of plagioclase and opaque minerals and the value of $A = L / (4\pi S)^{1/2}$ (average of the largest five opaque grains in each PTS) of six samples of the AL-group (Acapulco, ALH-77081, ALH-78230, Y-791491, MAC88177, and EET84302,28) are plotted in Fig. 5. EET84302 is much larger in the opaque grain size and in the total amount of opaque minerals and in A than the other five samples. Acapulco has larger grain size and larger A value and slightly smaller amounts of plagioclase than other acapulcoites. The silicate grains of Acapulco are also larger than those of other acapulcoites. The meteorites that are similar to Acapulco (*e.g.*, PALME *et al.*, 1981; ZIPFEL *et al.*, 1995) are called acapulcoites, but Acapulco is not a very typical acapulcoite in this sense.

Some opaque grains seem to align to a line, horseshoe shape or in circles that are the parts of a big network. Opaque grains have shapes that grew between silicate grains in these six samples. The opaque minerals were not totally melted, because materials formed from melt should show smooth shape. We assume the Fe-Ni-S eutectic melts were produced in minor amounts, but even the presence of minor melts will facilitate the migration of materials to grow larger grains to be larger filling available spaces, resulting in a complex shapes. We do not assume that the opaque minerals had been totally melted. If one larger grain grow bigger and large amounts of materials were supplied, the grains would be connected and large opaque grains with complex shapes like those in EET84302 may be formed. If the process goes one step further, an iron meteorite

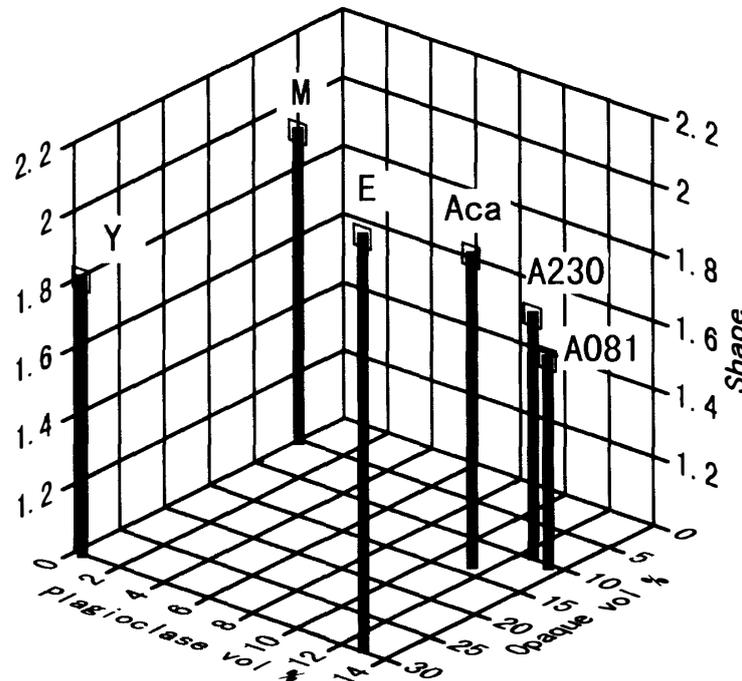


Fig. 5. Diagram of shape parameters $A = L/(4\pi S)^{1/2}$ of opaque grains versus volume % of opaque minerals and plagioclase. L is length and S is area of an opaque grain.

Shape = $A = L/(4\pi S)^{1/2}$. Aca = Acapulco. A081 = ALH-77081. A230 = ALH-78230.

Y = Y-791491. M = MAC88177. E = EET84302, 28.

with silicate inclusions can be formed on the parent body of the *AL* group. However, we are not relating *AL* group to any existing iron meteorite groups. MAC88177 has small amounts of opaque minerals but it has a large value of A . It may have experienced a migration of a large amount of materials. The complexity of the opaque grain shapes may have resulted from the minor melting and migration of low-temperature melting materials such as the Fe-Ni-S eutectic melt and Ca-Al-rich melt through grain boundaries.

