PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A
MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

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Phil. Trans. R. Soc. Lond. A 1987 **323**, 287-304
doi: 10.1098/rsta.1987.0086

Phil. Trans. R. Soc. Lond. A **323**, 287–304 (1987)
Printed in Great Britain

Local and exotic components of primitive meteorites, and their origin

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Most of the material of chondrites was heavily reprocessed in the early Solar System, and hence retains only a dim memory of its interstellar origin even in the least altered meteorites. Opaque matrix, the most primitive material, seems to have taken up Fe^{2+} and changed its mineralogy and texture. Chondrules and Ca, Al-rich inclusions have been further altered by melting, oxidation or reduction, loss of volatiles, etc. None the less, small amounts of exotic components have survived, as indicated by isotopic anomalies. A dust component enriched in ^{16}O is the most abundant and widespread. It survives in Ca, Al-rich inclusions as anomalous spinel grains, but is recognizable even in highly evolved meteorites and planets from variations in bulk oxygen isotopic composition. A few inclusions show small nucleosynthetic anomalies for many elements (Si, Ca, Ti, Cr, Sr, Ba, Nd, Sm), always coupled with mass fractionation of several of these elements, as well as O and Mg. Seven extinct radionuclides (^{26}Al, ^{41}Ca, ^{53}Mn, ^{107}Pd, ^{129}I, ^{146}Sm, and ^{244}Pu) have been recognized from their decay products, and provide clues to the chronology and nucleosynthetic sources of the early Solar System.

Highly volatile elements, such as C, N and the noble gases, show especially large and numerous isotopic anomalies. The noble-gas components include Ne-E (monoisotopic ^{22}Ne from the β^+ decay of 2.6 a ^{22}Na), Xe-HL (enriched 2-fold in the light and heavy isotopes), and Xe-S (enriched in the even-numbered, middle isotopes 128, 130 and 132). They are located in carriers that themselves are anomalous, e.g. carbon enriched up to 2.4-fold in ^{13}C or depleted by more than 30% in ^{15}N, or spinel enriched in ^{16}O and ^{13}C. Other anomalies include nitrogen enriched 2-fold in ^{15}N and hydrogen enriched 5-fold in D; probably relict interstellar molecules. A variety of astronomical sources seem to be required: novae, red giants, supernovae and molecular clouds.

1. INTRODUCTION

All meteoritic matter originally came from outside the Solar System and thus is exotic in the broadest sense of the term. However, much of this material was reprocessed in the early Solar System (by vaporization, melting, mixing, isotopic exchange, chemical reactions, etc.), and became isotopically homogeneous. Such commonplace material is properly called 'local', leaving the term 'exotic' or 'presolar' for material that still retains an anomalous isotopic signature. Exotic material sometimes occurs as discrete, potentially separable grains, but often has been assimilated by large amounts of local matter, and is recognizable only by its anomalous isotopic composition.

Qualitatively, a similar picture may be expected for comets. But as comets formed in more distant, cooler parts of the Solar System, reprocessing may have been less intensive and pervasive. Thus the local material may be less processed and less dominant, with the exotic material more abundant and diverse. None the less, primitive meteorites are a good frame of reference for interpretation of cometary data. I shall therefore first review the properties and classification of chondrites (§2) and then their local and exotic components (§§3 and 4).

2. PROPERTIES AND CLASSIFICATION OF CHONDRITES

Chondrites are stony meteorites containing chondrules, millimetre-sized silicate spherules that appear to be frozen droplets of a melt. They consist largely of *olivine* [$(Mg,Fe)_2SiO_4$], *orthopyroxene* [$(Mg,Fe)SiO_3$], and *plagioclase feldspar* [solid solution of $CaAl_2Si_2O_8$ and $NaAlSi_3O_8$]. In the more primitive chondrites, glass occurs in place of crystalline feldspar.

Chondrules are embedded in a ground-mass or *matrix*. In the less primitive chondrites, the matrix is somewhat finer-grained than the chondrules, but otherwise has the same mineralogy and composition. Millimetre-sized grains of *metal* (nickel-iron with 5–60% Ni) and *troilite* (FeS) are also present. In the more primitive chondrites, at least part of the matrix is very fine-grained (to *ca.* 10^{-6} cm) and richer in Fe^{2+} than are the chondrules.

Five chondrite classes are recognized, differing in the proportions of oxidized to reduced iron. *Enstatite* (E) chondrites are highly reduced, containing iron only as metal and FeS. *Carbonaceous* (C) chondrites are highly oxidized, containing mainly Fe^{2+}, Fe^{3+}, and little or no Fe^0. The middle ground is occupied by three classes of intermediate oxidation state and variable iron content: H, L, and LL chondrites. (The letters refer to total iron content: high, low and low-low.) Collectively, they are often called *ordinary* (O) chondrites.

Van Schmus & Wood (1967) have further subdivided each of these classes into '*petrologic types*', numbered from 1 to 6. These types were originally designed to reflect increasing chondrule-to-matrix intergrowth (probably due to thermal metamorphism in the meteorite parent bodies, at T up to 1100 °C), but have turned out to correlate well with compositional trends, e.g. volatile content. Probably these types represent an accretion time sequence, the higher types having accreted earlier, and therefore being located in deeper and warmer locations in the parent body.

Only the most primitive chondrites, up to petrologic type 3, are of interest to us, as they are least affected by metamorphism and hence have best preserved their exotic components as well as a record of processes in the solar nebula. Regrettably, even these meteorites are not pristine. Types 1 and 2 – found only among C-chondrites – have undergone hydrothermal alteration in their parent bodies (DuFresne & Anders 1962; R. N. Clayton & Mayeda 1984). Type 3, in turn, has been slightly metamorphosed, at T up to *ca.* 300–400 °C. Fortunately the least metamorphosed type 3s can be recognized from their low thermoluminescence sensitivity and other criteria, and on this basis, type 3s have been divided into 10 subclasses, 3.0 to 3.9 (Sears *et al.* 1980; Anders & Zadnik, 1985).

C3 chondrites contain a minor but very important textural component: millimetre-sized, often irregular *inclusions* of refractory, Ca, Al-rich but Si-poor minerals such as melilite, $Ca_2(Al_2,MgSi)SiO_7$ or *spinel*, $MgAl_2O_4$. They are known by the acronym CAI.

