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REFRACTORY INCLUSIONS IN THE ALLENDE METEORITE

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INTRODUCTION

The fall of the Allende Type 3 carbonaceous chondrite in Chihuahua, Mexico, on February 8, 1969 (King et al 1969, Clarke et al 1970) marked the beginning of a decade of remarkable discoveries which caused an unprecedented, explosive increase in our knowledge of physical and chemical processes that occurred at the birth of the solar system. Prior to its fall, studies of carbonaceous chondrites were severely limited by the rarity of these types of meteorite, their small sizes, and the relatively small sample sizes of their chips and powders obtainable from museum curators who were cognizant of the scientific value of these meteorites and their short supply. At a time when many of the world's geochemical laboratories were in close communication with one another and were perfecting their analytical techniques to derive the maximum information from a minimum of sample in preparation for the return of the first lunar rocks only six months thence, there were suddenly several tons of a single carbonaceous chondrite available for study. Large amounts of material were collected within a few days of the fall, before terrestrial weathering could cause significant degradation of it (Levy et al 1970), and were distributed widely within the scientific community shortly thereafter (Rancitelli et al 1969). So much material was then available that Morgan et al (1969) proposed that a large amount of homogenized powder be prepared and disseminated as a sorely needed standard for interlaboratory comparison of meteorite analyses. This suggestion was promptly taken up by the Smithsonian Institution (Mason 1975) and it is hoped that the compilation of papers (Jarosewich & Clarke 1980) presenting analytical results on 4 kg of such material will soon be published.

Individual stones weighing several kg each were cut into thin slices

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(Clarke et al 1970, Grossman & Ganapathy 1975), thereby exposing to view for the first time hundreds of cm^2 of slab surfaces of a carbonaceous chondrite. Such a view is shown in Figure 1, from which Allende is seen to consist of a heterogeneous mixture of different types of inclusion, distinguishable from one another here by their different sizes, shapes, and colors, embedded in a fine-grained, black matrix. Studies of individual inclusions demonstrate that these different inclusion types have different mineralogical and chemical compositions and, therefore, their own independent histories. Except for its size, Allende does not appear to be unusual among other Type 3 carbonaceous chondrites. The heterogeneity of these meteorites and the fact that each inclusion type has its own origin are important reasons why many earlier studies of carbonaceous chondrites based on analyses of small, bulk samples had such a relatively low information return. In the case of Allende, major advances were made because the large amounts of material available permitted access to and sampling and

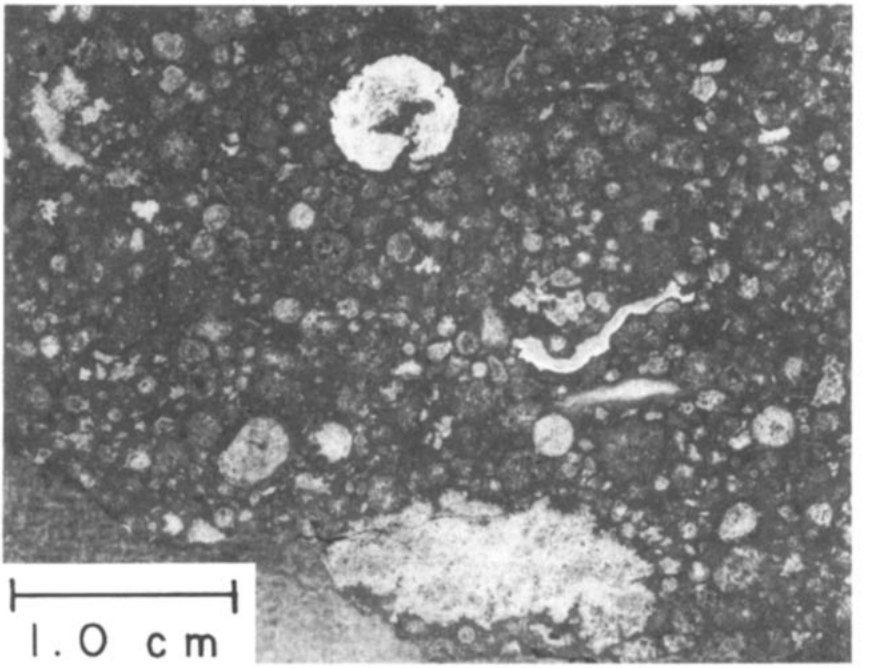

Figure 1 Slab surface of the Allende meteorite. Light-colored object at top center is one of the coarse-grained inclusions which are the subject of this paper. The two narrow, elongated objects at middle right are fine-grained inclusions (Grossman et al 1975, Grossman & Ganapathy 1976a) and the large object at bottom center is an amoeboid olivine aggregate (Grossman & Steele 1976, Grossman et al 1979a). A dark clast (Fruland et al 1978) is barely visible at middle left. Most of the other light-colored objects are chondrules.

Table 1 Chemical formulae of minerals mentioned in the text

Type of compound	Mineral	Chemical formula
Oxides	Corundum	Al_2O_3
	Hibonite	$CaO \cdot 6Al_2O_3$
	Perovskite	$CaTiO_3$
	Spinel	$MgAl_2O_4$
	Thorianite	ThO_2
	Baddeleyite	ZrO_2
	Magnetite	Fe_3O_4
Silicates	Wollastonite	$CaSiO_3$
	Rhönite	$Ca_4(Mg, Al, Ti)_{12}(Si, Al)_{12}O_{40}$
Melilite Series	Gehlenite (Ge)	$Ca_2Al_2SiO_7$
	Åkermanite (Åk)	$Ca_2MgSi_2O_7$
Plagioclase Series	Anorthite (An)	$CaAl_2Si_2O_8$.
	Albite (Ab)	$NaAlSi_3O_8$
Clinopyroxenes	Diopside	$CaMgSi_2O_6$
	Hedenbergite	$CaFeSi_2O_6$
	Fassaite	$Ca(Mg, Al, Ti)(Si, Al)_2O_6$
Olivine Series	Forsterite	Mg_2SiO_4
	Fayalite	Fe_2SiO_4
Garnets	Grossular	$Ca_3Al_2Si_3O_{12}$
	Andradite	$Ca_3Fe_2Si_3O_{12}$
Feldspathoids	Nepheline	$NaAlSiO_4$
	Sodalite	$3NaAlSiO_4 \cdot NaCl$

analysis of individual, mm- to cm-sized inclusions. Often, individual inclusions were studied by a combination of different techniques in one or more laboratories. This approach required new techniques for handling and analysing samples in the μg to mg size range, many developed in the lunar sample program, because of the number of different experiments being performed on splits of small amounts of materials.

This paper reviews the literature on only those objects in Allende commonly referred to as "coarse-grained inclusions." Although these inclusions comprise little more than 5 per cent of the mass of Allende, they have been the subject of more than 85 per cent of the papers published about this meteorite. A brief review of some of the other types of Allende inclusion is given by Wark (1979). Table 1 lists the chemical formulae of minerals discussed in this review.

PETROGRAPHY AND MAJOR MINERALS

The coarse-grained inclusions are usually the largest, most prominent, white inclusions seen on slab surfaces (Figure 1). They range from <1 mm to 2 cm in size and usually contain crystals at least 0.5 mm and often up

to 2–3 mm in longest dimension. They can be completely spherical, a fact which led some workers, e.g. Clarke et al (1970), Gray et al (1973) and Martin & Mason (1974), to call them chondrules, but are often highly irregular and convoluted in shape (Figure 2). Some are fragments of once-larger inclusions. Some have natural cavities containing wollastonite needles (Fuchs 1971, Grossman 1975a, Allen et al 1978). Others are doughnut-shaped, their central holes filled with minerals typical of the matrix of Allende. Some early work (Arrhenius & Alfvén 1971, Grossman 1973, Seitz & Kushiro 1974) suffered from failure to distinguish these inclusions from other Ca-rich bodies in this meteorite (Grossman & Ganapathy 1975) and more recent work (R. N. Clayton & Mayeda 1977, Wasserburg et al 1977, Lorin et al 1978, MacPherson & Grossman 1979) indicates that important information has been overlooked in considering all members of even this restricted group of inclusions to have a common origin.

In a petrographic and electron microprobe survey of a large number of coarse-grained inclusions, Grossman (1974, 1975a) found that there were basically two kinds: those with minor ($<5\%$) clinopyroxene and those with major ($>35\%$) clinopyroxene. He pointed out that pyroxene in the first kind, Type A, is relatively pure diopside ($Al_2O_3 < 9\%$, $Ti < 0.7\%$) and is restricted to thin ($<50\ \mu m$) rims around the inclusions, while that in the second kind, Type B, occurs as coarse crystals in inclusion interiors and contains 15–22% Al_2O_3 and 1.8–10.8% Ti. Clarke et al (1970) were the first to call these Ti-, Al-rich clinopyroxenes "fassaite." One inclusion, called "intermediate" by Grossman (1975a), has major amounts of pyroxene intermediate in composition between these two (9–18% Al_2O_3, 0.9–2.4% Ti). Later work showed, however, that fassaite is also present in Type A inclusions, both as a minor phase in their interiors (Allen et al 1978) and in their rims (Wark & Lovering 1977a). The latter workers also found diopside in rims on Type B inclusions. Because rims are clearly distinguishable petrographically from inclusion interiors, account for such a small volume fraction of each inclusion, and probably have little genetic relation to the interiors, it now seems unwise to use compositions of rim phases as part of a classification scheme. Thus, ignoring rim phases, all primary pyroxenes in the interiors of Type A, Type B, and intermediate inclusions contain $>9\%$ Al_2O_3 and $>0.9\%$ Ti and are all classifiable as fassaites (Deer et al 1978). The major difference between Type A and Type B inclusions is still the abundance of pyroxene and, if this is the sole criterion for distinguishing inclusions, the intermediate one would now belong to Type B. Because fassaite is the only dark-colored, coarsely crystalline phase in the inclusions, the presence of abundant dark material is often sufficient to distinguish a Type B from a Type A on a slab surface.

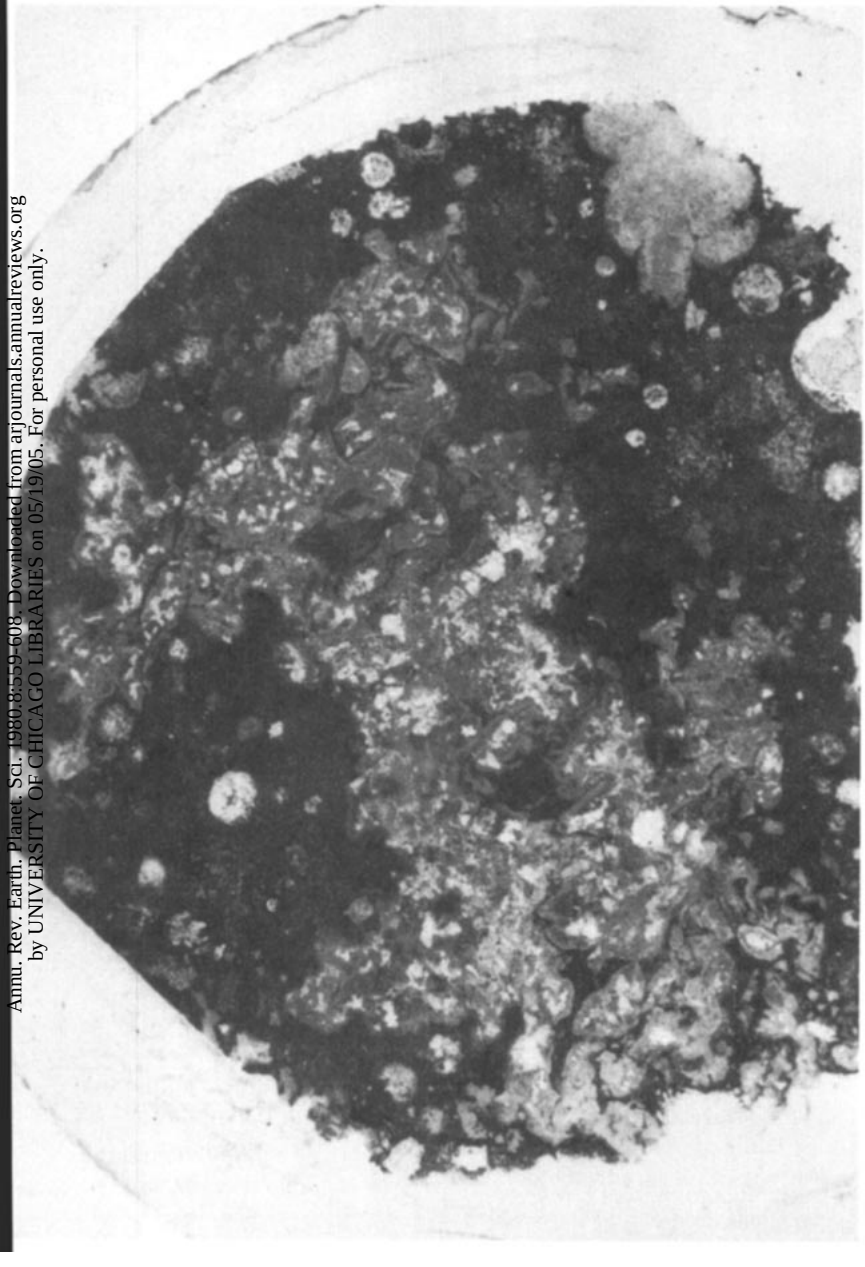

Figure 2 Fluffy Type A inclusion, TS24F1 of Grossman (1975a), consisting of melilite (white), hibonite and spinel (not visible), and fine-grained alteration products (grey). In places, highly contorted external surface of inclusion is marked by a light-colored rim, which separates the interior of the inclusion from the matrix of Allende (black). Transmitted light; width of field 2.5 cm.