The members of *WI*-group show more remarkable shapes of opaque minerals. YUGAMI *et al.* (1998) reported that Y-75305 includes large grains with an apparent shape that looks like parts of a ring. This texture can not be explained by gravitational separation in a molten magma, because it may form a sphere. They also emphasized that the largest opaque grain in Y-8005 looks like a miniature iron meteorite. The metal grains of Y-8005 have complicated shapes that resemble the opaque grains in EET 84302. These grains exist only on one side of an arc, and this texture is similar to that of Y-75305. Y-8005 is interesting because of its heterogeneity. Among the PTSs studied in this report, Y-74025 contains large metal and troilite veins forming a troilite-enriched region.

Since one or a few grains of the very large opaque minerals occupy the large percentages of the total area of the opaque minerals within each PTS of Y-75305, Y-74025 and Y-8005, the opaque minerals around the larger grains are small and less abundant. These meteorites have heterogeneity of distribution of opaque minerals in

the scale of one PTS. The very large opaque grains are not always present in all PTSs of these samples studied. There is heterogeneity among different PTSs of the same meteorite.

Because the segregation of metal in the chondritic materials is a basic process in formation of planetesimals, the formation process of the IAB irons in the winonaites-IAB parent body would be applied to deduce the formation of the *AL* group. The presence of metal-depleted lodranite implies the formation of metallic aggregate in other places. Although such metallic aggregate has not been represented in our meteorite collections, metal-rich part of EET84302 (TAKEDA *et al.*, 1994) can be interpreted as evidence of metal segregation. Now, a large mass of metal has been found in an acapulcoite (McCOY and CARLSON, 1998).

The driving force to migrate Fe-Ni-S-P eutectic melt in low gravity field has not been well understood. Large interface energies between solid silicates and the eutectic melt may be responsible for such migration, if we accept an idea proposed by a microgravity experiment for material transport (PREDEK *et al.*, 1987; IMAISHI *et al.*, 1980). Local temperature gradient by a shock event might promote further migration. Aggregation of the eutectic melt droplets will favor the lowering of the entire surface energy of the system. The value of $A = L/(4\pi S)^{1/2}$ of each grain seems to increase when the grain is not very large because elongated metal grains between silicate grains are connected. The value seems to decrease when the grains grow as large as the largest grain in Y-8005 to minimize the surface energy.

4.3. Chemical variations of minerals

The *fe*#s ($= 100 \times \text{Fe}/(\text{Mg} + \text{Fe})$ atomic ratio) of the mafic silicates of the *WI* group (*fe*# = about 2–7) overlap with those of the Mg-rich members (*fe*# = about 3–6) of the *AL* group. The presence of coarse-grained acapulcoites (some of former lodranites) does not support the old classification scheme of acapulcoite/lodranite. Fe/Mg reverse zoning at the rims of olivine can be attributed to reduction during cooling on the basis of the diffusion simulation of the cations (MIYAMOTO and TAKEDA, 1994). Primitive achondrites with larger *fe*#s (than 10) of silicates that include Ca-phosphate do not include schreibersite, and those with smaller *fe*#s (than 10) that include schreibersite do not include Ca-phosphate. Those with much smaller *fe*#s include daubreelite. Such trends can be explained by oxidation-reduction condition involving phosphate-phosphide conversion (IKEDA *et al.*, 1997; KIMURA *et al.*, 1992).

Winonaites generally have smaller *fe*# of mafic silicates than most acapulcoites (KIMURA *et al.*, 1992). Although these two groups are from different parent bodies, some records of common processes such as Mg/Fe zoning at the rims of mafic silicates were found. Tierra Blanca is a Fe-rich member of winonaites and ALH-81187 is a Mg-rich member of acapulcoites. The *fe*#s of mafic silicates of Tierra Blanca are larger than those of ALH-81187 (Fig. 4). The mafic silicates of both Tierra Blanca and ALH-81187 show reverse zoning of *fe*# at the rims of grains (Fig. 4). Typical acapulcoites do not show reverse zoning of *fe*# (e.g., ALH-81261, Fig. 4).