3. 'LOCAL' COMPONENTS OF CHONDRITES AND THEIR ORIGINS

To a first approximation, chondrites are condensates from the solar nebula, as first recognized by Wood (1958). Thus a convenient if oversimplified framework for interpreting chondrites is the equilibrium condensation sequence of a solar gas (Larimer 1967; Grossman 1972; Lewis 1972; Grossman & Larimer 1974). Figure 1 shows this sequence. A more detailed version of the 300–1800 K portion is given in Figure 2. As chemical equilibrium is independent of the path, these diagrams apply equally well to isobaric heating or cooling and to isothermal compression.

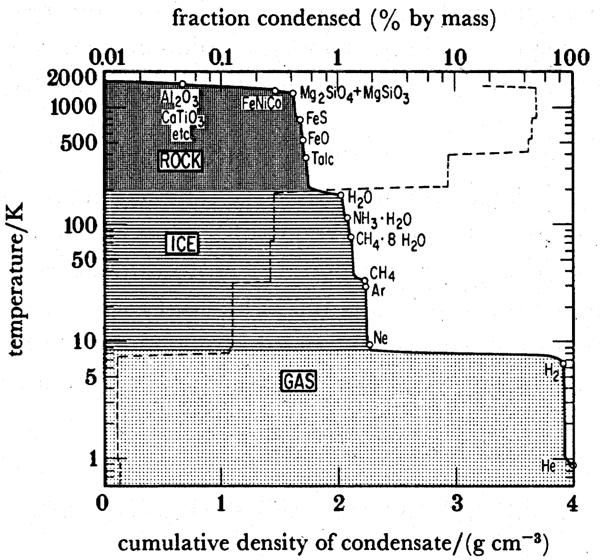

FIGURE 1. Condensation of solar gas at 10^{-4} atm† (Lewis 1972). Equilibrium condensation of a solar gas explains the composition of planets, at least to first order. Inner planets consist of rock, comets and satellites of outer planets consist of rock and ice, and the outer planets consist of all three.

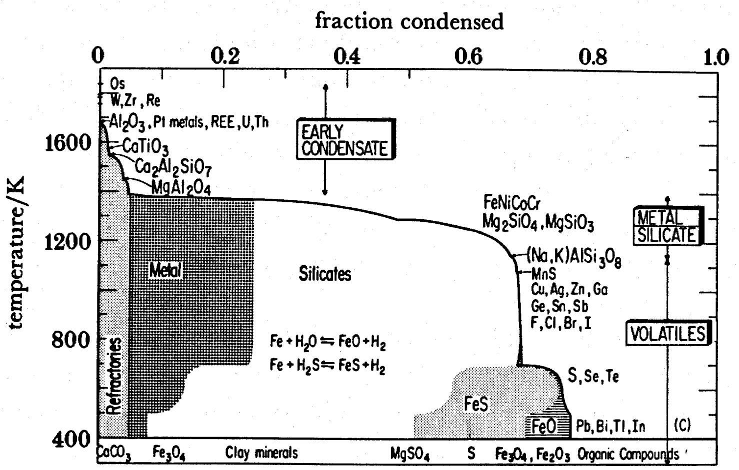

FIGURE 2. 'Rock' portion of the diagram in figure 1, on linear scales Below *ca.* 700 K, metal begins to react with H_2O and H_2S in the nebula, yielding FeO (which enters silicates) and FeS. The mineral assemblage at 400–500 K resembles that of ordinary chondrites. Below 400 K, many major changes take place, yielding a mineral assemblage resembling that of carbonaceous chondrites.

Figure 1 helps rationalize the composition of planets and other small bodies in the solar system. The inner planets and the asteroids consist only of *rock*; comets and the satellites of the outer planets consist of *rock and ice*, whereas the outer planets consist of *rock*, *ice*, *and gas*.

This appealing picture is oversimplified in several respects. (1) The various chondrite classes differ somewhat in bulk composition (Fe/Si, Mg/Si, Ca/Si, etc.), and so at best only one class can be a *total* condensate. All others must have been fractionated, presumably by gain or loss of major condensates (Larimer & Anders 1970). (2) The nebula was not wholly gaseous; some presolar solids survived (R. N. Clayton *et al.* 1973, see also §4). (3) Chemical equilibrium may not have been reached, especially at lower temperatures. (4) The composition may have

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† 1 atm = 101 325 Pa.

deviated from solar, owing to gas-dust fractionations, etc. None the less, the condensation diagram is a good frame of reference, yielding detailed predictions that can be compared with observations. I shall review the principal components of chondrites against this framework.

(*a*) *Matrix*

Only a few meteorites of petrologic types 3.0–3.4 have retained pristine matrix, unaffected by metamorphism or hydrothermal alteration (Wood 1962; Huss *et al.* 1981; Nagahara 1984). This opaque (or 'Huss') matrix contains micrometre- to submicrometre-silicate grains, amorphous material and submicrometre-sized grains of Fe-Ni, FeS, and Fe_3O_4. The Fe_3O_4 and the high Fe^{2+}-content of the silicates (the olivine has up to 50 mol % Fe_2SiO_4), are consistent with equilibrium in a solar gas at not more than about 400 K (Grossman 1972):

$$3Fe + 4H_2O \leftrightharpoons Fe_3O_4 + 4H_2, \tag{1}$$

$$2MgSiO_3 + 2Fe + 2H_2O \leftrightharpoons Fe_2SiO_4 + Mg_2SiO_4 + 2H_2. \tag{2}$$

In a solar gas, reaction (1) goes to the right below 400 K, whereas reaction (2) reaches 50 mol % Fe_2SiO_4 at 470 ± 40 K.

Was the matrix totally reprocessed in the solar nebula or has it retained some of its exotic features? At least some of the minor components of matrix must be exotic, because they are isotopically anomalous (§4*d*). However, we know embarrassingly little about the bulk of opaque matrix, as it has been recognized only recently and has not received much study; in contrast to the more common but less primitive types of matrix (Ashworth 1977; Hutchison *et al.* 1979). One feature that suggests major reprocessing in the solar nebula is the high Fe^{2+} and Fe_3O_4 content. Reactions (1) and (2) are heterogeneous, and are barely fast enough at the high temperatures (400–500 K) and densities [$n(H_2) \approx 10^{14}\ cm^{-3}$]. The required combination of moderate T and large $n(H_2)$ is not available in any known astrophysical environment other than the solar nebula.†

Actually, reactions (1) and (2) could proceed at higher temperatures if the H_2O/H_2 ratio were greater than solar. The easiest way to raise this ratio would be by a dust:gas fractionation, because dust is a major carrier of oxygen (Larimer & Bartholomay 1983; Wood 1984). As dust would tend to settle toward the nebular midplane where accretion takes place, some such fractionation is expected. We shall return to this problem in §3*b*.