Type A Inclusions

MacPherson & Grossman (1979) pointed out that two subtypes exist. Those in the most common subtype are extremely contorted in shape, are very heavily altered and contain abundant cavities (Figure 2). These we will call "fluffy." The others are more nearly spherical, less heavily altered and are more compact. Of the primary phases in both subtypes, melilite is by far the most abundant, $>75\%$ by volume. In fluffy Type A's, melilite ranges in composition from $Åk_0$ to $Åk_{33}$, but in compact ones, from $Åk_8$ to $Åk_{80}$. (As seen in Table 1, melilite refers to a solid solution series between pure gehlenite and pure åkermanite. The notation $Åk_x$ indicates a phase intermediate between these two end-members which is composed of x mole % åkermanite and $[100-x]$ mole % gehlenite.) Spinel, nearly pure $MgAl_2O_4$ in composition (Grossman 1975a), is usually the next most common phase, 5–20% by volume of the primary phase assemblage. Fluffy Type A's sometimes contain rare, orange-red, V-rich spinels (up to 5% V_2O_3, 20% FeO). Hibonite (Keil & Fuchs 1971, Haggerty & Merkel 1976, Allen et al 1978) is usually the next most abundant phase, $\sim 5\%$, followed by perovskite, ~ 1–3%. Occasionally, rhönite occurs as a minor phase in Type A's (Fuchs 1971, El Goresy et al 1977b).

From published petrographic descriptions, inclusions A and B of Fuchs (1971), 6/16 of Blander & Fuchs (1975), TS27F1 of Grossman (1975a), the inclusion in Figure 5 of Kurat (1975), and CG-11 of Allen et al (1978) are fluffy Type A's. Textural relations among primary phases are often obscured or obliterated by veins and cavities containing secondary alteration products. Where visible, most of the primary assemblage consists of interlocking masses of blocky or lath-shaped melilite crystals which can be up to 200 μm in longest dimension and often contain deformation lamellae. A wide range of objects are poikilitically enclosed by melilite (i.e. totally enclosed within single melilite crystals): rounded prisms of fassaite ($\leqq 30$ μm), spinel octahedra ($\leqq 10$ μm), rounded, elongated hibonite grains ($\leqq 10$ μm), rhönite, euhedral perovskite crystals ($\leqq 5$ μm), rounded fassaite grains ($\leqq 30$ μm) enclosing perovskite, and spinel containing and/or rimmed by perovskite ($\leqq 5$ μm). Euhedral fassaite ($\leqq 110$ μm) and ragged, elongated hibonite grains are sometimes found between melilites. Euhedral hibonite crystals, up to 250 μm long, are sometimes intergrown with and embayed by melilite and contain perovskite ($\leqq 10$ μm). A peculiar feature of CG-11 is the reversed zoning of its melilites whose compositions vary from $Åk_{17}$ in cores to $Åk_3$ in rims (Allen et al 1978). In TS27F1, a severely deformed, 500 μm melilite grain is contiguous to a polycrystalline mosaic of interlocking melilite grains (10–30 μm) and ragged spinels, suggesting shock-induced recrystallization.

Allen et al (1978) studied grain-to-grain differences in trace and minor element contents of spinels and perovskites in a fluffy Type A and found that concentration variations of a factor of six were common within each of these phases.

Grossman (1975a) described and illustrated a compact Type A, TS2F1. This type of inclusion is usually composed almost entirely of several 2–3 mm blocky crystals of melilite, poikilitically enclosing spinel, perovskite, fassaite, and rhönite. Spinel crystals are euhedral, commonly reach more than 50 μm in size, and may form chains up to 50 crystals long. Spinel, 1–6 μm perovskite cubes, purple anhedral perovskites ($\sim$70 μm), and 1–10 μm prisms of fassaite occur as isolated grains within melilite. The long axes of the fassaite prisms are aligned in three directions within single melilite crystals and one of these directions is parallel to a set of edges on the perovskite cubes. Also within melilite, perovskite grains ($\leq$5 μm) are found at spinel-melilite contacts and wedged between spinel crystals and intergrowths of vermiform perovskite with fassaite occur, sometimes with rhönite (El Goresy et al 1977b) and/or spinel. The only mode of occurrence of hibonite in compact Type A's is as acicular crystals within 100 μm of their innermost rim layer.

The literature contains much more petrographic, as well as chemical and isotopic, information about Type B inclusions than Type A's, a fact that may lead the casual reader to the conclusion that Type B's are much more abundant. Grossman (1975a) pointed out, however, that a significant fraction of the Type A's are only a few tenths of a mm in size, are perhaps fragments of once larger inclusions, and are thus not particularly prominent on slab surfaces. Therefore, fewer thin sections are made of these inclusions and, due to their relatively small sizes, they are discriminated against in chemical and isotopic studies. The two inclusion types are probably comparable in abundance, though not in mass.

Type B Inclusions

Petrographic details and illustrations of Type B inclusions are found in Clarke et al (1970), Blander & Fuchs (1975) (inclusions 6/1 and 10/2), Kurat et al (1975) (an inclusion in Bali), R. N. Clayton et al (1977) (sample A13S4), Fuchs (1978), and El Goresy et al (1979b). Grossman (1975a) surveyed the petrography and mineral chemistry of a large number of Type B's, two of which, among others, were further studied by petrographic and electron and ion microprobe techniques by Hutcheon et al (1978a, b), Steele et al (1978), and Steele & Hutcheon (1979).

Figure 3 shows a typical Type B inclusion. Such inclusions contain 35–60% fassaite, 15–30% spinel, 5–25% plagioclase, and 5–20% melilite. Spinel, nearly pure $MgAl_2O_4$, occurs as grains from <1 μm to >100 μm

in size, poikilitically enclosed by 0.1–5 mm long laths of plagioclase, An_{100}, and melilite, $Åk_{15-80}$, and by anhedral, sub-equant fassaite grains up to 3–4 mm in size. Grains of fassaite, anorthite, and shocked melilite fit snugly together, with fassaite grains often filling angular interstices between anorthite and melilite laths. Anorthite laths sometimes pierce fassaite grains. Occasionally, corrosion textures are observed which suggest that melilite has replaced anorthite and fassaite in some cases and that melilite and fassaite have replaced anorthite in others. Characteristic of many Type B's is a thick outer mantle of melilite with minor spinel which surrounds a core of fassaite, anorthite, and more Åk-rich melilite (Kurat et al 1975, R. N. Clayton et al 1977). The presence or absence of such a mantle is the basis of Wark & Lovering's (1977a) subdivision of Type B inclusions into B1's and B2's, respectively. In some B1's the outer reaches of this mantle have a zone of 1–25 μm long hibonite needles which project inward toward the center of the inclusion (Blander & Fuchs 1975,

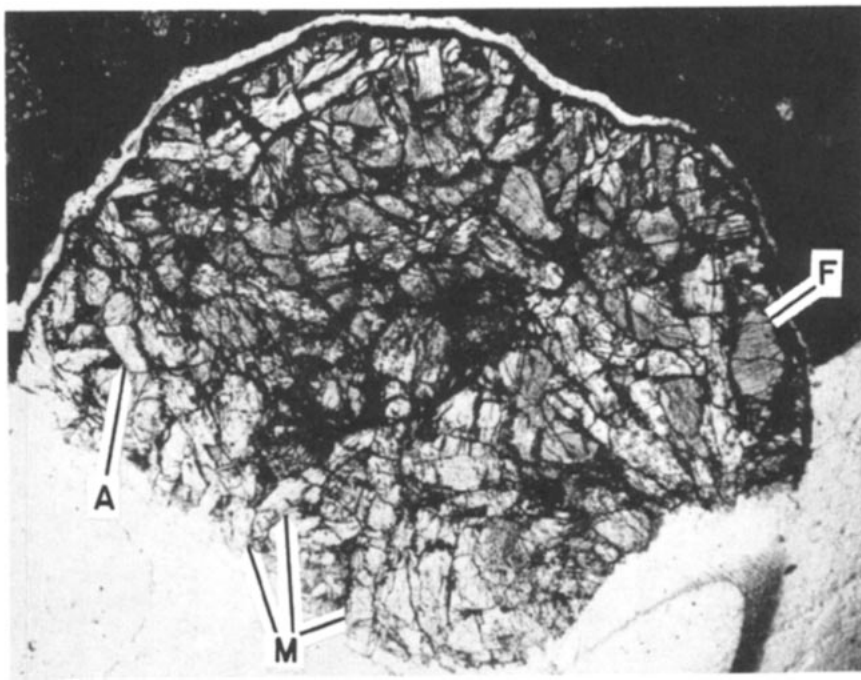

Figure 3 Portion of Type B inclusion, containing long, narrow melilite crystals (white, *M*), anhedral, sub-equant fassaites (grey, *F*), sparse, stubby laths of anorthite (white, *A*), and spinel as inclusions in these phases, especially fassaite. Thin white rim is epoxy where the inclusion has separated from the dark Allende matrix during preparation. Transmitted light; inclusion is 1.1 cm in diameter.

Wark & Lovering 1978a). MacPherson & Grossman (1979) noted that melilite laths in these mantles become progressively more Åk-rich along their lengths toward the center of the inclusion and that stubby, radially oriented laths appear to have had their growth interfered with by longer, neighboring laths growing at angles to them. Grossman (1975a) described fassaite crystals containing very dense concentrations of spinel at one end and virtually none at the other. El Goresy et al (1979b) described spinel "framboids" inside pyroxene and melilite crystals. These are spheroidal shells, up to several hundred μm in diameter, of spinel crystals which enclose fassaite, melilite, anorthite, and spinel. Wark et al (1979) also noted these, but considered "ribbons," continuous chains of spinel on Types B1 and B2 inclusion rims, to be larger versions of them and "palisades," long, curving chains of linked individual spinel crystals in Types A and B2 interiors, to be segments of larger ones.

Compared to those in fluffy Type A inclusions, grain-to-grain variations in trace and minor element contents of spinels within the same Type B inclusion are small, only about a factor of two (Steele & Hutcheon 1979), although huge variations in Na and Mg contents of anorthites were reported in another Type B by Hutcheon et al (1978b).

Other Types

Although use of the term "intermediate" to describe inclusions with fassaites of particular compositions was found superfluous above, that for which Grossman (1975a) coined the term differs from Types A and B also in having an anorthite-rich (50%), melilite-free mode. Wark & Lovering (1978a) also use the term "intermediate" or Type I for several other inclusions that contain $\sim$50% anorthite, but which are melilite-bearing. The latter differ also from Types A and B by having "chilled" textures, a different outer rim sequence, and different trace phases.

Other melilite-free inclusions from Allende and Leoville were described by Lorin et al (1978). They consist of ophitic fassaite-anorthite intergrowths, contain spinel framboids, but have no rims. They may be related to Grossman's (1975a) "intermediate" inclusion, a re-examination of which in this laboratory revealed an ophitic texture and spinel framboids.

Blander & Fuchs (1975) illustrated an inclusion, their sample 9/16, in which euhedral forsterite and spinel crystals, some in framboids, are poikilitically enclosed by fassaite and in which melilite is $Åk_{67-88}$. It is similar to one studied by Dominik et al (1978), except that in the latter, spinels are either coated by perovskite and enclosed by forsterite or fill interstices between forsterites. Furthermore, this entire assemblage has a thick mantle of fassaite which encloses spinel ribbons, melilite ($Åk_{88}$), and anorthite. Forsterite contains 1.2–1.8% CaO. It is very similar to inclusion

A16S3 (R. N. Clayton et al 1977). Another olivine-bearing inclusion (Haggerty 1978a) differs from these in containing no melilite.

Valence of Titanium

The presence of Ti^{3+} in Allende fassaites is indicated by their dark color, pleochroism, and features in their polarized absorption spectra (Dowty & Clark 1973a,b, Burns & Huggins 1973), as Ti is the only transition metal present in more than trace amounts. Dowty & Clark (1973a) also found that about 70% of the Ti in their pyroxene had to be calculated as Ti^{3+} to obtain a chemical formula with 4.00 cations per six oxygen atoms. Mason (1974) and Grossman (1975a) further noted that the cation deficit calculated assuming that all Ti is Ti^{4+} increases with Ti content of the fassaite. Rhönite was first discovered by Fuchs (1971) as a minor phase in the inclusions and was later found to contain large amounts of Ti^{3+} (Haggerty 1976, 1977, Fuchs 1978).

Haggerty (1976, 1977, 1978b) proposed the use of Ti^{3+}/Ti^{4+} ratios in phases in the inclusions as indicators of condensation temperatures, based largely on his measurements of such ratios in phases exhibiting critical textural relationships with one another which imply a crystallization sequence. The validity of this cosmothermometer is in doubt, however, as one critical texture is the commonly observed symplectic intergrowth of spinel, perovskite, fassaite, melilite, and hibonite, which Haggerty interprets as decomposition products of rhönite, an inference disputed by El Goresy et al (1977b) and by Fuchs (1978). Furthermore, some of the critical textures are in Type B inclusions which, as shown later, are probably melt products whose textures no longer reflect the condensation sequence.