The chemical composition of Tierra Blanca orthopyroxene is almost constant, but the *fe*# in olivine shows reverse zoning (Fig. 4). The *fe*# of Tierra Blanca olivine decreases at the rim (from about 5 to about 3.5). These values are larger than some

other winonaites. The $fe\#$ of the ALH-81187 olivine is almost constant (about 4) and $fe\#$ of the ALH-81187 orthopyroxene decreases at the rim (from about 6.8 to about 4). The $fe\#$ of the rim of ALH-81187 orthopyroxene grain is similar to the $fe\#$ of the ALH-81187 olivine. The cooling rates of Tierra Blanca and ALH-81187 have been calculated by using the same method as that of MIYAMOTO and TAKEDA (1994). Tierra Blanca cooled faster than ALH-81187 (YUGAMI *et al.*, 1995).

Considering the fact that diffusion of Fe in olivine is faster than that of orthopyroxene, it is suggested that the difference between $fe\#$ s of orthopyroxene and olivine and the reverse zoning of mafic silicates in Tierra Blanca and ALH-81187 can be explained by the difference in reduction in the solid state.

The reduction of Tierra Blanca was terminated in an earlier stage of the reduction process than ALH-81187, because olivine is more likely to be reduced than pyroxene. Winonaites and acapulcoites were formed in different parent bodies, but it seems that they suffered from reduction processes. The difference of $fe\#$ s of most acapulcoites and winonaites may have originated during the initial crystal growth involving partial melting and reduction before the solid state reduction occurred. It indicates that there have been two kinds of reduction, one is reduction during the crystal growth and the other is reduction in the solid state at the final stage of cooling when the planetesimals were disrupted or broken up. It is possible to envision that winonaites with very small $fe\#$ s of the cores of mafic silicates are products of reduction in the solid state terminated at a later stage than ALH-81187. Acapulcoites and winonaites are not essentially different from each other.

ALH-81187 is similar to ALH-81261 in their grain size of silicates and FeNi metals that look like they are disseminated into the space between silicate grains. Although both meteorites are acapulcoites, ALH-81187 is richer in pyroxene and metal than ALH-81261, and $fe\#$ s of pyroxene and olivine of ALH-81187 are smaller than those of ALH-81261, especially those at the rims of pyroxene grains. Olivine is more likely to be reduced than pyroxene, because of the smaller diffusion coefficient of Mg than that of Fe, and olivine decomposes to pyroxene, metallic iron and silica by reduction if it is exposed to a low oxygen fugacity environment. The dusty inclusions in the central regions of grains of both meteorites are similar to those of EET84302 (TAKEDA *et al.*, 1994), which were partly but not completely melted. They suggested that the dusty cores might be a remnant of the precursor materials. It is also likely that the inclusions in the outer part of the grains of ALH-81187 in particular were produced by reduction in the solid state, if we correlate it with the disturbed grain boundaries produced by a shock event. The texture of ALH-81187 may indicate that the precursor material before shock may have been similar to ALH-81261.

The Y-74357 lodranite shows dusty inclusions in olivine, which is partly reduced but completely homogenized to give Mg-rich composition closer to winonaites. However, the pyroxene crystal shows reduction only at the outer region (about $30\mu\text{m}$). MIYAMOTO and TAKEDA (1994) attributed this phenomenon to the difference in diffusion of Mg/Fe between pyroxene and olivine and the reduction that took place when the parent body was exposed to vacuum with some reducing agent after being broken up and while the interior was still retained at high temperature. They reported that the cooling rate of $1.5^\circ\text{C}/\text{year}$ from 1000 to 600°C gives the best fit for the observed Fe-Mg profile