Granted that Fe^{2+} and Fe_3O_4 contents of matrix were established in the solar nebula, it does not follow that all other features were, too. Very little work has been done on matrix; in particular, no systematic search has been made for surviving interstellar features. Several isotopically anomalous, hence presolar components have been found in chondrites [C, H, N and noble gases; §4*d*] and though none of them have been seen *in situ* – or otherwise conclusively linked to matrix – their presence shows that at least some heat-sensitive presolar components got into meteorites without serious damage. Thus it is quite conceivable that matrix of primitive meteorites is only lightly reprocessed interstellar dust (Wood 1973, 1981; Anders 1965; Cameron 1973; D. D. Clayton 1982; Huss 1987). This is a testable proposition that should be checked experimentally.

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† There is a minor loophole in this argument. If the iron in interstellar grains had been very finely dispersed (monolayer or individual atoms) at some stage of its history, then it may have become oxidized before its arrival in the Solar System. However, such ultrafine material would react with the nebular gases during its reorganization to larger crystals, and so its final oxidation state would still be established in the nebula.

(*b*) *Chondrules*

Chondrules formed by flash melting of pre-existing solids, not by direct condensation from gas (Nagahara 1981; Wood 1984). The melting time was short (seconds to minutes), judging from the failure of three reactions to go to completion: loss of Na and other volatiles (Schmitt *et al.* 1965; Tsuchiyama *et al.* 1981), reduction of Fe^{2+} to Fe by nebular hydrogen (Wood 1984), and isotopic exchange with oxygen of the nebula (R. N. Clayton 1981). The cooling time likewise was short (seconds to hours, depending on textural type), as shown by laboratory experiments (Tsuchiyama *et al.* 1980; Planner & Keil 1982; Nagahara 1983).

The short cooling times imply that the chondrule-forming region was *small*, allowing rapid heat loss by radiation. The existence of five major chondrite classes requires at least five separate domains, differing in oxidation state, bulk chemistry and oxygen isotope composition.

Chondrules are complementary to other major components of the meteorite (metal, *total* matrix), being depleted in siderophiles and volatiles but enriched in refractories (Grossman & Wasson 1983, 1985; Wood 1984). Interestingly, the bulk chondrite always is close to solar composition, which suggests that these components are cogenetic and accreted *comprehensively*. The parent material may have been the opaque matrix, perhaps after a preliminary stage of reprocessing (Rambaldi & Wasson 1984). It is important to distinguish this primitive, opaque matrix from more fractionated and recrystallized types, which probably represent 'failed chondrules': material that overshot or undershot the melting range (Wood 1984).

Chondrule formation seems to have involved some recycling, as demonstrated by relict crystals (Nagahara 1981; Rambaldi *et al.* 1983) and compound chondrules (Gooding & Keil 1980). It was followed by accretion of rims of matrix-like material (Allen *et al.* 1980).

It seems that chondrule formation was a pervasive process that occurred throughout the inner solar system and reprocessed some 50–90% of the material. The inner planets are depleted in some of the same elements as the chondrites (e.g. alkalis, S, Se, Ga, Ge, etc.) and by Occam's Razor, it seems reasonable to invoke the same process (Morgan & Anders 1980).

The mechanism of chondrule formation has not yet been conclusively identified. The complementarity of chondrules and matrix speaks against models that make chondrules and matrix in different places and mix them afterwards. The most promising model involves heating of interstellar matter as it falls into the solar nebula. Direct 'infall heating' by aerodynamic drag (Wood 1984, 1985) turns out to be quantitatively inadequate, but heating by viscous interaction with the nebula seems more promising (Wood 1986). The chondrule-forming zone in this model is a thin, hot layer of the nebula, as implied by the short cooling times of chondrules. However, this model is very recent, and still has to pass many quantitative tests.

The redox state (i.e. Fe^{2+} content of olivine) of primitive chondrules is quite variable even within a single chondrule, ranging from less than 1–40 at.%. At 1500 K in a solar gas, the equilibrium value would be only 0.001%, and so either the gas was non-solar or the parent material was highly oxidized and did not fully equilibrate with the ambient gas upon melting. Wood (1984), who favours the former alternative, has shown that Fe^{2+} contents of 0.5–30 at.% could be attained if dust were enriched 10^2 to 10^4-fold relative to gas, thus raising the H_2O/H_2 ratio in the system. However, it seems unlikely that chondrules fully equilibrated with the nebula, as they did not reach uniform Fe^{2+} contents, did not lose all their volatiles, and did not isotopically equilibrate their oxygen. It is more likely that chondrules formed from an Fe^{2+}-rich precursor (opaque matrix or a reprocessed derivative thereof), which was *partially* reduced and devolatilized on melting.

Even for the highly reduced E-chondrites, it appears that the precursor material was relatively oxidized (Rambaldi *et al.* 1984). Conditions must have become more reducing with time (Grossman *et al.* 1985), presumably because of a dust-gas fractionation in the nebula that raised the C/O ratio above the critical value of 0.8 (Larimer 1975; Larimer & Bartholomay 1979, 1983).

(*c*) Ca, Al-*rich inclusions* ('CAI')†

Chemically and mineralogically, CAI look like an *early condensate* from a solar gas (Larimer & Anders 1970; Marvin *et al.* 1970; Grossman 1972). They contain the suite of refractory minerals and trace elements expected to condense in the first *ca.* 6% of the 'rock' fraction, between 1800 and 1400 K (figure 2). However, figure 2 applies equally well to evaporation and to condensation, and so some authors have argued that CAI are *evaporation residues* (of presolar matter) rather than condensates (Kurat 1970; D. D. Clayton 1975; Wood 1981).

It turns out that the truth lies in between, perhaps closer to the latter view. In the most conclusive test thus far, Niederer & Papanastassiou (1984) have shown that the isotopic fractionation patterns of Ca and Mg require a complex sequence of both processes. Of course, both are natural extensions of the chondrule-forming process. In some cases, this process overshot the mark, vaporizing part of the chondrule and leaving a residue enriched in refractories such as Ca, Al, and Sc (Osborn *et al.* 1974). As the vapour would have recondensed eventually in the (generally cool) nebula, the chondrule-forming process thus would yield condensates, in addition to evaporation residues and fully or partially melted chondrules.

Two main classes of inclusions are recognized on the basis of grain size. *Coarse-grained* inclusions are uniformly enriched in refractory elements to about 17 times that of Cl-chondrite levels (Grossman *et al.* 1977), and hence may be interpreted as a *total condensate* of the first 6% of 'rock' (figure 2) or as an *equilibrium evaporation residue* of the last 6%. The latter view seems to be closer to the truth, judging from the presence of isotopically anomalous spinel (§4*a*), which could not have condensed from or equilibrated with the nebula.