Proposed Modes of Origin

SOLID CONDENSATES Much of the interest in coarse-grained inclusions stems from their mineralogical similarity to phase assemblages predicted from thermodynamic calculations to be high-temperature condensates from a gas of solar composition. Early work on condensation calculations and their application to these inclusions was reviewed by Grossman & Larimer (1974). In her study of a Type A inclusion in the Vigarano meteorite, Christophe Michel-Lévy (1968) was the first to suggest that the inclusions are samples of high-temperature condensates, in this case, those predicted in the calculations of Larimer (1967). Marvin et al (1970) and Larimer & Anders (1970) noted the similarity between the minerals in such inclusions from Allende and Ca-, Al-, Ti-rich compounds which Lord (1965) had calculated were near saturation at high temperature in a gas of solar composition and they ascribed a high-temperature condensation

origin to them. The work of Grossman (1972) was the first theoretical investigation of the sequence of condensation of phases from a cooling gas of solar composition at constant pressure where full equilibrium calculations were employed. Those calculations for the abundant condensable elements were extended to include more solid solutions and were updated in response to newer thermodynamic and solar system abundance data in a series of papers (Grossman & Clark 1973, Grossman & Olsen 1974, Grossman 1975b, 1977a,b, Lattimer & Grossman 1978, Lattimer et al 1978, Grossman et al 1979b). Grossman (1972) showed that Ca, Al, and Ti would condense totally as corundum, melilite, perovskite, and spinel at temperatures above the point where significant fractions of the most abundant condensable elements, Mg, Si, and Fe, would begin to condense. He showed further that the assemblage melilite-spinel-perovskite is in complete chemical equilibrium with a gas of solar composition. Using results from Lattimer & Grossman (1978), the temperature range indicated is 1533–1438 K at a reference total pressure of 10^{-3} atm and about 75° lower for each tenfold decrease in pressure considered. This is precisely the mineral assemblage of Type A inclusions, except for hibonite and very minor amounts of rhönite and fassaite. Even the åkermanite content of melilite in these inclusions is in accord with predictions for high-temperature condensates (Grossman 1972, 1975a). Using many arguments first introduced by Grossman & Clark (1973), Allen et al (1978) searched for, but could not find, any textural or mineralogical evidence for the crystallization sequence expected if the fluffy Type A inclusion they were studying had originally crystallized from a melt. MacPherson & Grossman (1979) felt that the highly irregular shapes and intense alteration of fluffy Type A's implied that they formed by accretion of grains or grain-clusters into highly porous aggregates and suggested that those inclusions are composed of direct gas-to-solid nebular condensates that have not been melted or metamorphosed since they accreted. It follows that all phases present in these inclusions have to be considered as candidates for high-temperature condensates, even though lack of thermodynamic data for some of them (hibonite, fassaite, and rhönite) prevents predictions of their stabilities in a solar gas (Allen et al 1978). Hence, hibonite is generally considered the first major element-bearing phase that condensed, instead of corundum which plays this role in the calculated condensation sequence (Grossman 1972, Blander & Fuchs 1975). Carrying the analogy further, hibonite must be a relict phase in these melilite-, spinel-rich inclusions since the latter phases are calculated to coexist only after corundum has reacted completely with the gas to form them. There are also good theoretical reasons to believe that fassaite could be a high-temperature condensate. Diopside is such a phase (Grossman

1972) and Grossman & Clark (1973) estimated that large amounts of $CaTiAl_2O_6$ and $CaAl_2SiO_6$ might dissolve in it in the temperature range of interest. The phase Ti_3O_5 reaches unit activity in a solar gas at temperatures slightly below the condensation temperature of forsterite (Lattimer & Grossman 1978) and it is thus certainly conceivable that significant amounts of trivalent Ti could have condensed in solid solution in pyroxene at higher temperature. It also follows that textural and chemical properties of phases in these inclusions should be directly interpretable in terms of physico-chemical conditions in the solar nebular gas. An example of this approach is the suggestion by Allen & Grossman (1978) that melilite grains in a fluffy Type A whose cores are richer in the low-temperature åkermanite component than their rims may have condensed during a period of falling pressure. This approach is still largely unexplored, however, because of the relatively little work done on fluffy Type A's, because of the hesitation of many workers to believe that any coarse-grained inclusions escaped melting, and because of the extensive obscuration of primary features in these inclusions by secondary alteration products. The major mineral phases of compact Type A's are also identical to those expected from high-temperature condensation, but, in these inclusions, textural evidence for an origin by aggregation of grains is less compelling than in fluffy Type A's. They are much less altered, implying that they were not as porous. Their shapes are near-spherical, suggesting that they may have crystallized from melt droplets. Hibonite, supposedly the first condensate, occurs only at their margins, rather than in their centers. In an SEM (scanning electron microscope) study of a compact Type A, Hutcheon (1977) observed epitaxial growth of perovskite on surfaces of spinel crystals which, in turn, are poikilitically enclosed by melilite. He pointed out that the implied crystallization sequence, spinel, perovskite, and then melilite, is not predicted by the equilibrium condensation model. Again, however, relatively little work has been done on Type A's.

LIQUIDS Far more petrographic work has been done on Type B's and it is upon studies of these that most arguments for a liquid origin for coarse-grained inclusions are based. Ever since their discovery, they were referred to as chondrules (Clarke et al 1970) because their near-spherical shape and the large sizes of their constituent crystals suggested that they were solidified melt droplets (Mason 1975). The term "igneous texture" was used to describe these tightly interlocking assemblages of coarse crystals (Mason & Martin 1974). Furthermore, the crystallization sequence inferred from textural relations in these inclusions was found to be the same as that expected for solidification of melts of similar

composition (Clarke et al 1970, Mason 1975). The validity of the latter argument is questionable, however, since the synthetic system with which the inclusions were compared contained no Ti whose presence in two valence states in the inclusions may have an important effect on the stability of fassaite, one of the major phases. Blander & Fuchs (1975) made detailed petrographic observations of Type B inclusions, pointed out how these textures were identical to those seen in terrestrial igneous rocks of different composition, and concluded that the inclusions must have crystallized from melts. *But it has never been shown that the same textures cannot be produced by condensation of silicates from slowly cooled vapors of moderate pressure.* Blander & Fuchs (1975) argued against a condensation origin for the same inclusions by showing also that the crystallization sequence recorded in their textures is different from that predicted by the equilibrium condensation model. Similarly, Hutcheon (1977) observed epitaxial growth of perovskite on spinel crystals that are poikilitically enclosed by fassaite and pointed out that the implied crystallization sequence, spinel, perovskite, and then fassaite, does not conform to the predictions of this model. Against this, it must be argued that we simply may not know the exact condensation sequence because of the unknown effect of fassaite upon it, that departures from equilibrium may have occurred during condensation, and that phases may not have accreted in their order of condensation. Among the most compelling of Blander & Fuchs' arguments that Type B inclusions are not direct solid condensates is that they contain melilite and spinel, which condense above forsterite, and anorthite, which condenses below forsterite (Lattimer & Grossman 1978), but they do not contain forsterite itself. Because of the fassaite problem, however, even this argument may not be definitive, particularly since condensation calculations predict that anorthite forms by a reaction between spinel and diopside, the most likely phase that would be replaced by fassaite in the condensation sequence.

A revealing example of the ambiguity of the textural evidence in deciding between a liquid or solid origin lies in the different interpretations of spinel framboids. El Goresy et al (1979b) suggest that they formed by condensation of spinel around preexisting solid condensates and were themselves overgrown by later solid condensates. Wark et al (1979), on the other hand, favor an origin by incorporation of small droplets with solid spinel rims by larger droplets. Framboids have never been observed in fluffy Type A's.

Perhaps the strongest case for a once-molten Type B is the one studied by MacPherson & Grossman (1979). The fact that melilite crystals have had their growth interfered with by one another implies that the inclusion formed by in situ growth of melilite, rather than by random accretion of

condensate melilite grains that had been suspended in the gas. Furthermore, because the low-temperature åkermanite component increases in concentration along the length of these crystals toward the center of the inclusion, they grew from the edge of the inclusion inward as temperature fell. It is difficult to see why condensation of solids from a vapor should lead to formation of a spherical shell that was filled in by later condensates. It is much easier to see how the observed textures could have been produced by crystallization of a suspended melt droplet, which would have cooled and solidified from the outside in by radiating heat away from its surface. This inclusion must have been molten, but whether this is true for other Type B's that do not show these chemical and textural properties is less clear.

How liquids of these compositions could have formed is also not known. Grossman & Clark (1973) showed that equilibrium condensation at pressures slightly above 10^{-3} atm would occur at temperatures slightly above minimum solidus temperatures for these compositions, producing partial melts. Complete melting would require condensation at much higher pressures than are normally considered for the inner part of the solar nebula (Cameron & Pine 1973), but which may have prevailed in giant gaseous protoplanets, which have been proposed as a source for these inclusions by Podolak & Cameron (1974) and Consolmagno & Cameron (1980). Blander & Fuchs (1975) argued that even low pressure condensation would produce melt droplets of these compositions because metastable, subcooled liquids would condense instead of the equilibrium solids. It is also possible that such melt droplets are secondary in origin, formed by melting of preexisting solid condensates in energetic nebular processes, such as lightning discharges (Whipple 1966, Cameron 1966, Sonnett 1979) or impacts between small, high velocity solid objects (Whipple 1972, Cameron 1973, Kieffer 1975).

In studies of coarse-grained inclusions in Lancé (Kurat 1970, 1975) and Bali (Kurat et al 1975), an origin by crystallization from liquids was inferred from textures. These workers were convinced that the bulk chemical compositions were established in gas-condensed phase reactions but, noting the thermodynamic equivalence of partial condensation and partial evaporation, they suggested that these liquids formed as evaporation residues by repeated impact events on parent bodies of chondritic composition. Chou et al (1976) also argued that the inclusions are volatilization residues, but of interstellar matter. The latter view was shared by Notsu et al (1978) and Hashimoto et al (1979) who produced Ca-, Al-rich residues in high-temperature vaporization experiments on a bulk sample of Allende and on the fine-grained fraction of the Murchison C2 chondrite, respectively. We will return to some of these ideas later.

Phase relations Knowledge of the sequence of crystallization of minerals expected from melts having the compositions of coarse-grained inclusions would be an invaluable aid to interpretation of their textures. Very little is known, however, about the low-pressure, liquid-crystal phase relations in the system applicable to these objects, $CaO–Al_2O_3–MgO–TiO_2–Ti_2O_3–SiO_2$. Although no compositions studied by Yagi & Onuma (1967) along the join $CaMgSi_2O_6–CaTiAl_2O_6$ are applicable to the inclusions, two studied by Yang (1976) and two by K. Onuma & Kimura (1978) in the plane $CaMgSi_2O_6–CaAl_2SiO_6–CaTiAl_2O_6$ appear to be close to inclusion bulk compositions. In none of these studies, however, were oxygen fugacities buffered so that Ti^{3+} could coexist with Ti^{4+}. This is also true in Seitz & Kushiro (1974), who determined the crystallization sequence in a melted fine-grained inclusion. Although that particular inclusion is close in composition to the coarse-grained ones discussed herein, it contains 2.3% FeO and 1.1% Na_2O which are not present in the primary phase assemblage of coarse-grained inclusions. Using petrographic and electron microprobe techniques, Butler (1977) studied a slowly cooled blast furnace slag that was remarkably similar in composition to coarse-grained inclusions, except for a total of 4% Fe, S, K_2O, Na_2O, and MnO. He found that melilite crystallized before Ti-, Al-pyroxene, in agreement with the crystallization sequence inferred by MacPherson & Grossman (1979) for their Type B inclusion. This reviewer considers Butler's (1977) results to be particularly significant because the slag crystallized under reducing conditions, as indicated by the presence of metallic Fe, and the pyroxene stoichiometry suggests that Ti may be present both as Ti^{3+} and Ti^{4+}.

MICRO-MINERALOGY AND MICRO-TEXTURES

Trace Phases

Palme & Wlotzka (1976) discovered an Fe-Ni particle containing minor to major amounts of Mo, Ru, Rh, W, Os, Ir, and Pt in a Type B inclusion. Grossman (1973) had calculated that Mo, Ru, W, Os, and Ir could have condensed totally as pure metals at temperatures above or within the range of condensation temperatures of the major phases in the inclusions and had predicted their presence therein. Palme & Wlotzka (1976) considered the condensation of those elements, together with Re, Pt, Rh, Fe, Ni, and Co, into a common alloy using an ideal solution model. Because the bulk composition of their metal particle has roughly chondritic proportions of refractory siderophiles, they inferred from their calculations that it last equilibrated with a solar gas between 1460 and 1500 K at 10^{-3} atm, in accord with temperatures inferred above from the silicate and oxide

assemblage. Within the particle, however, are regions in which some refractories are concentrated relative to others and regions of sulfides. These were attributed to low-temperature exsolution from a primary condensate alloy and to a secondary alteration process involving introduction of volatile S. At about the same time, the tremendous potential of the SEM/X-ray analyser for characterizing such phases was demonstrated by Lovering et al (1976) who reported four trace phases, <2 μm in size, rich in rare earth elements (REE), Th, Nb, U, and Zr from Type B inclusions and by Wark & Lovering (1976) who analysed ten refractory siderophile-rich alloys, 0.5–3 μm in diameter, from a Type A. For more details of the compositions of the refractory lithophile-rich phases and others, see Lovering et al (1979). On the basis of previous condensation calculations, which showed that all these elements are refractory in a gas of solar composition (Grossman & Larimer 1974), these phases were all interpreted as condensates.

In a series of contributions (El Goresy et al 1977a, 1978a,b, 1979a), it was shown that the trace phases have two major modes of occurrence, either individual metal nuggets, $\sim$1–2 μm in size, or complex assemblages, "Fremdlinge," of metal alloys, sulfides, phosphates, oxides, and silicates. These aggregates are usually 3–15 μm in size and are composed of smaller grains, some in the submicron range. Although Wark & Lovering (1978a, b) and Lovering et al (1979) find Fremdlinge only in Type B inclusions, El Goresy sees them in both A's and B's, but notes that individual beads and Fremdlinge are not usually found together in the same inclusion.

