PART VI

PRIMITIVE METEORITES

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INTERSTELLAR MATERIAL IN METEORITES: IMPLICATIONS FOR THE ORIGIN AND EVOLUTION OF THE SOLAR NEBULA

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Several isotopic anomalies, unexplained by known nuclear or non-nuclear processes within the solar system have been attributed to the preservation of pre-solar variations. The largest of these (in number of atoms) in an ^{16}*O-excess (up to 5%) in "high-temperature condensate" minerals in primitive meteorites. Some of these same minerals have an excess of* ^{26}*Mg, probably a decay product of* ^{26}*Al, that could have been a major source of heat for melting and metamorphosing planetesimals. Excesses of* ^{22}*Ne and of isotopes of Xe, found in carbonaceous chondrites, may have origins in presolar solid particles. Large variation in the isotopic abundances of nitrogen and carbon in meteorites may also represent isotopic heterogeneity in the solar nebula. Most of these "Isotopically anomalous" elements are found to be highly concentrated in minute phases within the meteorites, rather than being uniformly distributed. The identification and characterization of these carriers of presolar materials constitutes the principal thrust of current research in this area.*

INTRODUCTION

In most currently popular theories of formation of the solar system, an initially cold cloud of gas and dust is thought to have collapsed gravitationally, releasing sufficient energy to heat and vaporize some or all of the dust. On subsequent cooling, the vapors recondensed and the massive objects of the solar system grew by accretion and agglomeration of the condensates. Until recently it was believed that the nebula was chemically and isotopically homogeneous, as a result of complete mixing of atoms of diverse nucleosynthetic origin, either in interstellar space prior to formation of the nebula, or in the nebula itself prior to condensation. This paper deals with the recent observations which show that the nebula was not completely homogenized, and that it contained relics of presolar materials which are now observable in the primitive meteorites.

The interest in finding and characterizing presolar particles lies in three major areas: (1) as samples of ancient objects which may carry a record of the environment in space (*e.g.*, cosmic rays) at times hitherto inaccessible to observation; (2) as tracers for processes in the earliest history of the solar system, and (3) as possible samples of single nucleosynthetic events. Discrete presolar particles have not yet been identified in meteorites, and their existence is inferred from the isotopic "anomalies" discussed below.

The strongest evidence for an initially homogeneous solar nebula came from the apparent absence of variable isotopic composition in elements whose isotopes are believed to have been formed by different nucleosynthetic processes in different astrophysical environments (the p, r, s processes, etc. of Burbidge *et al.* 1957). The first suggestion of a departure from uniformity (except for well-understood effects of radioactive decay, cosmic-ray induced nuclear reactions and mass-fractionation) was made by Reed and Jovanovic (1969) for the element mercury in which they observed occasional enhancements in the ^{202}Hg/^{196}Hg ratio in some meteorites. Black (1972) found that neon in carbonaceous chondrites contained a component, enriched in ^{22}Ne, which had no obvious origin within the solar system, and he attributed to it a presolar origin. Perhaps because neon and mercury occur in meteorites only in trace amounts, there were sufficient doubts that some nuclear process had been overlooked (either in the laboratory or in the solar system), so that there was not widespread agreement that the neon and mercury observations provided strong evidence for presolar matter in meteorites.

Clayton *et al.* (1973) found large excesses of ^{16}O in the so-called high-temperature condensates in carbonaceous chondrites: these minerals rich in calcium, aluminum and titanium which probably formed from the hot nebular gas before condensation of the much more abundant magnesium silicates (Grossman 1972). Mass fractionation can be ruled out as the cause of the effect, since the ^{18}O/^{17}O ratio is virtually constant and "normal" in the same phases that show ^{16}O-excesses. The observed ^{16}O-excesses range up to 5% of the total oxygen in individual samples. Since oxygen is the most abundant element in the meteorites, the number of atoms required to make this "isotopic anomaly" exceeds that for the trace-element isotopic anomalies by many orders of magnitude. Nuclear reactions within the early solar system, such as destruction of ^{18}O and ^{17}O by proton bombardment can be shown to be quantitatively inadequate (D. D. Clayton *et al.* 1976).