Fine-grained inclusions have highly fractionated abundance patterns, being enriched in refractories (but by variable factors) and in volatiles such as alkalis and halogens (Grossman & Ganapathy 1976). They contain not only the usual refractory minerals, but also feldspathoids such as nepheline ($NaAlSiO_4$) and sodalite ($3NaAlSiO_4.NaCl$). In terms of the condensation diagram (figure 2), they can be described as discontinuous condensates, which lost material condensing between 1800 and *ca.* 1600 K (heavy lanthanides and other highly refractory elements) and 1400–1200 K (metal and silicate), but collected elements condensing in other intervals, sometimes disproportionately so.

Some important if ill-understood clues to the formation conditions of coarse-grained CAI come from 'Fremdlinge'‡: 10–100 μm aggregates containing metal; alloys of Pt metals, Ga, Ge, Mo, Sn, and Re; Ca-phosphates; and sulphide or oxide minerals of other rare elementss such as Sc, V, Zr, Nb, Mo and W (El Goresy *et al.* 1978; Armstrong *et al.* 1985). Some alloys can be rationalized in terms of condensation temperature, structure, or chemical reactivity; others, such as Os, Ru, Rh, cannot (T_{cond} = 1910, 1650 and 1470 K at 10^{-3} atm; structure HCP for Os, Ru but FCC for Rh).

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† The literature on this topic is vast. For a more thorough treatment, see Grossman (1980), Wood (1981), Wark & Lovering (1982), Kornacki & Wood (1984) and Huss (1987).

‡ German for 'strangers', to connote a separate, possibly extrasolar, origin.

It seems that these grains condensed under conditions highly conducive to nucleation and hence low supersaturation. Thus atoms could choose their preferred condensation sites and even a rare element such as Re was able to form its own phase rather than alloying with the previously condensed, structurally similar and more abundant Os. Yet owing to the rarity of these metals (solar $Os/H = 2\times10^{-11}$), it would take some 600 years for a 1 μm Os grain to form at $P = 10^{-4}$ atm, correspondingly less at higher P (Palme & Wlotzka 1976). It is hard to see how the required high temperatures were sustained for so long a time, and how they were regulated to maintain low supersaturation. But the growth time is proportional to $1/P$, and so shorter times would suffice at higher pressures. Perhaps this is an argument for models postulating higher pressures (protoplanets, Cameron (1978); denser gas regions surrounding planetesimals, Hayashi *et al.* (1985)).

To complicate matters, the oxide, sulphide, and metal phases in Fremdlinge require a very wide range of oxygen fugacities and temperatures (Armstrong *et al.* 1985). Most of the trace elements in question are about as rare as Os, and so the same dilemma about growth time applies, but with the added complication of a wide spectrum of 'microenvironments', differing in oxygen fugacity and gas composition. The problem cannot be solved by relegating it to some presolar stage, as no known astronomical object offers the required combination of temperature, density, time and composition. Supernovae, in particular, are too tenuous and short-lived for significant grain growth. Though *abundant* elements such as Mg or Al can yield micrometre-sized condensates in the few months available for condensation (Lattimer *et al.* 1978), *rare* elements such as Os cannot; an average Os atom will have only 10^{-12} collisions with another Os atom during the entire cooling interval from 1600 to 0 K.

By default, it seems necessary to invoke the solar nebula or the meteorite parent bodies as the site of the 'microenvironments'. If the alloys formed by condensation, then the required chemical variety (especially the high oxygen fugacities) could be achieved either in the nebula itself or in the protoatmosphere of growing asteroids or protoplanets (Cameron 1978; Hayashi *et al.* 1985). In the nebula, high oxygen fugacities could be produced by gas-dust fractionation; in a protoatmosphere, by evaporation of accreting ice. In both settings, further variety would result from settling of dust through gas regions depleted in certain elements by prior condensation.

On the other hand, if the alloys formed by exsolution during metamorphism in the meteorite parent body (Armstrong *et al.* 1985), then the required changes in chemical environment could be caused by migration and escape of major volatiles, such as CO, H_2O, H_2S, etc. In any event, it seems that the 'Fremdlinge' are not so 'fremd', after all.

(*d*) *Carbon and organic matter*

Primitive meteorites contain up to 4% C, with organic carbon dominant in C1s and C2s, but elemental carbon dominant in C3s (table 1). Abundances of total C, H, and N also decline in this order, with a sharp drop beyond C3. (As C-chondrites of higher petrologic types are very rare, H4 to H6 chondrites have been substituted.) The condensation temperatures from the solar nebula have been estimated from various 'cosmothermometers', such as isotopic fractionations or trace element abundances (Larimer 1978).

The organic carbon consists mainly of an intractable, aromatic polymer similar to coal in its structural units (Hayatsu *et al.* 1977), and a variety of extractable organic compounds, including alkanes, alkenes, arenes, alcohols, carboxylic acids, amino acids, nitrogen

TABLE 1. CARBON IN CHONDRITES

type	temperature/K condensation	temperature/K metamorphism	carbon total (ppm)	carbon organic (%)	carbon C† (%)	carbon CO_3^{2-} (%)	H (ppm)	N (ppm)
C1	360	—	36000	95	~ 1	~ 4	7900	2800
C2	400	—	23000	95	~ 1	~ 2	8900	1500
C3V	420	600	6700	$\leqslant 20$	> 80	—	500	61
H4	450	900	1300	< 10	> 90	—	5	47
H5	470	1000	1100	< 10	> 90	—	4	43
H6	490	1200	1060	< 10	> 90	—	4	50

† Elemental C or solid solution of C in γ-Fe.

heterocyclics (including the purine and pyrimidine bases of DNA and RNA), etc. (Hayatsu & Anders 1981).

The alkanes consist mainly of the normal, straight-chain isomers, with lesser amounts of four or five slightly branched isomers (mono- and di-methyl). As there exist many thousands of possible isomers (e.g. 10359 at C_{16}), the dominance of only five or six implies a highly selective process: presumably the Fischer–Tropsch synthesis, which involves catalytic hydrogenation of CO. Indeed, laboratory experiments show that all organic compounds reliably identified in meteorites can be produced from CO, H_2 and NH_3 in the presence of catalytically active meteoritic minerals, such as Fe_3O_4 and clays (Studier *et al.* 1968; Hayatsu & Anders 1981).