Both El Goresy et al (1978b) and Wark & Lovering (1978b) agree that the individual metal nuggets contain Mo, Ru, Rh, W, Re, Os, Ir, and Pt in roughly chondritic proportions, although these elements may be segregated from one another into separate regions within each nugget, as seen by the former workers and inferred by the latter workers. Usually, these nuggets contain substantial amounts of Fe ($\sim$10%) and Ni ($\sim$1%), but some in Type B's have much lower concentrations. Wark & Lovering (1978b) made the important observation that nuggets within a single host melilite crystal are closer in composition to one another than to nuggets within another crystal in the same Type A inclusion, implying that the inclusion was never molten. This reviewer agrees with their assessment that it is of utmost importance to confirm this observation with more statistics, but suggests that results be obtained separately for fluffy and compact Type A's. El Goresy et al (1978b) also made an important observation whose confirmation would have a profound effect on views of the origin of the inclusions. Inside some metal grains, they found several percent Zr, with no evidence for its presence as a separate oxide phase. This is important because Zr condensation calculations show that even in a

gas as reducing as one of solar composition, ZrO_2 is 10^7 times more stable than metallic Zr at its condensation temperature. This factor is so great that even if tremendously nonideal solutions of Zr in platinum metal alloys are postulated, Zr should still condense in oxidized form. Furthermore, it is doubtful that, after condensing in this way, Zr could have been reduced to the metal during later, lower-temperature secondary reactions. If the observation is correct, it would be difficult to deny that the grains in question formed under much more reducing conditions than those of a solar gas.

Blander et al (1980) discovered Pt-metal nuggets in a Type B in which refractory siderophiles were not in cosmic proportion to one another. They proposed isolation of grains from gas at a temperature above that where all these elements would be totally condensed, later oxidation by other elements in the inclusion, uncertainties in condensation models due to assumptions of ideal solution, and a source region of nonsolar composition as possible explanations. As in previous work (Fuchs & Blander 1977), they stressed the possible effects of supersaturation, perhaps due to coating over of early formed nuclei by other condensates, on deviation of the subsequent course of condensation from the equilibrium path.

El Goresy et al (1978b) classify Fremdlinge into three types. Type 1 are compact and contain 50–70% by volume of Fe-Ni metal, sulfides and refractory siderophile alloys, and the remainder phosphates, oxides, and silicates. Re, Os, Ir, Pt, Rh, and Ru are not alloyed with Fe-Ni, but are present as submicron grains, either individually or in predominantly binary, ternary, or quaternary alloys. Situations are common in which a grain of pure Os is found only a few μm from one of pure Ir (Wark & Lovering 1978b). Zn, Ga, Ge, Sn, and As are sometimes alloyed with Ni, Fe, and/or Pt metals. In the Fe-Ni alloys, a wide range of Ni/Fe ratios can be found within the same inclusion. Mo and W form sulfides and alloys with Pt metals. V and Nb occur in oxides and sulfides. Silicates are clinopyroxene, melilite, anorthite, wollastonite, nepheline, sodalite, and K-, Na-, Ca-phases. Type 2 contain 70–90% Fe-Ni. Mo and W sulfides and Pt metal alloys occur inside silicates and Fe-Ni. Type 3 contain 0–70% Fe-Ni which, with sulfides, usually occupies the core of the Fremdling, around which is a spongy rim of oxides, phosphates, and silicates. Pt metal alloys are found throughout. Other phases are ZrO_2, pyroxene, spinel, anorthite, nepheline, and sodalite. Individual Fremdlinge are sometimes enriched in a single element, such as Mo, V, or Sc, relative to all the other rare elements normally encountered.

El Goresy et al (1978b) and Wark & Lovering (1978b) consider the individual metal nuggets to be solar nebular condensates, largely because they contain solar proportions of refractory siderophiles. The former

workers ascribe the origin of Fremdlinge to accretion of components that condensed in a large number of events from a large number of chemically distinct reservoirs, many of which were nonsolar in composition and possibly pre–solar nebular in origin. Many constituents of the Fremdlinge, Ca-phosphates, nepheline, sodalite, Fe-Ni sulfides, and alloys of volatile transition metals such as Ga, Ge, Zn, and Sn, are simply not stable in a gas of solar composition at the high temperatures at which their enclosing host phases condense. The first major problem for El Goresy's model is thus preservation of these phases during the later high-temperature event that produced the major phases in the inclusions, a fact that caused El Goresy et al (1978b) to assert that not even the major phases condensed from a gas of solar composition.

The second major problem for this model is that it assumes that all minerals in the Fremdlinge and textural relations between them were produced during condensation and have not been modified during later melting events. Liquid-crystal phase equilibrium data are lacking for multicomponent Pt metal systems. From the high melting points of the end-members, however, it is conceivable that some alloy compositions would remain solid even at the high temperatures required for total melting of the major phase assemblage of the inclusions, as the siderophiles would not dissolve in molten silicates. This would not be the case for the lithophiles, however. Even though melting points of some phases present, such as thorianite and baddeleyite, are $>2500°C$, they may dissolve totally in silicate melts at much lower temperatures. Surely, the Fe-Ni sulfide fraction and nepheline and sodalite would have melted if the major primary phases did. Thus, many mineralogical and textural features which El Goresy attributes to condensation would not have survived such a melting event. As discussed above, fluffy Type A inclusions may have never been molten and Type B1 inclusions like that studied by MacPherson & Grossman (1979) probably were. Which other inclusions were molten is still not clear. Although El Goresy has not yet combined his data on Fremdlinge with complete petrographic descriptions of their host inclusions, such a synthesis would be extremely valuable.

Wark & Lovering (1978b) challenged El Goresy's basic assumption that all features in Fremdlinge predate the phases that enclose them, arguing that open-system secondary alteration processes are responsible for many of their characteristics. Their observation that corroded Fe-Ni grains are surrounded by magnetite and intersect cracks filled with it is certainly conclusive evidence that this has occurred in some cases. Magnetite is only stable in a solar gas below 400 K. Furthermore, the silicate assemblage of Fremdlinge includes anorthite, wollastonite, nepheline, and sodalite, phases characteristic of secondary alteration products that form

at the expense of primary silicate and oxide phases of the inclusions (Allen et al 1978) and therefore must postdate any melting event. If the silicates and magnetite are indeed secondary, the possibility exists that all of the enigmatic volatiles entered Fremdlinge in such reactions. El Goresy et al (1978b) insist, however, that the overwhelming majority of Fremdlinge show no connection to magnetite-filled cracks. What fraction of the Fremdlinge have been so affected is still unclear. Nor is it clear what effects, other than introduction of volatiles, may be due to secondary alteration. Neither this process nor exsolution (Wark & Lovering 1978b), however, seems capable of producing such features as virtually pure Os and Ir grains only a few μm apart, entire Fremdlinge composed predominantly of a single refractory element, inclusions of MoS_2 in Fe-Ni that has not reacted to form a sulfide (El Goresy et al 1978b), or Ca-phosphates containing 3% RuO_2 (El Goresy et al 1979a). These and other observations suggest that different constituents of Fremdlinge, some of which are now intimately associated with one another, were formed in different physico-chemical environments. In addition, Ru-bearing phosphate must have formed in a system far more oxidizing than a solar gas.

Alteration Products

Many papers dealing with petrography of coarse-grained inclusions mention dark, opaque, fine-grained regions in thin sections of them, but it was the SEM work of Allen et al (1978) that provided the most information about this material in fluffy Type A's. They found that such regions consist of cavities and veins whose walls are lined with euhedral crystals of grossular and which contain mats of acicular wollastonite crystals, euhedral nepheline crystals projecting inward, and anorthite. Some wollastonite needles are bent through nearly 90°, but are not broken, indicating great strength such as is found for vapor-deposited whiskers. Their presence and euhedral crystals partially filling void spaces are clear evidence for their origin by condensation from a vapor. The fact that this material corrodes and veins major mineral phases means that minerals in the cavities are secondary alteration products of those primary phases. Thus, after formation of hibonite, melilite, and spinel in fluffy Type A's, these minerals reacted partially with a gas phase to form the alteration products. Allen et al (1978) found that alteration products occupy more than 75% of the inclusion they studied. The intense alteration suffered by these inclusions implies that the reacting vapor had easy access to the primary phases, one of the major reasons why MacPherson & Grossman (1979) believe these inclusions were loose aggregates at that stage.

Large amounts of similar alteration products are found in Type B's

where they are not as abundant as in A's. The small grain sizes of the alteration phases make the altered regions too opaque to study by normal optical techniques. Hutcheon et al (1978b) studied several Type B's by cathodoluminescence. This technique has the advantageous capability of portraying different phases in these regions in characteristic and vivid colors. This study revealed that chemical and/or structural changes have occurred in anorthite, melilite, and fassaite next to fractures and embayments containing alteration products. Since the delicate textures of the alteration products would have been destroyed during melting, their presence in once-molten inclusions implies that vapor phase reactions occurred after melting and crystallization.

Interpretation of the alteration products as condensates from a vapor presents important difficulties. Neither wollastonite nor nepheline nor sodalite can condense from a gas of solar composition at equilibrium. Either the alteration process was not an equilibrium one or the reacting gas was not solar in chemical composition. Arrhenius & Alfvén (1971) and Arrhenius (1972, 1978) took the coexistence of volatile Na in nepheline and sodalite with refractories to mean that they condensed simultaneously due to ionization-controlled sublimation from a low-pressure, high-temperature plasma in which grains were not in thermal equilibrium with the gas. From the textural relations, however, it is now clear that Na was deposited in the inclusions in an event that postdated formation of the primary refractory phases. The presence of nepheline, anorthite, and wollastonite in Fremdlinge led to the suggestion, above, that some of them may have been affected by the same alteration process as the major phases. Although the mechanism for introduction of Na is not understood, other volatiles such as are found in some Fremdlinge may have accompanied it.

Both Clarke et al (1970) and Fuchs (1974) noted that grossular decomposes to wollastonite, melilite, and anorthite above 1073 K. Unless grossular nucleated metastably above this temperature, this is an upper limit to the condensation temperature of this phase in the alteration products. Fuchs (1974) interpreted such grossular in a Type B inclusion as a devitrification product of a glass below 1073 K, supporting the contention of Blander & Fuchs (1975) that the inclusions crystallized from a melt. The textural observations of Allen et al (1978) suggest that this interpretation is incorrect.

Rims

Even before the fall of Allende, Christophe Michel-Lévy (1968) had noted a spinel-olivine rim around a Type A inclusion in the Vigarano meteorite. Frost & Symes (1970) saw pyroxene rims on a perovskite-

bearing inclusion and Kurat (1970) described nepheline-, perovskite-, and diopside-rich rims on Type A's in Lancé. In Vigarano, Reid et al (1974) illustrated rims on Type A's. In Allende, Blander & Fuchs (1975) described diopside rims around cavities inside a Type A inclusion and perovskite-spinel and Na-rich rims on Type B's. Diopside rims around Type A's and lining cavities in their interiors, spinel rims on Type B's, and olivine rims around inclusions of both types were reported by Grossman (1975a).

These reports were greatly augmented and clarified by SEM observations of polished sections and the data were synthesized into the following picture by Wark & Lovering (1977a,b, 1978a). The outermost region of every coarse-grained inclusion consists of a narrow (usually $\lesssim 60$ μm) sequence of mineralogically distinct zones. Furthermore, there are mineralogical differences between the rims on Type A, on Type B, and on intermediate inclusions. From inside to outside, the sequence in Type A rims is Fe-bearing spinel + perovskite, nepheline + sodalite + melilite + forsterite + anorthite, fassaite which gradually changes its composition outward to diopside, and, finally, hedenbergite + andradite. Allen et al (1978) discovered in a fluffy Type A a 300 μm thick layer consisting largely of porous masses of clinopyroxene prisms on the exterior surface of the outermost zone seen by Wark & Lovering. Type B rims consist of Fe-bearing spinel + perovskite, forsterite + nepheline, and hedenbergite + diopside.

Textures within each zone are not clastic. There is usually little void space and crystals are snugly intergrown with one another. The resulting massive appearance of the rims suggests that their constituents crystallized in situ and are not accreta that originally crystallized elsewhere. Because of the paucity of FeO in the minerals in inclusion interiors and its presence in rim phases, Christophe Michel-Lévy (1968) thought that rims are high-temperature reaction products between inclusion interiors and the FeO-rich matrix of the meteorite, a suggestion also made by Reid et al (1974). Because the diopside rims observed by Kurat (1970) and Grossman (1975a) are FeO-free, these workers stated that this phase could not be a reaction product between the inclusions and the meteorite matrices but, instead, between them and a gaseous, pre-agglomeration medium. Perhaps the best evidence that reaction with the matrix was not involved is the observation of both Blander & Fuchs (1975) and Wark & Lovering (1977a) that rims are absent from jagged surfaces of inclusion fragments, but present on the smooth, rounded surfaces of the same fragments, even though all surfaces are in contact with the matrix. This implies the following sequence: inclusion solidification, rimming, fragmentation, and incorporation into the meteorite.