The ^{16}O anomaly is distributed very heterogeneously within the carbonaceous chondrites on a scale at least as small as a few microns (R. N. Clayton *et al.* 1976), implying the presence of ^{16}O-rich solid particles at the time of formation of the early solar nebular condensates. The most likely carriers of the ^{16}O refractory solid particles which acquired their extreme isotopic compositions in an α-process nucleosynthesis, and solidified before being mixed with the heavier oxygen isotopes produced elsewhere. If this explanation is correct, one might expect correlated isotopic anomalies in the other elements with which the oxygen was chemically bound in the solid grains. In the following sections, the present state of "isotopic anomalies" in meteorites is summarized.

ISOTOPIC VARIATIONS WITH POSSIBLE NUCLEOSYNTHETIC ORIGIN

Carbon

^{13}C/^{12}C ratios are about 8% greater in oxidized carbon compounds than in reduced compound in the C1 carbonaceous chondrites (Clayton 1963; Briggs 1963). Since carbon has only two stable isotopes, it is not possible to distinguish unambiguously between mass fractionation and nuclear effects as causes of the observed variations. Although the difference is very large for a single stage chemical isotopic fractionation, the experiments of Lancet and Anders (1970) show that kinetic effects in Fischer-Tropsch reactions between hydrogen and carbon monoxide at reasonable temperatures are sufficient to account for the observations. Although the possibility of a nuclear origin of the isotopic variations remains, the case for it must be considered very weak.

Nitrogen

Like carbon, nitrogen has only two stable isotopes. Ratios of $^{15}N/^{14}N$ are systematically high in carbonaceous chondrites and low in enstatite chondrites, the former being about 4% greater than terrestrial atmospheric nitrogen, the latter being about 4% less (Kung 1976). This variation is considerably greater than that observed among all terrestrial samples analyzed, but still may be due to mass fractionation associated with gas loss in the solar nebula. One unusual carbonaceous chondrite, Renazzo, was found to have a $^{15}N/^{14}N$ ratios 17% greater than terrestrial nitrogen, which might indicate that nuclear processes were involved . Apparently, $^{13}C/^{12}C$ ratios have not yet been measured in Renazzo.

Oxygen

All meteorites show the effect of variable ^{16}O abundance, presumably as a result of incorporation of variable proportions of ^{16}O-rich presolar material into the parent bodies as they grew (R. N. Clayton *et al.* 1976). The C3 carbonaceous chondrites contain about 1% more ^{16}O than the C1 carbonaceous chondrites. In individual mineral fractions of C3 meteorites, the ^{16}O enrichment reaches 5%.

Neon

The ^{22}Ne-rich component in carbonaceous chondrites, labelled neon-E by Black (1972), was discovered in step-wise heating experiments in which differently sited components of the gas are released in different temperature ranges. Recently Eberhardt (1974) has succeeded in concentrating the neon-E component by chemical dissolution of most of the meteorite (Orgueil, a C1 carbonaceous chondrite), thereby removing the more abundant "planetary" components. The $^{22}Ne/^{20}Ne$ ratio in the residue was enriched by a factor of 3.6 over the value for the whole meteorite, and indications are that neon-E may be pure ^{22}Ne. Although neon-E is clearly the product of nuclear reactions, debate continues over the time and place of the reaction: within the solar system or before it (Heymann and Dziczkaniec 1976; D. D. Clayton *et al.* 1976), and whether it is a decay product of 2.6-year ^{22}Na (Herzog 1972), or is formed directly by α-particle reactions in ^{14}N in a star (Cameron and Truran 1976).

No close correlation is expected for neon "anomalies" and oxygen "anomalies," since neon was trapped from the nebular gas at low temperatures, whereas the ^{16}O-rich matter was incorporated at high temperature into the early condensates.