Figure 3 illustrates the thermodynamics of this process in a solar gas. Let us consider isobaric cooling at 10^{-5} atm, although isothermal compression at *ca.* 400 K would give similar results.

At high temperatures carbon exists mainly in the form of CO. Near 600 K, hydrogenation to CH_4 should commence, and be 50% complete at 590 K. However, this reaction is very slow in the absence of catalysts, and as the dominant minerals at this temperature (Fe, Mg-silicates, FeNi coated with FeS) are very poor catalysts (Anders *et al.* 1973), most of the CO survives metastably.

At 520 K, disproportionation to C and CO_2 should set in. Again, good catalysts are lacking, and so much of the CO survives. Finally at 400 K, the anhydrous mineral assemblage dominant up to that point (figure 2) transforms to clays and Fe_3O_4, both of which are good catalysts for the hydrogenation of CO. However, at this low temperature, CH_4 – though still the most stable product – is no longer the only possible one. Many other organic compounds can form metastably, and actually do, being favoured by the reaction mechanism.

The formation of organic compounds thus is triggered by the appearance of catalysts at 400 K. Indeed, meteorites rich in clay minerals (C1, C2) are rich in organic compounds, whereas meteorites poor in clay minerals (C3) have little or no organic matter, containing most of their carbon as elemental C (table 1).

The aromatic polymer forms as a secondary product in this scheme, by aromatization of the primary aliphatic products. Such aromatization is known to occur on Fe_2O_3 (Galwey 1972), and has also been seen in a Fischer–Tropsch synthesis extended over six months (Hayatsu *et al.* 1977).

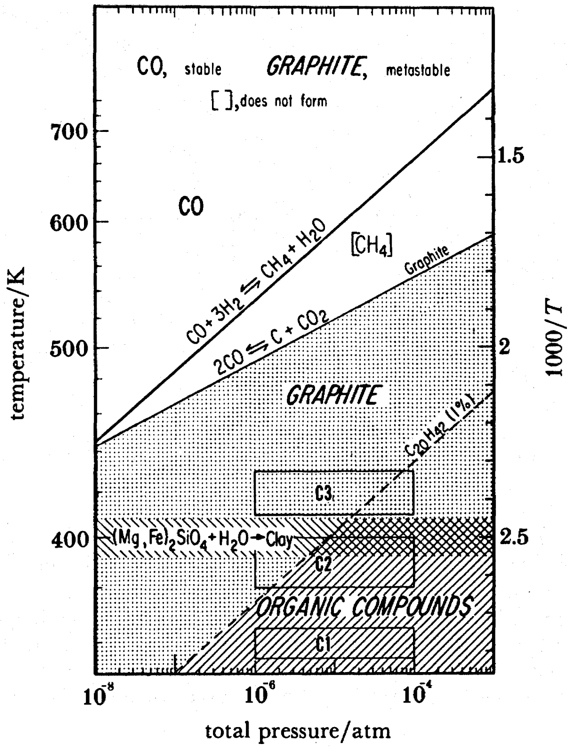

FIGURE 3. Chemical state of carbon in the solar nebula (after Hayatsu & Anders 1981). Each solid line gives the temperatures at which 50% of the carbon should have reacted according to the equilibrium shown. In each field, the principal stable products are shown in Roman type and metastable products, in italics; those that do not form for kinetic reasons are enclosed in brackets. This diagram applies to isobaric cooling as well as to isothermal compression.

CO survives down to 400 K, owing to the lack of good catalysts. At this temperature, clays form from anhydrous silicates, catalysing hydrogenation of CO to complex organic compounds (Fischer–Tropsch reaction).

The chemical state of carbon in C-chondrites agrees with that predicted from their formation conditions (indicated by boxes), as inferred from various cosmothermometers and -barometers (Larimer 1978). C3 chondrites contain mainly elemental carbon, whereas C1 and C2 chondrites contain mainly organic carbon (table 1).

4. EXOTIC COMPONENTS OF CHONDRITES

(a) *Oxygen*-16

The most abundant and ubiquitous exotic component is ^{16}O (R. N. Clayton *et al.* 1973; D. D. Clayton 1981). Its presence can be demonstrated by a 3-isotope diagram, a variant of which is shown in fig. 4. On this diagram, all samples related by *mass fractionation* lie along a line of slope $\frac{1}{2}$ (reflecting the mass differences $^{17}O-^{16}O$ and $^{18}O-^{16}O$), whereas *mixtures of independent components* lie along 'mixing lines' of variable slope joining the two components.

All terrestrial and some meteoritic samples lie on the fractionation line, but many meteoritic samples do not, suggesting the presence of an independent component. This component is most clearly characterized by the anhydrous minerals of C2 and C3 chondrites (especially CAI), which lie on a line of slope *ca.* 1 and hence must be mixtures of two components of nearly identical $^{17}O/^{18}O$ ratio but different ^{16}O content. The ^{16}O-rich endmember is represented by spinel and pyroxene of $\delta^{18}O = -40‰$ and $\delta^{17}O = -42‰$. The same endmember seems to be required for ordinary chondrites and for certain anomalous inclusions [Clayton 1981, §4*b*], and by

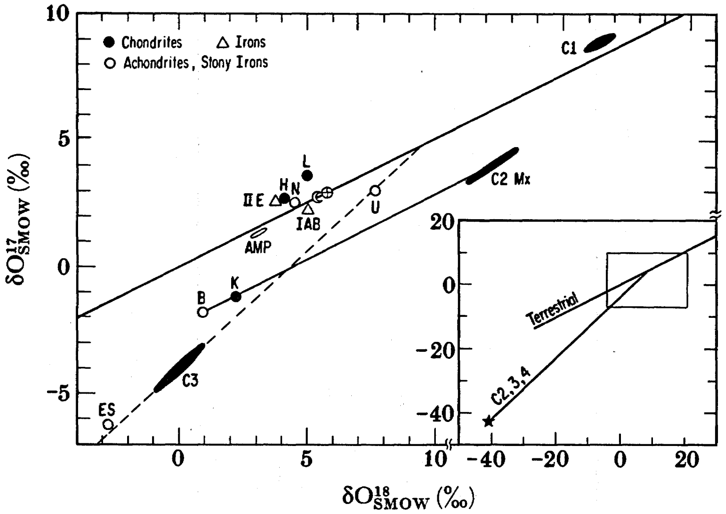

Figure 4. Three-isotope plot shows variation of meteoritic $^{18}O/^{16}O$ and $^{17}O/^{16}O$ relative to standard mean ocean water (SMOW) (R. N. Clayton *et al.* 1973). Inset gives total observed range for individual minerals; main graph shows bulk meteorites or matrix. All terrestrial samples lie on a mass-fractionation line of slope $\frac{1}{2}$. The anhydrous minerals of C2–C4 chondrites, on the other hand, lie on a mixing line of slope *ca.* 1, corresponding to constant $^{17}O/^{18}O$ but variable ^{16}O. The latter presumably comes from an ^{16}O-rich component, typified by Allende spinel (star in inset). All meteorites fall off the terrestrial line to varying degrees, indicating that their ^{16}O contents differ from that of the Earth and hence preclude a genetic relation.