The rim sequence could have been produced by reaction with a single medium, perhaps even at fixed pressure and temperature, such as in the case of alteration zones formed in terrestrial contact metasomatic processes. In this event, rim mineralogy would be strongly dependent on the composition of the inclusion interior. Thus, simply because they have different rim sequences from one another does not necessarily lead to Wark & Lovering's (1977a) conclusion that Type A and Type B1 inclusions were rimmed in different physico-chemical regimes, because the chemical compositions of these different inclusion types are very probably different. On the other hand, each layer could have formed by precipitation from, or reaction with, a separate physico-chemical environment, in which case the mineralogy would not necessarily depend on the composition of the inclusion interior. Only if the layers formed in this way does the Wark & Lovering suggestion follow. It is the thick melilite mantles of Type B1 inclusions that differentiate them from B2's. Thus, during rim formation, each of these inclusion types had different mineral assemblages in chemical communication with the external medium and, if the first of the above alternatives is correct, a different rim sequence should have formed on each. If the second alternative is correct and all Type B's formed in the same place, they should all have the same rim layers. Which is correct is not yet known, as Type B2 rims have not been studied as thoroughly as B1 rims (Wark & Lovering 1977a).

Observations that rims overlie alteration products imply that the alteration event predated rimming (Allen et al 1978). Because rims formed before the inclusions were incorporated into the meteorite and because alteration products never breach the rim, the source of vapor for the alteration process could not have been the Allende matrix. Perhaps the alteration phases were the first result of the same process that later formed rims.

Serious problems arise if rims are interpreted as condensates. Nepheline, sodalite, and andradite are not known to condense from a gas of solar composition at equilibrium. Either they condensed in a nonequilibrium process or they formed from a gas whose composition was not solar. This is also suggested by the occurrence of diopside exterior to nepheline and sodalite in Type A rims, indicating condensation of Na-bearing phases prior to diopside. New approaches will be required to learn more about the origin of alteration products and rims. One promising possibility is the inversion of condensation calculations. In this approach, any mineral assemblage of interest could be assumed to be an equilibrium condensate assemblage and the composition of the gas phase with which it is in equilibrium could be calculated. If the compositions so calculated have any astrophysical significance, new insights about the inclusions would be suggested.

BULK CHEMICAL COMPOSITION

Major Elements

Complete major element compositions of individual coarse-grained inclusions have been published for the following samples: one, labeled inclusion 1, in Gray & Compston (1974), three in Graham (1975), five, labeled Group I, in Conard (1976), nine, labeled Group I, in Mason & Martin (1977), four, Groups I, III, V, VI, in Taylor & Mason (1978), and another, CG-11, in Davis et al (1978b). The latter is an extremely heavily altered fluffy Type A whose composition is 29.4% CaO, 37.6% Al_2O_3, 4.3% MgO, 1.0% TiO_2, 25.1% SiO_2, 1.7% FeO, and 0.8% Na_2O. From its mineralogical description, the one analysed by Gray & Compston (1974) is probably also a fluffy Type A. Its composition is almost identical to that studied by Davis et al (1978b). Although the types of inclusions studied by Taylor & Mason (1978) are unknown, those analysed by Graham (1975), Conard (1976), and Mason & Martin (1977) are probably Type B's. Their compositions are in the range 21–37% CaO, 21–33% Al_2O_3, 5–15% MgO, 1.0–1.7% TiO_2, 24–36% SiO_2, 0.3–3.1% FeO, and 0.06–1.7% Na_2O. As Grossman & Larimer (1974) and Grossman (1975a,b, 1977a,b) pointed out, these bulk compositions are close to those of high-temperature condensate assemblages calculated to form before condensation of significant fractions of Mg, Si, and Fe, except for the presence of relatively small amounts of FeO and Na_2O which must be ascribed to introduction of volatiles during secondary alteration and rimming processes (Wänke et al 1974, Grossman 1975a, Grossman & Ganapathy 1975).

Do Types A and B inclusions have the same bulk chemical composition? Do they plot precisely along the predicted trajectory of condensates in composition space? These are important, first-order questions that have not been answered by the above data for the following reasons. First, because crystal sizes of the major phases are often large compared to the inclusions in which they are found and because inclusions like the B1's have thick, monomineralic mantles, the size of an unbiassed sample for major element analysis should be comparable to that of the entire inclusion. Samples smaller than this will only be representative if they are splits of homogenized powders made from nearly entire inclusions and this was definitely not the case in Conard (1976), whose samples were obtained and described by Gray et al (1973), in Davis et al (1978b), or in Taylor & Mason (1978). There is no indication that such precautions were taken by Gray & Compston (1974), Graham (1975), or Mason & Martin (1977). Second, because of the presence of relatively large amounts of secondary alteration products, bulk analysis will not in general yield

the composition of the primary mineral assemblage alone (Davis et al 1978a), even when great care is taken to ensure representativeness. Davis et al (1978a) concluded that Type A's are lower in MgO than B's, based on data for CG-11 and unpublished data from this laboratory for three other A's and several B's. The significance of this conclusion, however, was obscured by the above problems. Wark & Lovering (1978a) reached a similar conclusion about MgO and found other differences as well, but their broad-beam electron microprobe analyses of polished sections apparently also suffer from contamination by alteration products and may have the matrix correction problems often encountered when this technique is applied to coarse-grained samples. These criticisms also apply to the data used by McSween (1977). Better estimates of major element compositions of primary phase assemblages are those of Grossman (1975a). Those were computed from means of large numbers of spot microprobe analyses of the major phases and ranges of modal analyses, excluding alteration products, observed in many inclusions. From these, MgO and SiO_2 are lower and CaO higher in Type A's than B's. This technique was applied individually to two Type A and three Type B inclusions by Grossman & Ganapathy (1976b). Although the above results were confirmed for MgO and CaO, the SiO_2 data overlap in this study. It is clear that the latter approach must be applied to many more inclusions before the questions posed above can be answered with precision.

Trace Elements

PREDICTIONS OF CONDENSATION MODELS Results of calculations of equilibrium condensation temperatures of many trace elements from a gas of solar composition are given by Grossman (1973, 1977b), Grossman & Larimer (1974), Boynton (1975a, 1978a), Grossman & Ganapathy (1976b), Palme & Wlotzka (1976), Ganapathy & Grossman (1976), Grossman et al (1977), Davis & Grossman (1979), and Blander et al (1980). They show that Os, Re, Ir, Ru, W, and Mo could have condensed completely as pure metals and that Zr, Hf, Y, Sc, and some REE could have condensed as pure oxides at temperatures above the accretion temperature of the inclusions (1438 K at 10^{-3} atm, for example). None of the pure crystalline phases of Pt, Rh, V, Ta, Th, U, Pu, and the remaining REE for which thermodynamic data exist are capable of condensing these elements totally above this temperature. Calculations show, however, that these elements can do so if they form ideal solid solutions either with other siderophiles in the case of Pt and Rh (Palme & Wlotzka 1976) or in condensed oxides and silicates of more abundant elements in the case of Th, U, and Pu (Ganapathy & Grossman 1976), REE except Eu (Grossman

& Ganapathy 1976b), Ta, and V. It is thus not certain that these elements can condense above this temperature because the complete absence of activity coefficients for them in relevant phases dictates use of simple ideal solution models, which overestimate condensation temperatures for those components that actually exhibit positive deviations from ideality (Boynton 1975a, 1978a). On the other hand, perhaps only a few of the elements discussed here condensed in the forms calculated, but rather as more stable pure compounds, such as some of the phases found in Fremdlinge, for which there are no thermodynamic data. Substantial concentrations of those trace elements that can condense above the accretion temperature of the inclusions should be found in the inclusions, because those elements may have condensed in solid solution in major condensate phases, they may have nucleated upon them, or they may have acted as condensation nuclei for them (Grossman 1973).

GENERAL OBSERVATIONS The Allende literature contains many trace element or isotopic analyses of objects for which there are either no accompanying mineralogical or petrographic descriptions or descriptions that are not detailed enough to tell if the inclusions in question are coarse-grained ones. The following are sources of trace element data for bulk inclusions that are either definitely or probably coarse-grained: one in Gast et al (1970), eight in Gray et al (1973; labeled Ca-Al chondrules), 15 in Grossman (1973; omitting sample 8), two in Tanaka & Masuda (1973; inclusion O and Ca-Al rich chondrule), one in Wetherill et al (1973; low Na), one in Osborn et al (1974; Ca-Al chondrule), one in Wänke et al (1974), 10 in a series of papers by Grossman & Ganapathy (1975, 1976b) and Grossman et al (1977), three in Chen & Tilton (1976; ChL-1, ChL-2, and WA), three in Chou et al (1976), five in Conard (1976; Group I), three in Tatsumoto et al (1976; N17, N18, N19), one in Palme & Wlotzka (1976), one in Drozd et al (1977; 3666-I1), 10 in Mason & Martin (1977; Group I), three in Nagasawa et al (1977; 5, 6, 7), one in Davis et al (1978b), and four in Taylor & Mason (1978; Groups I, III, V, VI).

The conclusion reached from all these data is that the average inclusion is enriched in each of the above refractory elements, except V, Rh, and Mo, plus Eu, Ba, Sr, and Nb, by a factor of 15–20 relative to Type 1 carbonaceous (C1) chondrites. These elements span a tremendous range of chemical properties and geochemical behavior. The only thing common to them all is that their condensation temperatures from a gas of solar composition are above or within the range of condensation temperatures of the major mineral phases in the inclusions. Furthermore, excluding samples that were obviously grossly contaminated by Allende matrix

material during their extraction from the meteorite, all the more volatile elements, with the occasional exception of Na, Cl, Br, and Au, are depleted in coarse-grained inclusions relative to C1 chondrites by amounts ranging from 10% to a factor of 320. *The trace element results are thus very powerful evidence that coarse-grained inclusions formed in a gas-condensed phase fractionation process.* It is concluded that Eu, Ba, Sr, and Nb condense in the same temperature range as the other refractories, even though calculations have not yet shown how. Although V (Conard 1976), Rh (Taylor & Mason 1978), and Mo (Mason & Martin 1977) are also enriched relative to C1's, they are not as enriched as other refractories. It should be noted, however, that calculations show that V is one of the last refractories to condense as a pure oxide (Grossman & Larimer 1974) and, unless it formed a solid solution instead, it would not have condensed totally above the accretion temperature of the inclusions. Similarly, Rh is the last refractory siderophile to condense totally into an alloy (Palme & Wlotzka 1976). Mo is a mystery, however, as it is one of the first elements to condense as a pure phase. Perhaps its low enrichment is due to an error in its Cl abundance (Palme & Wlotzka 1976, Blander et al 1980).

Although trace element data cannot be used to chose decisively between a condensation or evaporation origin for the inclusions, they do constrain the type of partial evaporation process that would be acceptable. The key observation is the equal enrichment of refractory siderophiles and refractory lithophiles. In the model advocated by Kurat et al (1975), the inclusions result from repeated cycles of volatilization and condensation during meteorite bombardment of chondritic bodies. These processes should have been accompanied by melting events on the parent body, during which lithophiles would have fractionated from siderophiles due to the tendency of the latter to form melts that are immiscible with and denser than lithophile-rich melts. Thus, although partial evaporation of dust aggregates is a possibility (Chou et al 1976), the trace element composition of the inclusions seems to rule out parent-body volatilization processes.

Condensation calculations predict even lower concentrations of volatiles than are found in the inclusions, except for such elements as Fe, Ni, Co, and Cr (Grossman et al 1979b), which may have begun to condense just above the accretion temperature of the inclusions. Na and Cl are known to be major constituents of phases in alteration and rim assemblages. Wänke et al (1974) found volatiles to be enriched in the outer part relative to the inner part of an inclusion, indicating their association in this and other inclusions with secondary alteration products, rims, and adhering particles of Allende matrix.

Grossman & Ganapathy (1976b) and Grossman et al (1977) determined

concentrations of 21 refractory elements in nine inclusions and found highly variable ratios of some elements to others in individual inclusions, implying that refractories fractionated from one another during condensation and entered inclusions in separate components. When the mean concentration of each refractory element was divided by its C1 abundance, however, almost the same number arose in each case, 17.5 ± 0.4 (see Figure 4). Thus, as a group, coarse-grained inclusions did not fractionate refractories from one another relative to C1 chondrites, suggesting total condensation of each element from a gas of solar composition and indiscriminate incorporation of all condensate components into coarse-grained inclusions as a whole. The composition of C1 chondrites is generally regarded as that of the total condensable matter of the solar system. An enrichment factor of 17.5 relative to C1 chondrites will result if all of a particular refractory element in C1 chondrites is concentrated into 100/17.5 = 5.7 wt % of the total condensable matter. As Grossman et al (1977) argue, this value is in excellent agreement with independent estimates from condensation calculations of the fraction of the total condensable matter that should have condensed above the accretion temperature of the inclusions. This is true, even allowing for the presence of

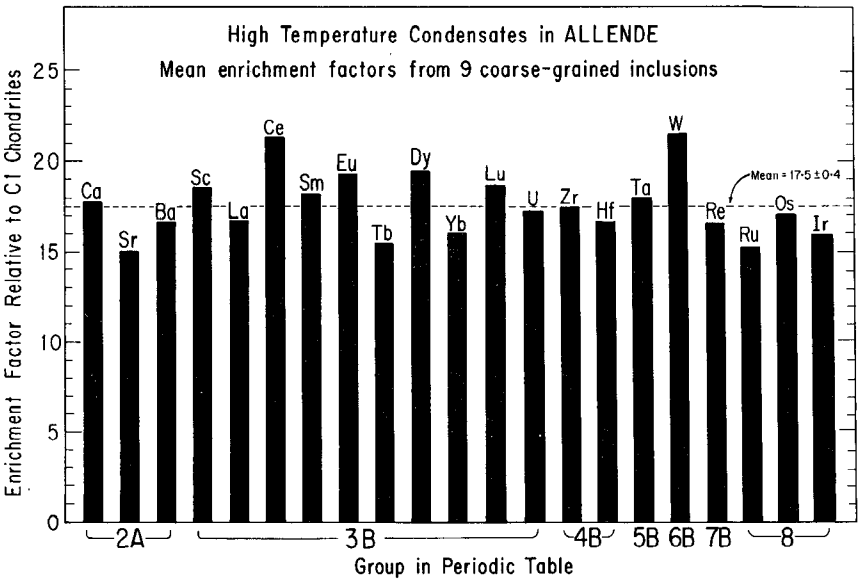

Figure 4 Refractory elements are uniformly enriched relative to C1 chondrites in this collection of coarse-grained inclusions. This indicates total condensation and indiscriminate incorporation of each of these elements into the inclusions, regardless of differences in their chemical properties. From Grossman et al (1977).

substantial amounts of alteration products, provided they introduced no additional refractories.