at the rim of orthopyroxene of Y-74357. Because of similar diffusion profiles observed in the ALH-81187 orthopyroxene to those of Y-74357 and partly reduced and homogenized olivine in both meteorites, we can envision similar thermal histories for these two meteorites. The ranges of reverse zoning at the rims of orthopyroxene grains of ALH-81187 are wider than those of Y-74357. It suggests that the cooling rate of ALH-81187 may be slightly slower than that of Y-74357. Then it may have been shocked and partly reduced while it was hot. The chemical compositions of ALH-81187 olivine show that it was apparently, partly reduced to the $fe\#$ as low as those of some winonaites, but the oxygen isotope ratios of ALH-81187 is within the range of acapulcoites. So it is provable that ALH-81187 was originally an acapulcoite like ALH-81261 and was partly reduced to the present state after shock.

It has already been suggested that the reflectance spectra of reduced and nickel-iron-rich primitive achondrites such as Y-74357 will give a better fit for those of the S asteroids (HIROI *et al.*, 1993). The S asteroids are known for what is called as "space weathering", which is not scientifically defined process. HIROI *et al.* (1993) and TAKEDA *et al.* (1994) suggested that the reduction process related to shock events such as observed for Y-74357 may have a relationship to space weathering. They may give spectra similar to those with space weathering, because ALH-81187 is much fine-grained and their grain boundaries are filled with opaque materials.

The reduced members of lodranites (e.g., EET84302 and Y-74357) contain Ni as Ni-rich schreibersite as in the IAB iron meteorites, whereas MAC88177 is richer in troilite than metal and contains Ni as taenite. The fact implies that the statement made earlier by KIMURA *et al.* (1992) on the classification criteria based on the $fe\#$ of the mafic silicates cannot be used as a criterion to classify the *WI* and *AL* groups.

The gradual decrease of CaO contents towards the rims of orthopyroxene is one of the characteristics of primitive achondrites. However, the zoning profiles of CaO differ from meteorite to meteorite (Fig. 3). Many lodranites (e.g., Y-791491) have higher concentrations of CaO in the core. Some lodranite orthopyroxenes with an oriented dusty core, where the CaO content is as low as chondritic orthopyroxene, show the increases of CaO to a level of the ordinary lodranites around the core and slight decrease at the rims. The preservation of CaO zoning in the orthopyroxene crystals of primitive achondrites suggests that the growth speed of these orthopyroxenes was not as slow as that in diogenites (cumulates of the HED parent body) in spite of the coarse-grained textures and uniform Mg/Fe compositions. Because diffusion of Mg and Fe is faster than that of Ca, Mg/Fe homogenization most likely has taken place even during a high temperature annealing episode in the solid state, as well as in magmatic processes.

Another implication of Ca zoning in the orthopyroxene to the formation process of lodranites is that removal of the plagioclase-rich melt was not equilibrium partial melting process and that growth of orthopyroxene was coupled with the removal of Ca in the melt by growth of other Ca-rich phases, such as augite and Ca-Na-rich plagioclase. If this zoning was produced during the crystal growth of orthopyroxene, it indicates that the CaO contents of melt decreased in the course of crystal growth. The extraction of Ca-rich partial melts in one place, which results in concentration of Ca-rich melt in other places where other Ca-rich phases were crystallizing, can account for the decrease of CaO at the rims. Significant variation of Ca content from one crystal to

another cannot be explained by an equilibrium partial melting model. A driving force for the melt accumulation is not well understood. We accept a model proposed for the IAB irons (TAKEDA *et al.*, 1997). The coexistence of FeNi metal and the plagioclase-augite-rich area in Caddo County suggested that the Fe-Ni-S eutectic melt moved side by side with the Ca-Al-rich liquid, although they have quite different densities. The low gravity field of the asteroids can account for this difficulty and ignore this difference. The distribution of plagioclase in Caddo County also suggests that the Ca-rich liquid accumulated and migrated, and crystallized as plagioclase-augite-rich materials.