Occam's Razor, other meteorite classes. Meteorites lying below the terrestrial line contain more of the ^{16}O-rich component than does the Earth, whereas those above the line contain less. The lateral dispersion may reflect mass fractionation between solid and nebular gas. For hydrothermally altered meteorites, i.e. C1s and C2s, there seem to have been further fractionations involving liquid water (Clayton & Mayeda 1984).

This diagram may be used to trace genetic relations among meteorites and planets. Objects lying on the same fractionation line (e.g. Earth, Moon) may be linked by simple mass fractionation and hence *may* have a common origin, either in the same body or in a homogeneous region of the nebula. Objects that do not lie on the same fractionation line contain different amounts of ^{16}O and hence presumably come from different bodies. There is no simple relation between ^{16}O content and primitiveness or (presumed) heliocentric distance. C1 and C2 chondrites, both more primitive than the Earth, lie on opposite sides of the terrestrial line.

(*b*) Mg, Si, Ca, Ti, Cr, Sr, Cd, Ba, Nd, Sm

The literature on the subjects of §4*b*, *c* is vast, and cannot be fully covered in this paper. The reader may wish to consult the reviews by Arnould (this symposium), Shima (1986), Wood (1981), Begemann (1980) and Wasserburg *et al.* (1980).

Mg and Si in CAI often show evidence of slight mass fractionation, apparently reflecting condensation, evaporation, or both (Lee & Papanastassiou 1974; Yeh & Epstein 1978; Molini-Velsko *et al.* 1983). So does Cd, a volatile trace element (Rosman *et al.* 1980).

All CAI from Allende & Leoville show small (*ca.* 0.8‰) excesses of ^{50}Ti (Niederer *et al.* 1981; Niemeyer & Lugmair 1981, Fahey *et al.* 1985). Presumably a separate dust component was present, enriched in ^{50}Ti (D. D. Clayton 1981). Similar enrichments have been found for Ca^{48} (Jungck *et al.* 1984) and ^{54}Cr (Birck & Allègre 1984).

The remaining elements are isotopically normal in most CAI, but show fractionation and unidentified nuclear effects in a few special inclusions, designated by the acronym 'FUN' (Wasserburg *et al.* 1977). The FUN inclusions also have unusual oxygen isotope compositions, lying on mixing lines between a common (gas?) composition (H in figure 5) and several points (B, C, D, E) along a mass fractionation line AG. This line emanates from the ^{16}O-rich composition A, representing the anomalous dust component in the nebula. Two models have been proposed for the FUN inclusions.

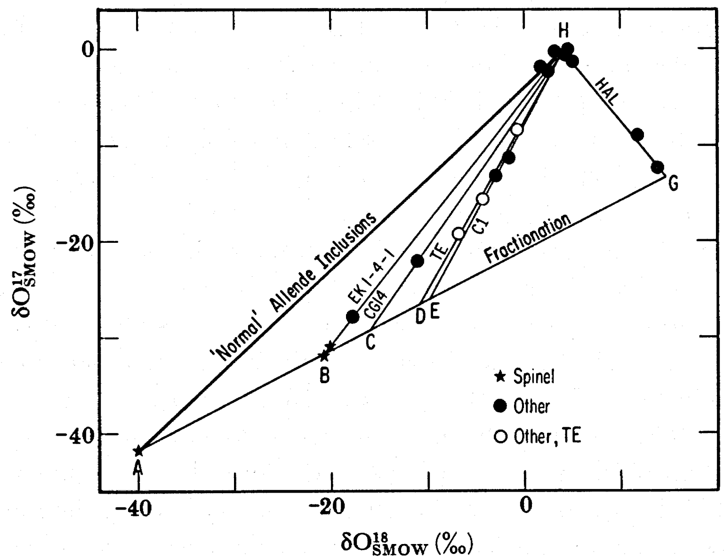

FIGURE 5. Oxygen in FUN inclusions (fractionation and unidentified nuclear effects) from Allende (R. N. Clayton *et al.* 1984). A few exceptional inclusions fall off the normal mixing line, and show FUN effects. Bulk samples and separated minerals of each inclusion form linear arrays converging on a common point H. Presumably these inclusions first formed by mass fractionation of dust (composition A) along the line AG, and later partially exchanged with nebular gas H. Spinel, with the slowest exchange rate, remained on the original fractionation line, whereas more reactive minerals (melilite, pyroxene) moved toward H.

(1) Dust component A underwent mass fractionation, yielding solids B, C, D, etc. Each of these polymineralic solids then reacted with the nebular gas H, under conditions where only the most reactive minerals achieved exchange equilibrium (Blander & Fuchs 1975). This left the minerals strung out along mixing lines BH, CH, etc. (R. N. Clayton & Mayeda 1977; Wasserburg *et al.* 1977; Lee *et al.* 1980).

(2) Solids A, B, C, etc. are mixtures of two mass fractionated components in a presolar locale, gas to the left of A and dust to the right of G. Upon arrival in the Solar System, these solids would partially equilibrate with nebular gas H, as above (D. D. Clayton 1981).

On either model, some *ad hoc* reason must be found why only the highly mass fractionated solids B, C, retained nuclear anomalies; in other words, why is F coupled with UN? One possibility is that a presolar melting process greatly coarsened B, C, D, etc. compared with A, thus minimizing uptake of late, fine-grained ingredients of the Solar System mix of heavy elements.

(*c*) *Extinct radionuclides*

Seven extinct radionuclides, with the half-lives given in parentheses, have left detectable decay products in meteorites: ^{41}Ca (1.0×10^{5} a), ^{26}Al (7.2×10^{5} a), ^{53}Mn (3.7×10^{6} a), ^{107}Pd

(6.5 × 10^6 a), ^{129}I (1.6 × 10^7 a), ^{244}Pu (8.1 × 10^7 a), and ^{146}Sm (1.03 × 10^8 a).† Some (^{41}Ca, ^{26}Al, ^{53}Mn) have been found only in CAI and others only in irons (^{107}Pd), but this reflects mainly detectability, not necessarily limited distribution. These nuclides typically occur at only 10^{-2}–10^{-5} the abundance of neighbouring nuclides, and so their decay products will be detectable only if parent and daughter element have been chemically fractionated to an appreciable degree. For this reason, extinct nuclides are rarely found in matrix, which – though quite ancient – is the least fractionated part of meteorites.