Applying linear regressions and factor analysis to their chemical data for bulk inclusions, Grossman et al (1977) concluded that Os, Ir, Ru, Re, and most of the W were carried into the inclusions either in a single refractory siderophile element alloy or in a group of such alloys that behaved like a single component, i.e. they did not separate from one another during their incorporation into the inclusions. Similarly, Sc, Zr, Hf, Ta, U, and REE except Eu entered in pyroxene or in one or more trace phases that did not separate from each other. Sr and some Eu were brought in with melilite, either in its crystal structure or in inclusions, and Ba was carried in alone in a separate phase.

Of all the inclusions for which bulk trace element analyses are reported, only two in Grossman & Ganapathy (1975, 1976b) and Grossman et al (1977) and one in Davis et al (1978b) are known to be Type A's. These are simply too few data points upon which to make comparisons between A's and B's. They were used, however, together with unpublished data from this laboratory for three additional Type A's to conclude that Sc-REE correlations are different in the two inclusion types (Davis et al 1978a). If this is verified by further studies of more Type A's, the conclusion would be that different refractory condensate components entered each type, implying that different physico-chemical environments were involved in the condensation of each. This would also seriously affect conclusions reached in the previous paragraph, as pooled data for Types A, B, and I inclusions were used in that study to arrive at correlations from which condensate components were inferred that were therefore constrained to be common to all types.

RARE EARTHS When normalized to C1 chondrites, REE in most coarse-grained inclusions show flat patterns at enrichments of 15–20, except for Eu. This is presumably due to the fact that, except for Eu, REE were totally condensed into one or more phases which were incorporated into each inclusion as a single component. Eu entered the inclusions, at least partially, in a separate component. Davis et al (1978a) noted that, while Type B inclusions can have positive or negative Eu anomalies, A's only have positive ones. Inclusions of both types are known with no Eu anomalies. All of the above are common REE patterns in coarse-grained inclusions, referred to as Group I patterns by Conard (1976), Mason & Martin (1977), and Nagasawa et al (1977).

A radically different REE pattern, referred to as Group II, was first found in Allende fine-grained inclusions by Tanaka & Masuda (1973). [Space does not permit detailed discussion of these, but petrographic

descriptions are in Blander & Fuchs (1975; inclusion 7/9), Grossman et al (1975), and Grossman & Ganapathy (1975) and chemical data in Conard (1976), Grossman & Ganapathy (1976a), Mason & Martin (1977), and Nagasawa et al (1977).] These are characterized by being relatively uniformly enriched in light REE by factors of 20–60, having deep negative Eu anomalies, steadily decreasing enrichments from Gd to Er, sharp positive Tm anomalies, and a steep drop through Yb to Lu. Boynton (1975a) used thermodynamic calculations to show that a pattern with these general characteristics would result in fine-grained inclusions if they condensed with the REE remaining in the gas after prior removal of the most refractory REE in an earlier condensate. This is very strong evidence that a REE-bearing component in these inclusions formed in gas-condensed phase fractionation processes. In particular, this component cannot be a vaporization residue, or else it would have the inverse pattern. This is supported by volatilization experiments on REE-doped, Ca-, Al-rich glasses which yield residues that are enriched in heavy REE relative to light ones (Nagasawa & Onuma 1979). Arrhenius (1978) argued that Group II patterns formed by plasma condensation and that thermodynamic calculations neglecting charged species are an oversimplification of the real situation, regardless of their success at modeling the patterns. Boynton (1975b) showed, however, that REE patterns unlike any so far seen in Allende would result if REE condensation were controlled by ionization potentials.

Boynton (1975a) assumed that discrepancies between the pattern observed by Tanaka & Masuda (1973) and the one he calculated by assuming ideal solution of REE in the removed phase are due to nonideal effects and computed "activity coefficients" that would make the two patterns match as closely as possible. Davis & Grossman (1979) felt that relative activity coefficients required by this model were too high for the temperature being considered, 1650 K, and found that when such a model was applied to 20 Group II patterns in the literature, relative activity coefficients required by the model had to vary by unreasonably large amounts from inclusion to inclusion over a narrow temperature range. Instead, Davis & Grossman (1979) showed that most discrepancies could easily be explained by incorporation of a variable, but usually small, amount of a second REE component with a flat REE pattern into each inclusion.

Boynton (1978a) derived relative activity coefficients for Th, U, Pu, and Cm based on those in Boynton (1975a) and found that nonideal effects of the calculated magnitude enhanced considerably the solar nebular volatilities of Th, U, and Pu relative to the ideal solution calculations of Ganapathy & Grossman (1976). He further asserted that these workers

were incorrect in concluding that Th, U, and Pu were each so refractory that they should be present in unfractionated proportion to one another in large collections of coarse-grained inclusions, claiming that, due to his newly calculated volatility for U, this element should be depleted relative to other refractories. If, as Davis & Grossman (1979) suggest, REE activity coefficients derived in Boynton (1975a) are vastly in error, then Boynton's (1978a) conclusions about Th, U, and Pu derived therefrom are also in error. This seems to be confirmed by Grossman et al's (1977) finding that U has the same enrichment factor as all other refractories in coarse-grained inclusions.

Group II REE patterns are sometimes found in coarse-grained inclusions: CG-5 (Type B) of Grossman & Ganapathy (1976b) and 4691 (Type B) of Mason & Martin (1977). The conclusions reached by Boynton (1975a) and Davis & Grossman (1979) are thus also applicable to these samples. Two REE components are present, one that was isolated from its gaseous reservoir after prior loss of a higher-temperature condensate and one from a source that suffered no prior loss. All solar nebular condensation calculations consistently underestimate the Tm abundance in inclusions with Group II REE patterns. Davis & Grossman (1979) suggested therefore that the first of the above components condensed under slightly more reducing conditions than those in a gas of solar composition, in which case Tm would have been more volatile. If these inclusions were molten after condensation, the above two components may no longer be physically separable. No petrographic differences are apparent between these and normal coarse-grained inclusions.

Eu and Yb are the most volatile REE according to calculations and, in fine-grained inclusions, sometimes show large negative anomalies superimposed on otherwise flat REE patterns. Such "Group III" patterns (Conard 1976, Mason & Martin 1977) are apparently also found occasionally in coarse-grained inclusions, as are so-called "Group VI" patterns, which are flat except for positive Eu and Yb anomalies (Taylor & Mason 1978). Group III inclusions must have incorporated a REE component that stopped equilibrating with the gas at a temperature high enough to prevent complete condensation of Eu and Yb, but low enough that all the more refractory REE had condensed totally (Boynton 1978b). Not only does the REE component in Group VI inclusions appear to have equilibrated at a low enough temperature that all REE condensed totally into it, but it also contains excess Eu and Yb, presumably due to prior condensation of all the more refractory REE in some other nebular reservoir. Finally, although most Group II inclusions have negative Eu and Yb anomalies, indicating incomplete condensation of Eu and Yb into them, some, including coarse-grained inclusions A-2 (Conard 1976) and

CG-6 (Grossman & Ganapathy 1976b) have positive Eu and Yb anomalies superimposed on an otherwise normal Group II pattern. It must contain the same two REE components as other Group II inclusions, but at least one of them, or perhaps a third, contains excess Eu and Yb that probably originated in the same way as the excesses in Group VI inclusions. No petrographic descriptions of coarse-grained inclusions with Group III or Group VI patterns have been published. No exceptional mineralogical characteristics were noted by Gray et al (1973) for A-2 or by Grossman (1975a) for CG-6, indicating that there are no other obvious signs of the unusual history recorded by its REE pattern.

MINERAL ANALYSES The ion microprobe has been used to study trace elements in individual mineral phases. Although Hutcheon et al (1977) reported <50 ppb Li in major phases of Types A and B inclusions, Phinney & Whitehead (1978) found ~ 1 ppm in all major phases in Type B's. Thus, Li does not appear to be enriched in the inclusions relative to C1's and is probably not refractory. The element Be, however, shows substantial enrichments in melilite and anorthite in Phinney & Whitehead's (1978) inclusion and may thus be refractory.

Trace element analyses of mineral separates from Type B inclusions, particularly for REE, are the subject of several papers. Mason & Martin (1974) found that melilite from one inclusion has a positive Eu anomaly and steadily decreasing enrichments from La to Sm while fassaite from the same inclusion has a negative Eu anomaly and steadily increasing enrichments toward heavy REE. These complementary patterns were rationalized on crystallochemical grounds. They pointed out, as did Grossman & Ganapathy (1976b), that this is difficult to reconcile with the fact that bulk trace element contents seem to be independent of inclusion mineralogy and argued for a two-stage process—incorporation of REE into the inclusions during accretion of condensate components and redistribution of REE during crystallization after a later melting event. N. Onuma et al (1974) made the same observation and reached the same conclusion but proposed an alternative, that melilite and pyroxene partitioned REE between them during condensation in a closed system. This remains a viable hypothesis, as the cases of independence of trace elements on mineralogy documented by Grossman & Ganapathy (1976b) could be explained by nebular heterogeneity. Nagasawa et al (1976) determined melilite/liquid REE partition coefficients, combined them with data from other experiments on pyroxene/liquid partitioning to predict the equilibrium REE distribution between melilite and pyroxene, and found that the latter is in good agreement with data for mineral separates studied by Mason & Martin (1974), N. Onuma et al

(1974), and Nagasawa et al (1977). This verifies the conclusion of Mason & Martin (1974) and N. Onuma et al (1974) that REE are in the melilite and pyroxene crystal structures. The fact that this could be verified for mineral separates which undoubtedly contain Fremdlinge implies that the bulk of the REE do not reside in trace phases. Nagasawa et al (1976) concluded, as did Mason & Martin (1974), that melilite and pyroxene co-crystallized under closed-system conditions within the inclusion.

A hibonite-rich sample from a fluffy Type A was found to be rich in REE, with positive Eu and Lu anomalies (Davis et al 1978b). Impure mineral separates from compact Type A's (samples 1 and 2) were analyzed by Nagasawa et al (1977).

ISOTOPIC COMPOSITION

Three reviews of the isotopic compositions of Allende inclusions have appeared recently (R. N. Clayton 1978, Podosek 1978, and Lee 1979), so there is no need here to re-explore all ramifications of the data. Only the bearing of isotopic data on the origin of coarse-grained inclusions will be reviewed.

Normal Inclusions

OXYGEN R. N. Clayton et al (1973) discovered large oxygen isotopic anomalies in all types of Allende inclusions, including coarse-grained ones. On a diagram such as Figure 5, all samples produced from an isotopically homogeneous reservoir by mass-fractionation accompanying normal chemical processes should lie along a line whose slope is 0.5, such as the dashed line for terrestrial materials. Data points for inclusions, however, plot along the line *AD* whose slope is close to unity. The oxygen isotopic compositions of the inclusions are thus controlled predominantly by nuclear, rather than chemical, processes. R. N. Clayton et al (1973) suggested that *AD* is a mixing line between normal solar system oxygen at higher $\delta^{17}O$ and $\delta^{18}O$ than *D* and another oxygen component whose isotopic composition is vastly enriched in ^{16}O at lower $\delta^{17}O$ and $\delta^{18}O$ than *A*. Noting that astrophysical environments exist where ^{16}O can be nucleosynthesized in the absence of ^{17}O and ^{18}O, such as the explosive carbon-burning zone of a supernova, they further proposed that grains must have condensed in a region so close to the site of ^{16}O nucleosynthesis that matter produced in this process had not yet mixed significantly with oxygen from other nucleosynthetic processes. They suggested that such ^{16}O-rich grains must have found their way into the interstellar gas cloud that underwent gravitational collapse and fragmentation to form the solar nebula and that they survived this process, presumably because tempera-

tures were not high enough or were not high for long enough to completely evaporate them. The solar nebula was thus pictured as being composed of gas containing a homogenized mixture of oxygen isotopes produced in a number of nucleosynthetic environments (normal solar system oxygen) and previously formed dust grains whose oxygen was vastly enriched in ^{16}O relative to it. It was proposed that the major mineral phases of the inclusions condensed from this gas with nearly solar oxygen isotopic composition, but that when the inclusions accreted, they also incorporated a few per cent of their oxygen in the form of preexisting ^{16}O-rich grains. That different inclusions plot in different places along *AD* was explained by different proportions of new and old condensates in each. Later work on mineral separates (R. N. Clayton et al 1977), however, revealed inconsistencies in this model. Fassaite and spinel from Type B inclusions were found to plot close to *A*, with nearly a 5% excess of ^{16}O relative to terrestrial samples. This meant that, if the exotic oxygen