Acapulco and EET84302 have similar dusty cores in the cores of orthopyroxene crystals. In the dusty cores, there are minute particles of FeNi metal and troilite, and the CaO contents of such cores is low (CaO=about 0.6 wt% (Acapulco), CaO=about 0.8 wt% (EET84302)). If this dusty core is a relict orthopyroxene of the precursor material, Ca-rich rim can be overgrown by materials from the Ca-rich partial melt.

4.4. *Implication for formation mechanism of primitive achondrites*

On the basis of all available data of *AL* and *WI* meteorites from Antarctica and elsewhere, it is pointed out that heterogeneous distribution of materials is an important aspect as discussed above. A hypothetical map of the distribution of opaque minerals and plagioclase on a larger scale was made to show the idea that local heterogeneity of materials in the *AL* parent body can explain the variability of abundance of minerals found in the meteorite samples (YUGAMI *et al.*, 1996a). The fine-grained region is acapulcoite such as ALH-78230 (Fig. 2d) and the coarse grained region depleted in plagioclase and augite is lodranite. An iron-meteorite-like region, where the Fe-Ni-S eutectic melt is concentrated, has been found in the mineral distribution map of Y-74025 (Fig. 2b). A region rich in plagioclase and augite has been found in Caddo County (TAKEDA *et al.*, 1997). Now, we have to examine what kind of heat source can induce such heterogeneous distribution.

MIYAMOTO and TAKEDA (1992) examined the internal heating model of the primitive achondrites by decay of ^{26}Al and showed that the interior of the body will be partly melted for the Al contents of Acapulco. Although there is no evidence for ^{26}Al , we can assume the presence of ^{26}Al for the chondritic source materials that were formed in the earliest solar system (BENEDIX *et al.*, 1998). Partial melting models of the *AL* group assume this internal heat source (*e.g.*, MCCOY *et al.*, 1993, 1996). However, the temperature-time variation requires slow heating and slow cooling for the ^{26}Al heat source, and the heterogeneous distribution of materials proposed in this paper, may not be produced by certain processes. For example, the partial melting model combined with the explosive volcanism model assumes the loss of the partial melts into space (MCCOY *et al.*, 1993, 1997b). This assumption is not consistent with our finding of the partial-melt-rich areas in EET84302 and Caddo County B3A (TAKEDA *et al.*, 1994, 1997).

The heterogeneous distribution of the materials prefers heterogeneous heat source as that produced extensively recrystallized chondrites by heterogeneous shock melting in part (TAKEDA *et al.*, 1984). However, it should be noted that no evidence of shock records has been preserved in this LL7 chondrite and H7 chondrite studied in this report. If once the melting takes place, the shock records will be erased by subsequent

crystal growth and thermal annealing. The presence of dusty cores in the Acapulco and EET84032 orthopyroxene crystals are similar to orthopyroxenes of partly melted, shock-recrystallized diogenites (e.g., Y-74013, TAKEDA *et al.*, 1981). In these diogenites, evidence of heterogeneous melting and crystal growth by shock effects has been documented. The driving force of partial melt to other places has not been too well known, but it requires heterogeneity in temperature distribution. The shock event may be partly responsible for such heterogeneous heating, but we do not propose that it is the only heat source. The ^{26}Al heating is also responsible for high temperature episode. The present melt should be called "melt formed by heterogeneous melting that has different compositions from place to place".

The uneven distribution of minerals in the H7 chondrite and Tierra Blanca may be explained by extensive brecciation, mixing and melting, when the recrystallization took place. Inhomogeneous distribution of melted portion indicates migration and segregation of the partial melts. However, this interpretation does not rule out the coexistence of fine-recrystallized portion and metal as was found in Caddo County (TAKEDA *et al.*, 1997).