Magnesium

Magnesium might be expected to exhibit variations of isotopic composition of nucleosynthetic origin which correlate directly with the oxygen isotope variations, since the ^{16}O-rich grains might contain refractory compounds such as spinel ($MgAl_2O_4$). The search for such variations revealed excesses of ^{26}Mg (Gray and Compston 1974; Lee and Papanastassiou 1974; Lee *et al.* 1976), which, it appears, are not primary nucleosynthetic effects, but are the result of decay of 720,000-year ^{26}Al. The ^{26}Mg excesses, the largest of which is an increase of 1.3% in $^{26}Mg/^{24}Mg$, have been observed so far only in the high-temperature condensates in the Allende (C3) carbonaceous chondrite. The evidence for an origin via ^{26}Al is the correlation between $^{26}Mg/^{24}Mg$ and Al/Mg in individual mineral samples from the condensate nodules in the meteorite. There is no mineral-by-mineral correlation between the ^{26}Mg excess and the ^{16}O excess, since the ^{16}O excess is not correlated with Al/Mg ratios.

The question of the origin of ^{26}Al, either in a presolar nucleosynthetic process or by proton irradiation in the solar nebula remains unresolved (Cameron and Truran 1976; Heymann and Dziczkaniec 1976). The apparent isochron observed in one chondrule by Lee *et al.* (1976) implies decay of ^{26}Al *in situ*. If this is the case, then ^{26}Al was present in some meteorite parent bodies in high enough concentrations to provide a major heat source, as is required to explain the early melting of the differentiated meteorites (achondrites and stony irons). If the ^{26}Al was produced before development of the solar nebula, the time interval between the end of nucleosynthesis and condensation of the solar system must have been not more than a few million years, rather than of the order of a hundred million years, as is implied from ^{129}I-^{129}Xe and ^{244}Pu-Xe chronologies.

The apparent absence of a direct nucleosynthetic isotopic anomaly in magnesium does not necessarily rule out the possibility that this element was a constituent of the ^{16}O-rich particles. Magnesium is not a sensitive indicator of nucleosynthetic origins, since all three isotopes are probably produced in the observed proportions in a single process: explosive carbon-burning (Arnett 1969), whereas carbon, nitrogen, oxygen, silicon, calcium and many heavy elements require a combination of nucleosynthetic processes in different locations to produce their observed isotopic abundances.

Krypton and Xenon

Aside from the special case of ^{129}Xe produced by decay of ^{129}I, the only one of the several components of heavy rare gases in meteorites for which a presolar origin has been proposed (Manuel *et al.* 1972) is the so-called CCF (carbonaceous chondrite fission) component (Reynolds and Turner 1964). Highly enriched samples of this component have been prepared from the Allende meteorite by chemical concentration of its host phase (Lewis *et al.* 1975). Manuel *et al.* (1972) observed that xenon has the curious property of being enriched in both the light and heavy isotopes, and suggested that the excesses result from addition of r-process and s-process products from a nearby supernova, whereas Anders *et al.* (1975) attribute them to *in situ* fission of an extinct superheavy nuclide. Both of these are exciting possibilities, and the problem is unresolved at this time.

Osmium

If p- and r-process nuclides were introduced into the solar nebula by a nearby supernova, as was suggested for xenon (Manuel *et al.* 1972), evidence for this should be present in other heavy elements. Of the isotopes of osmium, ^{184}Os is made in the p-process and ^{192}Os in the r-process. Because of its very refractory nature, osmium from an external source would probably not be vaporized in the solar nebula, and should be trapped in the early condensates. Grossman and Ganapathy (1976) found about a 1% excess of ^{192}Os, which was barely above their analytical uncertainty. Their result was based on a single Allende inclusion. Further analyses are required to assess the significance of their observation.

Mercury

The ^{202}Hg excesses observed by Jovanovic and Reed (1976) are too small to be observed in whole-rock analyses of meteorites, but are measurable in various fractions during step-wise heating of some carbonaceous chondrites and some ordinary chondrites. They attribute the excess to the decay of 300,000-year ^{202}Pb of extra-solar origin (analogous to one of the postulates given above for $^{26}\text{Al} \rightarrow {}^{26}\text{Mg}$).

SUMMARY

Until quite recently, it was accepted as one of the ground-rules of solar system cosmochemistry that the solar nebula was initially homogeneous isotopically, and that variations observed today must be accounted for by subsequent processes *within* the solar system. It is the failure to find such processes for a number of elements which has led to the proposals for presolar sources which have been discussed in this paper. Before such proposals receive widespread acceptance, it will be necessary to show that there is some coherence in the observations of various elements, so that one does not have a separate *ad hoc* explanation for each one. Probably insufficient data are yet available to form the complete picture. For example, among the light refractory elements, isotopic measurements are needed for silicon, calcium and titanium, which might be expected to show "anomalies" correlated with the oxygen anomaly.