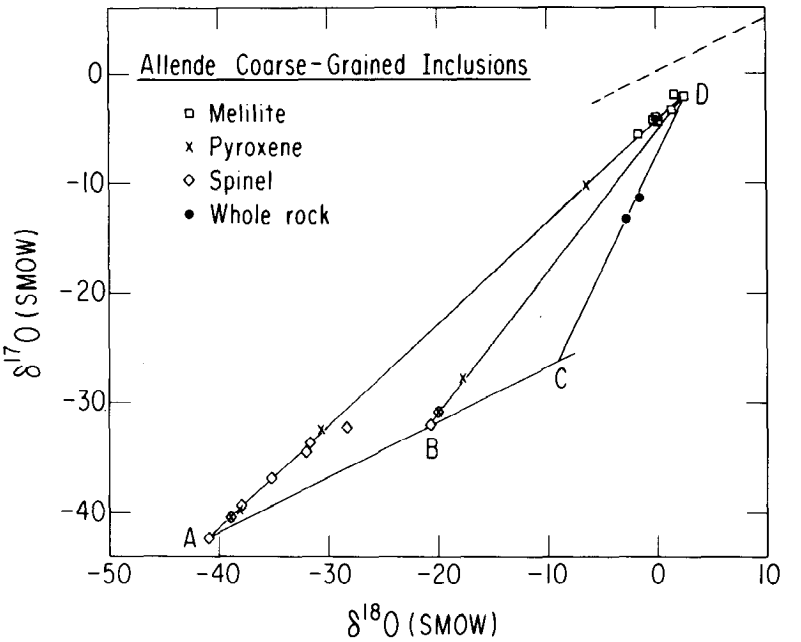

Figure 5 Variations in $^{17}O/^{16}O$ and $^{18}O/^{16}O$ ratios in Allende inclusions relative to the SMOW standard. Normal inclusions lie along *AD*, but FUN inclusions EK 1-4-1 and C1 plot along *BD* and *CD*, respectively. Melilite from normal inclusions and EK 1-4-1 always plots near *D* and fassaite and spinel near *A* and *B*. Dashed line for terrestrial samples and line *ABC* are mass-fractionation lines, having slopes of 0.5. From R. N. Clayton & Mayeda (1977).

component were pure ^{16}O, there would have to be about 5% interstellar grains in those mineral separates and even more if the exotic component had ^{17}O and ^{18}O in it. There simply were no candidate phases present at that concentration in the mineral separates, unless they were fractions of the fassaite and spinel themselves. This alone would not have been sufficient to dismiss the interstellar grain hypothesis, as Lattimer et al (1978) showed that most phases that condense from a gas of solar composition also condense from gases having even the extremes of composition that might be expected in many astrophysical environments. The real difficulty is that in every Type B inclusion investigated, the fassaites and spinels plot near *A* and the melilites near *D*. The interstellar grain hypothesis then requires the same ratios of exotic to nebular melilite, exotic to nebular fassaite, and exotic to nebular spinel in every inclusion and there seems to be no way to make the mixing process so precise.

The possibility remains that all crystals of fassaite and spinel in every inclusion have the same oxygen isotopic composition near *A* and all melilites have the same composition near *D*. It is extremely difficult, but not impossible, to reconcile this with an origin for Type B inclusions by crystallization from a melt. One possibility is that condensation occurred in a gas whose isotopic composition was near *A*, that melting took place during or after condensation, and that spinel and fassaite crystallized first upon cooling. The partially molten inclusions then found themselves immersed in a gas whose composition was close to *D*. The rate of oxygen exchange between gas and remaining liquid was rapid, so that melilite which later crystallized from the melt had more nearly normal oxygen isotopic composition. If fassaite and spinel exchanged their oxygen much more slowly with the melt, their much more ^{16}O-rich compositions could have been preserved. The required crystallization sequence is that observed by Seitz & Kushiro (1974) but, as already mentioned, those experiments may not be relevant to many Type B inclusions. Although spinel was undoubtedly the first-crystallizing phase in many of them, one of the inclusions analysed by R. N. Clayton et al (1977) was one in which MacPherson & Grossman (1979) suggested that melilite was first; yet, the oxygen isotopic distribution is the same in all of them. If all Type B inclusions were molten, it would thus have to be argued that the isotopic variations were produced after complete solidification (Blander & Fuchs 1975, Chou et al 1976, MacPherson & Grossman 1979). In such a model, all phases crystallized near *A* and attempted to exchange their oxygen with a gas near *D*. R. N. Clayton et al (1977) pointed out that, if oxygen diffusion is much more rapid in melilite than in fassaite and spinel, melilite would have approached *D* in composition, but fassaite and spinel would have been little changed from their initial composition. The presence of

alteration products and rims indicates that the inclusions did react with a gas phase after solidification of primary phases and before incorporation into the Allende parent body. Although it is not known whether alteration products or rims or both were analysed, the discovery by R. N. Clayton et al (1977) that a nepheline-, sodalite-, grossular-rich sample from near the rim of an inclusion plots right at *D* suggests very strongly that oxygen exchange and formation of alteration products and/or rims are related.

The alteration process would also work if the inclusions were never molten, but here there is an alternative, that fassaite and spinel condensed from a gas of isotopic composition *A* and melilite from another whose composition was close to *D*. In this model, condensation must have taken place more rapidly than homogenization of ^{16}O-rich gas with normal gas, which is also a requirement in the exchange model.

Melilite and spinel from compact Type A's have isotopic compositions similar to their counterparts in Type B's, but mineral separates from fluffy Type A's have not yet been analysed.

MAGNESIUM Whether excess ^{16}O entered the solar nebula in presolar grains or as gas, other elements should also show isotopic anomalies in Allende inclusions. Early work showed relatively small Mg isotopic variations (Gray & Compston 1974, Lee & Papanastassiou 1974) that were not due to mass-fractionation but that could not be unambiguously interpreted in terms of enrichment or depletion of specific isotopes, although Gray & Compston (1974) favored excess ^{26}Mg. Lee et al (1976) found clear evidence for excess ^{26} Mg in the coarse-grained inclusion studied by Gray & Compston (1974) and in other types of Allende inclusions. A study of mineral separates, each containing many crystals, showed that ^{26}Mg excesses are linearly correlated with ^{27}Al/^{24}Mg ratios of the major mineral phases in WA, a Type B inclusion (Lee et al 1977a). Spinel and fassaite, with very low ^{27}Al/^{24}Mg ratios, show virtually no excess ^{26}Mg. Melilite, with moderate ^{27}Al/^{24}Mg, has a 4‰ excess and anorthite, whose ^{27}Al/^{24}Mg ratio is over 200, has a 9% excess. The existence of a positive correlation between ^{26}Mg/^{24}Mg and ^{27}Al/^{24}Mg ratios fulfills a necessary condition for isochronism and is taken as clear evidence for in situ decay of ^{26}Al. One interpretation of the data is that the constituent phases crystallized from a homogeneous reservoir, either gaseous or liquid, of Mg and Al isotopes in which the initial ^{26}Al/^{27}Al ratio was $\sim 5 \times 10^{-5}$, the latter value being obtained from the slope of the correlation line. These results were confirmed by mass spectrometric measurements on directly loaded individual crystals of anorthite, melilite, and spinel (Lee et al 1977b) and by an ion microprobe study of anorthite (Bradley et al 1978) from the same inclusion. ^{26}Mg/^{24}Mg ratios of individual crystals

in polished sections of other Type B inclusions were measured by ion microprobe (Lorin et al 1977, Hutcheon et al 1978a,b). These data are in good agreement with those for WA in that they are correlated with $^{27}Al/^{24}Mg$ ratios along the same line, even within individual anorthite crystals (Bradley et al 1978, Hutcheon et al 1978b).

Taken together, the above studies show that ^{26}Al was present in the crystal structures of the major mineral phases of the inclusions and that it decayed in situ. As Esat et al (1979) cautioned, however, melilite from WA lies $\sim 1‰$ off the best fit line through the other WA data points. D. D. Clayton (1977a) used this observation as an argument in favor of his proposal that Allende inclusions are actually sintered mixtures of micron-sized presolar grains in which ^{26}Al had long since decayed away completely. He argued that the gross correlation between ^{26}Mg excesses and $^{27}Al/^{24}Mg$ ratios exists because the ^{26}Mg excesses were originally present in Al-rich interstellar phases and ^{26}Mg was kinetically hindered from diffusing far from the ^{27}Al originally associated with it during recrystallization into the mm-sized phases now observed. In this view, the correlation is merely a mixing line between phases that formed with large ratios of $^{27}Al/^{24}Mg$ and, as a result, of $^{26}Mg/^{24}Mg$ as well and phases with low ratios of $^{27}Al/^{24}Mg$ and $^{26}Mg/^{24}Mg$. The lack of collinearity of WA data points is taken as evidence for only slight spatial segregation of Mg from Al in this model. One of the major problems with this explanation, however, is that it ignores the fact that, if the presolar grains model is correct, major spatial segregation of Mg from Al *has* occurred in the process of making mm-sized grains of chemically distinct phases from the material required by D. D. Clayton (1977a) to be homogeneous in composition on a $10\,\mu m$ scale originally. An alternative, and far more probable, explanation for the lack of *precise* isochronism suggested by Esat et al (1979) is late disturbance of the Al-Mg system by an isotopic exchange process similar to that which affected oxygen in melilite.

Because of the short half-life of ^{26}Al (7.2×10^5 yr) and the fact that this isotope was most likely produced in the carbon-burning zone of a supernova, the previous existence of ^{26}Al in the Allende inclusions implies that such an explosion occurred at most a few million years prior to their formation and in the vicinity of the birthplace of the solar system. This led to the suggestion by Cameron & Truran (1977) that a nearby supernova explosion triggered the collapse of the interstellar cloud to form the solar nebula. An alternative suggestion, that ^{26}Al was produced locally in reactions involving energetic protons emitted from an early active sun, has been investigated by Heymann & Dziczkaniec (1976), D. D. Clayton et al (1977), and Lee (1978). Such models do not appear plausible, however, because isotope anomalies that have not been observed should have

also been produced in other elements and because a mechanism is unknown for production of the required high fluences of protons which, collectively, must have carried off a substantial fraction of the sun's gravitational binding energy.

R. N. Clayton et al (1973) originally proposed that interstellar grains were the carriers of ^{16}O because of the relative ease with which isotopically different gases could mix and erase spatial variations in isotopic composition. If, however, the supernova trigger is correct and if the ^{16}O comes from the same supernova, the time scale for condensation could easily have been shorter than that for gas mixing. Thus, it is not clear that gas is ruled out as the ^{16}O carrier, although Lattimer et al (1978) suggested reasons why grains are more likely to be responsible and computed condensation sequences for zones of different composition in expanding supernova ejecta. Even if grains were the original carriers, they may have evaporated locally in the contracting nebula, creating isotopically anomalous gas pockets in which rapid condensation may have occurred.

Ion probe studies of inclusions other than WA also show deviations from the normal Al-Mg isochron. Lorin & Christophe Michel-Lévy (1978a) found hibonite grains in what appear to be Type A inclusions from both Leoville and Allende that plot well above the normal isochron upon which fall data points for other minerals in the same inclusions. Similarly, hibonite in the fluffy Type A studied by Steele & Hutcheon (1979) plots above an isochron through data for melilite, spinel, and other hibonites which has a lower slope than usual. Such results were attributed to early formation of hibonite and other phases and exchange of Mg isotopes in melilite and anorthite during later reactions in which oxygen isotopes were also exchanged (Lorin & Christophe Michel-Lévy 1978b). This appears to be in conflict, however, with the observations of Hutcheon et al (1979) who found petrographic evidence that hibonite was more altered than melilite in another fluffy Type A, some of whose hibonite plots above the straight line through primary melilite and other hibonite points. Other explanations are possible. If Lorin & Christophe Michel-Lévy's (1978a,b) inclusions are fluffy Type A's, they may be mechanical mixtures of phases whose Al and Mg isotopic compositions were never homogenized with one another. Inferred differences in condensation age would be $>10^6$ years, too great to interpret the inferred differences in initial $^{26}Al/^{27}Al$ in this way. Instead, perhaps they record Al isotopic heterogeneities in the gas cloud from which these phases condensed.

No traces of radiogenic ^{26}Mg were found by Lorin et al (1978) in their study of the petrographically unusual ophitic inclusions. Anorthite in the core of one was very enriched in heavy Mg isotopes by a mass-fractionation process, while spinel and fassaite in the core and anorthite

near the edge were not. It is difficult to reconcile these internal Mg isotopic variations with a melting event, which is the favored explanation for the textures, unless isotopic compositions were modified later, by preferential exchange. Oxygen isotopic measurements of mineral separates from such inclusions might be revealing. Why the Mg isotopes are different in these inclusions is unknown, although their bulk compositions must be different from those of normal inclusions, another indication of a different history.

OTHER ELEMENTS Epstein & Yeh (1977) and Yeh & Epstein (1978) found Si isotopic variations in Allende inclusions that appear to be due to mass-fractionation. R. N. Clayton et al (1978) showed that this is true for both Type A and Type B inclusions and that the magnitude and sign of the variations are what would be expected for fractionation during high-temperature condensation of silicates from a gas whose $\delta^{30}Si = 0$. Patchett (1979) showed that mass-fractionation processes have enriched both coarse- and fine-grained inclusions in the light isotopes of Sr. These could be chemical processes specific to the condensation of Sr or characteristic of additional trace elements that entered the inclusions with Sr, such as Eu in the coarse-grained ones (Grossman et al 1977). Tatsumoto & Shimamura (1979) reported variations in the $^{238}U/^{235}U$ ratio in Type B inclusions which, because of their size and the mass numbers involved, are probably due to nuclear effects. Heydegger et al (1979) reported very small nuclear effects in the $^{50}Ti/^{49}Ti$ ratio in fassaites from Type B inclusions.