The original hope that extinct radionuclides would provide a detailed, relative chronology of the early Solar System has not materialized. At best, one can obtain an isochron giving the ratio of an extinct nuclide to a stable isotope (e.g. ^{26}Al/^{27}Al) in a given meteorite, referring to the time the system became closed to diffusion of the daughter (^{26}Mg in this example). But if two meteorites have different ratios, this can reflect not only differences in age but also differences in initial ratios, given the evidence for isotopic heterogeneity in the early Solar System (§4*a*, *b*). There is no way of separating these two variables.

In view of this ambiguity, very little can be said about the variation of extinct radionuclides with heliocentric distance. The distribution of ^{26}Al can perhaps be traced by its heat effects, as the usual ^{26}Al/^{27}Al ratio of 5 × 10^{-5} would cause even asteroids of *ca.* 10 km to melt. On this basis, one could argue that little or no ^{26}Al got into the Earth and other terrestrial planets, as the heat effects would be staggering (Urey 1955). However, as the accretion time of the terrestrial planets (*ca.* 10 Ma (Wetherill 1980)) is much longer than the half-life of ^{26}Al (0.72 Ma), slow accretion would solve the problem equally well. An argument in favour of patchy distribution of ^{26}Al is the difference in reflection spectra among the largest asteroids. 4 Vesta is differentiated to basalt and hence must have extensively melted, whereas 1 Ceres and 2 Pallas have relatively primitive, olivine–magnetite surfaces, implying little or no differentiation (Gaffey & McCord 1978).

(*d*) *Noble gases*, C, N, *and* H

Several isotopically anomalous components of these elements have been found in meteorites. The noble-gas components, though present at concentrations of only 10^8–10^{10} atoms g^{-1}, were the first to be identified, and then served as tracers in the search for their (likewise exotic) carrier phases. Most of these carriers are carbonaceous, but have not yet been fully characterized or isolated in pure form. They differ from each other in noble-gas release temperature, grain size, density, combustibility and isotopic composition. Pending further characterization, they are non-committally designated by Greek letters.

Table 2 lists the exotic noble-gas components and their carriers, as well as two exotic components of H and N that are not associated with noble gases.

Two types of Ne-E are distinguished, differing in release temperature and carrier. Ne-E(L) is virtually pure (not less than 99 %) Ne^{22}, and though the experimental limit for Ne-E(H) is less stringent (not less than 83 %), it, too, probably is pure ^{22}Ne. The only plausible source of monoisotopic ^{22}Ne is β^+-decay of ^{22}Na ($t_{\frac{1}{2}} = 2.6$ a). The short half-life of ^{22}Na implies that it was synthesized and trapped on an equally short timescale, i.e. under catastrophic conditions, presumably in a nova (Clayton & Hoyle 1976; Arnould & Beelen 1974). The high δ^{13}C values

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† For references, see Begemann (1980), Wasserburg *et al.* (1980), Birck & Allègre (1985), Crabb & Anders (1982), Lugmair *et al.* (1983) and Hutcheon *et al.* (1984).

TABLE 2. EXOTIC COMPONENTS OF NOBLE GASES, C, H, AND N

component	carrier	enriched in	release T/°C	δC^{13} (‰)	δN^{15} (‰)	abundance (ppm)	size/µm	probable source	reference
Ne-E(H)	spinel	Ne^{22}	1200	> 2000		1700	1–10	nova?	*a,e,i*
Ne-E(L)	Cα	Ne^{22}	600	340	> 0	5	1–10	nova	*a,c,e,o*
Xe-S	Cβ	Xe^{130}	1400	1400	> 0	2	0.1–3	red giant	*a,j,n,o,p*
Xe-HL	Cδ (diamond)	$Xe^{124,136}$	1000	-38	-330	400	0.005	supernova	*g,h,l,o,q*
D	polymer†	D	400	?	?	300	?	molecular cloud	*d,f,m,p*
Heavy N	?				973	< 60		nova?	*k*

† Minor (acetylenic?) component of meteoritic polymer.

(*a*) Alaerts *et al.* (1980); (*b*) Black & Pepin (1969); (*c*) Carr *et al.* (1983); (*d*) Hayatsu *et al.* (1983); (*e*) Jungck & Eberhardt (1979); (*f*) Kolodny *et al.* (1980); (*g*) Lewis *et al.* (1975); (*h*) Lewis *et al.* (1983); (*i*) Niederer *et al.* (1985); (*j*) Ott *et al.* (1985); (*k*) Prombo & Clayton (1985); (*l*) Reynolds & Turner (1964); (*m*) Robert *et al.* (1979); (*n*) Srinivasan & Anders (1978); (*o*) Swart *et al.* (1983); (*p*) Yang & Epstein (1984); (*q*) Lewis *et al.* (1987).

certainly are consistent with a nova origin, but the expected high $\delta^{15}N$ is yet to be confirmed, as the samples analysed were not pure enough (Lewis *et al.* 1983).

The association of Ne-E(H) with spinel is less certain now than it was two years ago. Niederer *et al.* (1985) have found heavy C of $\delta^{13}C > 2000‰$ in Ne-E-bearing spinel fractions. It is not clear whether this heavy carbon is present as discrete grains clinging to or enclosed in spinel or whether it is dissolved in spinel on an atomic scale.

Xenon-S shows the distinct signature of the s-process: strong predominance of even isotopes over odd ones (128, 130, 132 compared with 129, 131), rise from 128 to 132, and absence of Xe isotopes that do not lie on the s-process path (124, 126, 134, 136). Its carrier, Cβ, also shows the expected enrichment in ^{13}C. As the s-process is known to take place in red giants, Xe-S and Cβ must have formed in a red giant. However, it is not known whether they were expelled from the star during the red-giant stage (red giants lose mass at a rate of 10^{-7}–10^{-8} $M_\odot$ a^{-1}, forming circumstellar shells of carbon grains), or only later. Nor is it known whether Cβ represents general galactic background from many red giants or material from a single star in the OB association from which the Solar System formed (Reeves 1979).