As TAKEDA (1993) and TAKEDA *et al.* (1994) pointed out, a type of shock melting when planetesimals grew by slow-speed collisions in the early solar system, combined with the heating by decay of ^{26}Al , may be different from those produced by a small scale impact of a high speed projectile on the surface of a large planet. We are discussing a basic process for planetesimals, and not referring to the primitive achondrites in particular. The high-speed impacts produce shock veins as were found in chondrites, eucrites and some lunar samples. These examples of melting are not what we expect to see for the large-scale planetesimal collisions. Especially, when a body consisting of hard components and soft porous matrices is collided, the behavior may be quite different from the high-speed impact on a small body. The porous matrix material is preferentially melted (N. HIRATA, personal comm., 1998). However, further theoretical and experimental studies are required before we apply such model to formation of the primitive achondrites.

Evidence of migration of materials through grain boundaries and the presence of dusty cores as in the shock recrystallized chondrites and diogenites (TAKEDA *et al.*, 1981; TAKEDA, 1993) can be explained more easily by a formation model of primitive achondrites by heating due to non-equilibrium processes in addition to the ^{26}Al heating, etc.

The model of generating lodranite-like mafic-silicate-rich materials from chondrite-like source materials by partial melting due to decay of ^{26}Al and removal of Ca-Al-rich melt and Fe-Ni-S eutectic melts during crystal growth (TAKEDA *et al.*, 1994) expects that such materials rich in plagioclase (Ca-Al rich) and metal-troilite should be concentrated elsewhere. The discovery of plagioclase-augite-rich materials near by metal-rich meteorites as was found in Caddo County (TAKEDA *et al.*, 1997; YUGAMI *et al.*, 1998) gives strong evidence for the proposed model. Discovery of large grains of metal in some winonaites (Fig. 2b) also supports the above model. The model proposed in this work for the IAB irons will be useful for interpretation of the exotic clasts rich in alkali-feldspar and Cr-diopside found in the IIE irons (WASSERBURG *et al.*, 1968).

If the explosive volcanism is a dominant process, plagioclase-augite-rich materials

will be brought to surface and lost in space. The fact that plagioclase-augite-rich portions exist in Caddo County together with FeNi metal and the gradual difference of abundances of plagioclase and augite of the *AL* group, as was found in this study, suggests that the explosive volcanism is not a dominant process. The records kept in meteorites suggest that the local heterogeneous segregation of materials induced by difference in grain boundary energy and by collisional processes is much easier to occur than explosive volcanism. The local heterogeneity of materials observed in this paper may be the very first step of differentiation of chondritic materials to achondritic materials.

The last history of the primitive achondrite parent body can be traced by Mg-Fe variations of the mafic silicates described in this paper. The results indicate that there have been two kinds of reduction, one is reduction during the recrystallization and the other is reduction in the solid state at the final stage of cooling, when the planetesimal was disrupted or broken up. The difference of *fe#*s of most acapulcoites and winonaites may have been formed during the recrystallization involving reduction before the solid state reduction occurred. The chemical zoning at the rim of olivine can be attributed to the reduction at the final stage. If a fracture developed towards the interior of the parent body, while the interior is still hot, the vacuum-like environment will be introduced in the interior and the deduction of the rims of mafic silicates will take place with the presence of reducing agents or loss of S.

When the extensive fracturing and shock effects are recorded in the olivine crystals, minute metallic particles are produced at dislocations and defect sites, producing dusty appearance such as observed in ALH-81187. This kind of darkening may be responsible for proposed space weathering of the S-asteroids (HIROI *et al.*, 1993).

5. Conclusions

(1) Mineral distribution maps produced by EPMA and image processing show that fine-grained acapulcoites and winonaites, except for Y-74025, have fairly even distribution of minerals, indicating major recrystallization without extensive segregation.

(2) Coarse-grained varieties of primitive achondrites, reported previously, show extensive segregation of materials, presumably crystallized from the partial melts.

(3) Variations of CaO zoning in orthopyroxene suggest that melting of Ca-rich partial melt and their crystallization took place unevenly.

(4) Zonings of *fe#* at the rims of olivine in primitive achondrites suggest that reduction took place at the very last history of the thermal evolution of the parent bodies.

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