If incompletely homogenized supernova debris is the source of the ^{16}O excesses, then nuclear isotope effects should be present in many other elements in these samples. A major problem thus arises in that, aside from the aforementioned possibilities of nuclear effects in U and Ti, diligent searches for such effects in Ba (McCulloch & Wasserburg 1978a), Ca (Lee et al 1978), K (Begemann & Stegmann 1976, Birck et al 1977, Stegmann & Begemann 1979), and Si and Mg (above papers) in normal inclusions have been unsuccessful. This is a particularly chronic problem for models in which the inclusions are pictured as sintered aggregates of interstellar grains (D. D. Clayton 1977a) or volatilization residues of such material (Chou et al 1976, Notsu et al 1978, Hashimoto et al 1979).

FUN Inclusions

The story is different, however, in three other inclusions. Samples from two Type B's, C1 and EK 1-4-1, are the only ones, of a very large number of Allende inclusions whose oxygen isotopic compositions have been measured, which do not plot along *AD* in Figure 5 (R. N. Clayton et al

1977, R. N. Clayton & Mayeda 1977), but rather along *CD* and *BD*, respectively. Although pure mineral separates were not analysed in the case of C1, the familiar pattern in which melilite plots near *D* and spinel and fassaite at much lower $\delta^{17}O$ values, in this case near *B*, is observed for EK 1-4-1. R. N. Clayton & Mayeda (1977) suggested that the original bulk isotopic compositions of EK 1-4-1 and C1 were at *B* and *C*, respectively, lying along a mass-fractionation line *ABC* with the original bulk composition of usual Allende inclusions. They called upon a kinetic effect in the gas phase prior to condensation of these two inclusions to enrich them in the heavy oxygen isotopes relative to other inclusions and upon the same exchange process that modified the isotopic composition of melilite in the latter to do so in these inclusions also. It is also possible to replace the mass-fractionation step with a mixing process in which a second isotopically anomalous nucleosynthetic component lying beyond *C* is added to the inclusions (Lee et al 1978). This is considered unlikely, however, as the mixing line so generated would fortuitously have to have the slope of a mass-fractionation line, 0.5.

C1 (Lee et al 1976) and EK 1-4-1 (Wasserburg et al 1977) have also undergone large mass-dependent enrichments in their heavy Mg isotopes. The ratio of the Mg effect in C1 to that in EK 1-4-1 is the same as the inferred ratio of their original oxygen effects, but the latter are about one-half as great. As pointed out by R. N. Clayton & Mayeda (1977), coexistence of mass-fractionated oxygen and Mg is expected from their model, though the relative magnitudes may be a problem. Within each inclusion, all phases have the same Mg isotopic composition (Wasserburg et al 1977), except for C1 anorthite (Esat et al 1978). Evidently, in contrast to oxygen, later isotopic exchange with an external reservoir introduced insignificant Mg relative to that originally present, except for anorthite. Si is much more severely mass-fractionated toward the heavy isotopes in EK 1-4-1 (Yeh & Epstein 1978) and C1 (R. N. Clayton et al 1978) than in normal inclusions, C1 again being more fractionated than EK 1-4-1. In contrast, very small Ca mass-fractionations, opposite in sign to those for Mg, O, and Si, were found in these inclusions (Lee et al 1978).

These so-called "FUN" inclusions are remarkable in another way: they are the only ones to show clear-cut nuclear anomalies in elements other than oxygen: Ca (Lee et al 1978), Nd in EK 1-4-1 only and Ba (McCulloch & Wasserburg 1978a), Sm (Lugmair et al 1978, McCulloch & Wasserburg 1978b), Sr (Papanastassiou & Wasserburg 1978), Si (R. N. Clayton et al 1978, Yeh & Epstein 1978), and possibly Mg (Wasserburg et al 1977). Furthermore, in surprising contrast to oxygen, for each of these inclusions, the isotopic compositions of these elements are the same within analytical

uncertainty in all mineral separates and splits analysed, except for Mg in C1 anorthite.

Throughout this section, an alternative working hypothesis has been entertained that the inclusions were never molten and different phases within them obtained their different isotopic compositions by condensing in isotopically distinct locales. In the case of FUN inclusions, this model would have fassaite and spinel condense from a gas phase related to that which spawned their counterparts in other inclusions by a mass-fractionation process. The melilites in FUN inclusions would have to condense from a separate reservoir of more nearly normal oxygen isotopes, just as the model requires for other inclusions. These cannot be the same melilites as are in the usual inclusions, however, because they have the same nuclear anomalies in other elements as the fassaites and spinels in FUN inclusions. The hypothesis thus requires two isotopically distinct oxygen reservoirs within each of three reservoirs (one for usual inclusions, one for EK 1-4-1, and one for C1), which are isotopically different in other elements. This logical extension of the alternative hypothesis seems very improbable and implies that the hypothesis may have to be dropped as a possible explanation for even the usual Allende inclusions.

A third isotopically unusual inclusion is known, HAL. So far, only its Ca and Mg isotopic compositions have been reported (Lee et al 1979). Mass-dependent heavy Ca isotope enrichment has occurred and small nuclear effects are also present. If ^{26}Al was once present in HAL, its abundance was, as in C1 (Esat et al 1978), far less than in usual Allende inclusions. Esat et al (1979) reported that another inclusion, Egg-3, may be of the FUN-type, exhibiting mass-fractionated Mg, but also ^{26}Mg excesses.

One of the most fascinating things about C1 is that its gross mineralogical composition and textural characteristics are just like those of isotopically normal Type B inclusions (Gray et al 1973), indicating that the same physico-chemical processes leading to inclusion formation occurred in the isotopically distinct regions in which C1 and normal inclusions originated. The same can probably be said for EK 1-4-1, although here the state of the remaining sample makes detailed petrographic comparison difficult. HAL, however, is not only isotopically different from C1 and EK 1-4-1, but it is also mineralogically exotic (Allen et al 1980), classifiable neither as Type A nor Type B. HAL's mineralogical and isotopic differences stem either from alteration after condensation from an isotopically normal reservoir (Lee et al 1979) or from differences in physico-chemical conditions during condensation from an isotopically

distinct region (Allen et al 1980). REE patterns of Cl (Conard 1976) and HAL (Tanaka et al 1979) have pronounced negative Ce anomalies. These can only be produced during condensation if the gas phase is extremely oxygen-rich, such as in supernova ejecta from which H has previously been burned out (Boynton 1978c, Tanaka et al 1979). Calculations show that Ce becomes very volatile under these conditions, a prediction apparently confirmed by volatilization experiments in oxidizing gases (Nagasawa & Onuma 1979). REE in these two inclusions condensed in a chemically distinct environment from those in all other inclusions.

Lee (1979) showed that the simplest interpretation of nuclear effects in Ba, Nd, and Sm in FUN inclusions is that Cl incorporated an excess of nuclides synthesized in the *p*-process and EK 1-4-1 has excesses of both *p*- and *r*-process nuclides. Although it is possible that both nucleosynthetic processes can take place in different zones of the same supernova, the distribution of nuclear effects in Cl and EK 1-4-1 shows that matter from each of these processes was carried in a separate component. It is not known whether the carriers of *p*- and *r*-process–enriched heavy elements were solid or gaseous prior to mixing with mass-fractionated low-Z elements. Nor is it understood why nuclear anomalies in heavy elements occur only in inclusions with mass-fractionated low-Z elements. One attempt at explaining this (D. D. Clayton 1980) reinterprets the line *ABC* in Figure 5 as being due to mixing of isotopically anomalous interstellar dust with light-isotope enriched gas sputtered from it, but fails to explain many details of the observations, such as the very tight adherence of many inclusions to the line *AD*.

AGE INFORMATION

Information about the age of the inclusions has been obtained from the U-Pb, Rb-Sr, and K-Ar systems and from decay products of extinct nuclides.

Rb-Sr System

Gray et al (1973), Nyquist et al (1973), Wetherill et al (1973), Nagasawa & Jahn (1976), and Tatsumoto et al (1976) observed extremely low $^{87}Sr/^{86}Sr$ ratios in inclusions with very low Rb/Sr ratios. The lowest $^{87}Sr/^{86}Sr$ ratio measured by Gray et al (1973), 0.69877, is lower than all other solar system materials and identifies the inclusions as the oldest known solids in the solar system. Data from a variety of materials in Allende (Gray et al 1973, Tatsumoto et al 1976) and from mineral separates from individual inclusions (Nagasawa & Jahn 1976) fail to define isochrons,

leading to the conclusion that element redistribution has occurred, perhaps recently. The conclusions of these workers are unaffected by the findings of Patchett (1979), since Sr isotopic compositions were corrected for mass-fractionation in the Rb-Sr studies.

U-Th-Pb System

Pb isotopic compositions of a variety of materials separated from Allende define straight lines on $^{207}Pb/^{204}Pb$–$^{206}Pb/^{204}Pb$ diagrams, yielding model ages of 4.57×10^9 yr (Chen & Tilton 1976) and 4.55×10^9 yr (Tatsumoto et al 1976). Furthermore, the data define chords on concordia diagrams, suggesting initial ages of 4.57×10^9 and 4.55×10^9 yr and more recent disturbance at 0.28×10^9 yr and 0.11×10^9 yr, respectively. Although these results could be affected by U isotopic variations, the $^{238}U/^{235}U$ ratio is near normal in one of Tatsumoto et al's (1976) inclusions that departs significantly from their $^{207}Pb/^{206}Pb$ isochron (Tatsumoto & Shimamura 1979). In an ion microprobe study of Th-, U-rich micro-phases in Type B inclusions, Lovering et al (1976) detected no ^{204}Pb and used observed $^{207}Pb/^{206}Pb$ ratios to obtain an average age of 4.60 and a range of 4.53–4.65×10^9 yr for six different grains, presumably by assuming the terrestrial $^{238}U/^{235}U$ ratio. On some of the same grains and others, Wark & Sewell (1979) calculated a mean age of 4.56 and a range of 4.32–4.84×10^9 yr simply from the ratio of U to Th to Pb, assuming no common Pb and the terrestrial U isotopic composition. Perhaps some of the scatter in these ages is due to the U isotopic variations known to be present in bulk Type B inclusions (Tatsumoto & Shimamura 1979).

K-Ar System

Although one olivine-bearing coarse-grained inclusion is known to have an $^{40}Ar/^{39}Ar$ age equal to the canonical age of the solar system (Dominik et al 1978), three others have apparent ages greater than 4.6×10^9 yr, one studied by Jessberger et al (1977), 4.9×10^9 yr, and two by Jessberger & Dominik (1979), 4.90 and 4.98×10^9 yr. As the technique assumes the samples have normal $^{39}K/^{40}K$ ratios, one possible explanation for high ages is that the inclusions contain isotopically anomalous K. Stegmann & Begemann (1979) found, however, that the K isotopic composition in one of Jessberger & Dominik's (1979) inclusions with a high age was indistinguishable from that of terrestrial K. The reason for the high ages is unknown. One possibility, suggested by D. D. Clayton (1977b), is that the inclusions contain ^{40}Ar produced by decay of interstellar ^{40}K prior to formation of the solar system. Why presolar ^{40}Ar was not degassed during sintering and fusion of the interstellar grains to form the inclusions

is, however, a severe difficulty for this model. D. D. Clayton (1975) also proposed that ^{41}K excesses, from decay of now-extinct ^{41}Ca, should be found in materials derived from presolar grains and suggested that his model of forming inclusions by partial fusion of presolar condensates would be in great difficulty if ^{41}K excesses are not observed in Allende inclusions (D. D. Clayton 1977b). The latter appears to be the case, given the results of Begemann & Stegmann (1976), Birck et al (1977), and Stegmann & Begemann (1979).

Extinct Nuclides

In addition to ^{26}Al, other extinct nuclides have left their decay products in the inclusions. ^{136}Xe from fission of ^{244}Pu was measured in a Type B inclusion by Drozd et al (1977), in a melilite separate by Marti et al (1977), and in EK 1-4-1 by Papanastassiou et al (1978). The ratio of ^{244}Pu to other refractories is similar to that in other meteorites. The presence of fission tracks (Shirck 1974, Drozd et al 1977, Podosek et al 1977) shows that ^{244}Pu decayed after the major mineral phases formed. Scheinin et al (1976) and Lugmair et al (1978) claimed to have found a small excess of ^{142}Nd in fassaite from a Type B and attributed it to decay of extinct ^{146}Sm. Excess ^{129}Xe from ^{129}I decay was found by Scheinin et al (1976) in the same inclusion. Papanastassiou et al (1978) reported excess ^{129}Xe in EK 1-4-1 and noted that the inferred initial ^{129}I/^{127}I ratio is approximately that of other chondrites. Because I is probably in Cl-bearing sodalite, an alteration product, the excess ^{129}Xe implies that the alteration process occurred very shortly after inclusion formation.

CONCLUSION

Before the fall of Allende, the prevailing point of view was that the solar nebula was a well-stirred, homogeneous, quiescent gas cloud, hot enough in its center that preexisting interstellar dust grains were completely evaporated, with condensation occurring later, during a period of slow, monotonic cooling. A mere decade of research on Allende has changed this picture so dramatically that it will never be the same. In coarse-grained inclusions alone, only 5% of the meteorite, we have uncovered remarkably well-preserved evidence for a supernova explosion that occurred just before condensation, for incompletely homogenized material from several nucleosynthetic sources, and for solar nebular regions of different chemical and isotopic composition. It is doubtful that we would have come this far in such a short time had it not been for the large size of Allende, for major technological advances in analytical techniques and instrumentation, and for the excitement about and intense

interest in the problem of solar system origin shared by an international group of scientists with diverse approaches to the subject.

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