There are many important ramifications of the observations of nebular heterogeneities of probably presolar origin. The significance of ^{26}Al as an early short-term heat source was mentioned above, and was discussed in more detail by Lee *et al.* (1976). Solar system chronologies based on the Rb-Sr and U-Th-Pb systems generally have assumed the existence of a primordial strontium and lead of uniform composition throughout the nebula. This assumption in the case of lead has recently been questioned (Tatsumoto *et al.* 1976). Finally, some of the suggestions of sources for presolar matter involve a nearby supernova explosion immediately before collapse of the nebula itself (Sabu and Manuel 1976; Cameron and Truran 1976). It is obvious that the dynamical and chemical consequences of such an event should not be ignored.

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DISCUSSION

ARNOLD: What is the pattern of O isotopes in the carbonate fractions which show the large ^{13}C *anomalies of carbonaceous chondrites?*

CLAYTON: The $^{17}O/^{16}O$ *ratio in the carbonate oxygen have not been measured.*

SINGER: I am curious about the ^{15}N *anomaly which is so large. Can't it be localized in the meteorite, is the N bound in particular compounds, and are there correlated anomalies?*

CLAYTON: In the case of Renazzo, which has by far the largest ^{15}N *excess, nitrogen was extracted in a step wise heating experiment, and four separate fractions, comprising 97% of the total nitrogen, were collected between 750° and 1350° C. All had very similar isotopic compositions, so there is no sign of especial enrichment in a particular compound or site, which might be expected to yield nitrogen over a narrower temperature range. No correlated anomalies have been found for Renazzo; it is obviously important to get a carbon isotope analysis.*

DICKEL: For the light elements (D, C, and O) changes of a few % are very small compared to apparent ranges in the interstellar medium (well over 2-1) [even though uncertainties are large] and I would not be surprised to see changes of a few % over the size of the solar system by charge exchanges, etc.

PAPANASTASSIOU: For the astronomers in the group, the isotopic effects being discussed are two orders of magnitude (or more) larger than experimental uncertainties. These effects are much smaller than $^{13}C/^{12}C$ *variations in some interstellar clouds. Similarly nucleosynthetic models predict isotopic abundances usually up to a factor of two. Despite this the observed isotopic effects in meteorites are considered as very large and very real, while no such effects have been found in terrestrial samples.*

GOLD: I do not wish to imply that I can find processes that can account for the isotopic effects that have been found. However, I do wish to remind people how large the isotopic separation effects must be in circumstances that naturally occur in the solar system. For atmospheres that have a stable temperature gradient there is no problem: if we have an atmosphere heated from above and if its height in this stable regime is several scale-heights, then of course the lighter isotopes are concentrated by a large factor at the high levels. If there are rings or satellites that skim off the outermost levels--as Titan appears to be doing--any condensate these will have a great enrichment in the lightest isotope of any condensable gas. A large gaseous planet like Jupiter could cause thick deposits of $H_2{}^{16}O$ *for example to accumulate on a satellite placed at a suitable distance. Later processes may then mix this* *with "ordinary" oxygen in the rocks and produce isotopically anomalous materials. Even in the terrestrial atmosphere at a high level the isotopic effects must be very large.*

I wanted to say all this just to give an example showing that isotope separation is by no means an impossible process in the solar system. Whether we can know enough to describe in detail the processes that might have taken place in the early solar system is quite another matter. But at any rate the problem cannot be dismissed by saying that such isotopic effects cannot possibly result from fractionation processes.

CLAYTON: Mechanism of the sort proposed require the existence of planets and satellites prior to the formation of what are considered to be the most primitive meteorites. It would be remarkable if meteorite parent bodies formed at that time could end up with solar abundances of the elements, lead and strontium of primordial isotopic composition, non-extinct ^{129}I, ^{244}Pu, ^{26}Al, *etc.*

Extreme mass-fractionation processes in general will not be ruled out until clear examples of correlated isotopic anomalies in different elements are found.