The nucleosynthetic origin of Xe-HL is less well understood, though a supernova, with high rates of neutron capture and photonuclear reactions, may provide the right environments for simultaneous enrichment of heavy and light isotopes (Heymann & Dziczkaniec 1979). One interesting clue is the lack of enrichment at mass 129 relative to 128 or 130, although mass 129 – in contrast to the others – originally forms not as xenon but as a long-lived iodine isotope ($t_{\frac{1}{2}} = 16$ Ma). Apparently the atoms were trapped by a chemically non-selective process, such as ion implantation (Lewis & Anders 1981). This could happen if the supernova ejecta, travelling at *ca.* 10^4 km s^{-1}, overtook a shell of carbon grains ejected at the presupernova stage (D. D. Clayton 1982). The carrier, Cδ has been identified as diamond (Lewis *et al.* 1987), which presumably condensed metastably in the presupernova dust shell. This supports an earlier suggestion by Saslaw & Gaustad (1969) that diamond is a major constituent of interstellar dust.

Deuterium enrichments of up to 3000‰ have been found in stepped combustion or pyrolysis of the organic polymer from primitive chondrites (Kolodny *et al.* 1981). Although such enrichments could, in principle, be achieved by equilibrium fractionations, rates are far too low at the required temperatures of *ca.* 100 K (Geiss & Reeves 1981). More probably, these enrichments were caused by ion-molecule reactions (Kolodny *et al.* 1980; Geiss & Reeves 1981), which are believed to be responsible for the even greater D-enrichments in interstellar molecules (Smith, this symposium). The observed enrichment factor of *ca.* 50 relative to galactic hydrogen (figure 6) might represent either formation of the entire polymer by ion–molecule reactions at a rather high temperature (*ca.* 60 K) and consequently small fractionation, or admixture of a minor amount of highly enriched material, i.e. polymerized interstellar molecules.

The latter possibility seems more likely. On acid treatment or oxidation, the D-rich material reacts preferentially, which suggests that it contributes a minor fraction of the total (McNaughton *et al.* 1982; Yang & Epstein 1984). Detailed structural analyses of the polymer show that while most of it is aromatic, as discussed in §3*d*, a minor part (*ca.* 1%) is acetylenic (Hayatsu *et al.* 1983). The latter component yields fragments such as C_mH ($m = 3$–10)and C_mCN ($m = 3$–6), which are closely related to polycyanoacetylenes [$H(C{\equiv}C)_nCN$], a prominent class of interstellar molecules ranging up to $n = 6$.

A strikingly large enrichment of ^{15}N (up to 973‰) has been found in the brecciated stony

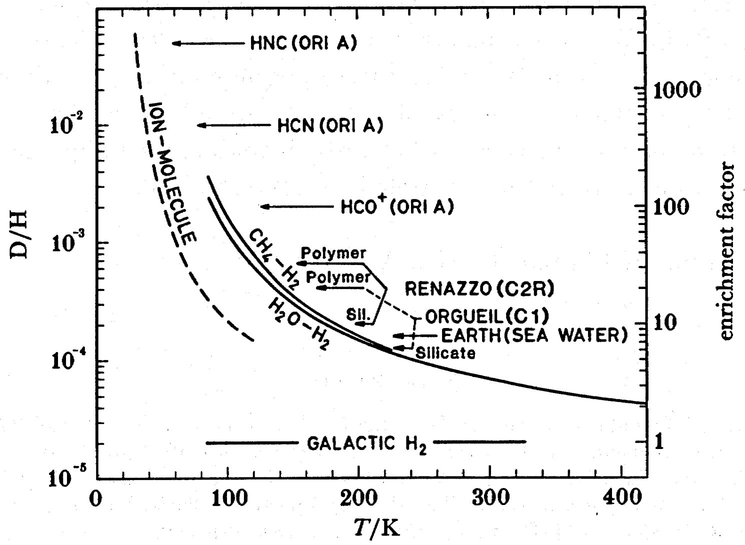

Figure 6. (Adapted from Geiss & Reeves (1981).) Organic polymer fractions from C-chondrites are strongly enriched in deuterium, up to 32 times relative to galactic hydrogen. Equilibrium isotope fractionations can produce such enrichments, but only at temperatures of *ca.* 100 K, where rates are prohibitively slow (e.g. the half-time of the Fischer–Tropsch reaction at 110 K is *ca.* 10^{30} years!) More probably, the fractionation is due to ion–molecule reactions, which are thought to be responsible for the large (up to 10^5-fold) D-enrichments in interstellar molecules. Only a minor, reactive part of the meteoritic polymer seems to be enriched in D, and probably represents relict interstellar molecules. The major part appears to be of local origin, as outlined in figure 3.

irons Bencubbin and Weatherford (Prombo & Clayton 1985). The nature of the carrier is not known. By their oxygen isotope composition the silicates may be related to C2R and C2M chondrites, and there are suggestions that the metal clasts may be direct nebular condensates (Newsom & Drake 1979). But otherwise these meteorites are far from primitive, having undergone severe brecciation and perhaps impact melting (Kallemeyn *et al.* 1978). At present, it is not possible to decide whether the nitrogen anomaly is nuclear or chemical, i.e. produced by ion–molecule reactions in a molecular cloud (Prombo & Clayton 1985).

5. Comets

We already know that the 'rock' fraction of comets is approximately chondritic, and that the 'ice' fraction differs appreciably from the equilibrium composition predicted by figure 1, because it contains mainly CO_2 rather than CH_4 as well as formaldehyde and other 'interstellar' molecules (Delsemme 1982; Huebner *et al.* 1982). For further characterization of comets, we would like to know to what extent they shared the 'reprocessing' of chondrites (Anders 1986). Do comets contain chondrules, CAI, Fe^{2+}-rich silicates, hydrated minerals, magnetite, etc.? Were they ever exposed to liquid water? Have they been metamorphosed? What kinds of organic compounds do they contain? Some clues have already been obtained from meteor spectra (Wilkening 1975) or stratospheric dust particles (Brownlee, this symposium; Fraundorf & Shirck 1979), and others will undoubtedly come from the Vega and Giotto missions. But most of the answers will require a sample return mission.

Such a mission is even more essential if we want to study the isotopic record of comets, particularly its presolar part, which ought to be clearer and more diverse than that in meteorites. Two isotopic anomalies – for Mg (Esat *et al.* 1979), and H (McKeegan *et al.* 1985) – have actually been detected in stratospheric dust particles, at least some of which are thought to be of cometary origin (Brownlee, this symposium). However, these particles are so small (less than 10^{-9} g) that only a few of the most abundant elements can be analysed isotopically. The remainder of the cometary record – including the unique presolar part – will remain inaccessible to us until a comet sample is returned to Earth.

This work was supported in part by NASA Grant NAG 9-52.

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