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## ORIGIN OF Ca-Al-RICH INCLUSIONS. II. SPUTTERING AND COLLISIONS IN THE THREE-PHASE INTERSTELLAR MEDIUM

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### ABSTRACT

The theory introduced by Clayton for the formation of the Ca-Al-rich inclusions within C3 meteorites is extended to an evolutionary history in a three-phase interstellar medium. Widespread supersonic turbulence in the hot interstellar medium is maintained by supernova shock waves, causing heavy sputtering of the refractory dust. Subsequent reaccumulation with varying dust/gas ratios or varying particle sizes leads to isotopically fractionated Ca-Al-rich accumulates. A new theoretical explanation of the correlation of mass-fractionated isotopes with unidentified nuclear anomalies follows from this picture, as do many other intriguing details. The Ca-Al-rich inclusions themselves are probably formed by the following sequence in the solar system: (1) cold accumulation of larger-than-average Ca-Al-rich particles containing supernova condensate (SUNOCON) cores into macroscopic ( $\sim 1$  cm) Ca-Al-rich agglomerates, probably by sedimentation; (2) fusion of the SUNOCONS into macroscopic minerals by exothermic chemical reactions that begin when the accumulate has been warmed, thereby releasing energy from the unequilibrated forms accumulated from the interstellar medium. We suggest that a fossil record of the presolar evolution of the interstellar medium has been discovered. If so, a new diagnostic of the dynamics of the interstellar medium is available.

*Subject headings:* abundances — interstellar: matter — meteors and meteorites — nebulae: supernova remnants — nucleosynthesis

### I. INTRODUCTION

This work seeks a connection between the separate dynamic phases of the interstellar medium and the chemical state of the early solar system. The approach will be to show that dust components in the ISM are expected to have specific bizarre chemical and isotopic patterns that are suggestive of those found in meteoritic inclusions. If that chemical state in the initial solar system was not erased by a high temperature state that volatilized the dust, then bodies now extant may retain a fossil record of the phases of the interstellar medium and of the residence times within them. Such a connection, if established, would enable a new discipline of study of the interstellar state. That discipline is already experimentally active in the study of meteoritics, but the connection that we draw between interesting meteoritic inclusions and the interstellar medium suggests a new prospect for astronomy. By a postulate of simplicity and by concentration on what seem to be the essential aspects rather than on the rather bewildering details of total meteoritic knowledge, we will arrive at a new interpretation of the chemical and isotopic anomalies within the meteoritic inclusions. If correct, a violent and ancient history of the interstellar medium (ISM) can be read in those small stones. We cannot get all details of such an ambitious effort right initially; indeed, this paper is entirely wrong if key isotopic fractionation patterns were established in the solar system, as the meteoritic community normally assumes. The contribu-

tion of this paper lies in the exposition of a new general picture within which the detailed solution may be found.

This idea addresses a growing cosmochemical puzzle—that those Ca-Al-rich inclusions (CAIs) of the C3 carbonaceous meteorite Allende that have been shown to have unidentified nucleosynthetic (UN) isotopic anomalies in the heavy elements are also the CAIs in which the oxygen, magnesium, and silicon show a strong mass-dependent isotopic fractionation. The combination of these two types of isotopic anomalies have been called FUN following Wasserburg, Lee, and Papanastassiou (1977). This FUN correlation, which has so far escaped explanation by solar system scientists, is nonetheless recognized as a leading clue to the origin of these interesting meteoritic inclusions and, by virtue of that connection, to the circumstances of the origin of the solar system itself. What we will show is that *the FUN correlation is expected to exist in the interstellar medium in Ca-Al-rich assemblies and, furthermore, it can be preserved during their accumulation into macroscopic bodies*. It remains for future interdisciplinary studies to ascertain whether this circumstance provides the roots of the CAIs observed in meteorites.

Paper I (Clayton 1977a) introduced a theoretical interpretation of these inclusions that will be adopted and greatly extended in this paper—that they are fused assemblies of the Ca- and Al-rich supernova condensate (SUNOCON) cores of interstellar grains. That general framework of presolar history was described thereafter

(Clayton 1978a). Clayton (1980a) demonstrated why the largest refractory SUNOCONs are expected to have the largest ratios of Al-rich core mass to Mg-rich mantle mass. On the other hand, chemists (e.g., Larimer and Anders 1967; Grossman 1972) have long argued that the solar system began as a hot gas, in order to account for the separate existence of the refractory assemblies like the Ca-Al-rich inclusions. Our own efforts have stressed the view (e.g., Clayton 1978a) that the solar system objects accumulated from a cold dusty gas containing precondensed refractory elements, not from a hot gas that is postulated to have sequentially condensed increasingly less refractory elements. We take it that meteorites accumulated from matter typical of cold molecular clouds. This routine chemical condition, which is homogeneous in its bulk composition, is alleged to contain within the distinct precondensates the probable seeds of most if not all of the chemical and isotopic anomalies (Clayton 1977a, b, 1978a). This approach to the isotopic anomalies remains a minority opinion, however. A much more popular contemporary picture has been the postulated injection of isotopically anomalous matter from a neighboring supernova, as advocated initially by Cameron and Truran (1977). Clayton (1979b) provides a comprehensive review of these competing pictures.

In this present work we lay out a plausible approach to the way that our model can explain the basic correlation noted in the second paragraph. That this approach seems to yield an explanation of the correlation may give it added support. This controversial thesis is crucial to the possibility of obtaining laboratory samples that retain a fossilized record of the ISM. That the *r*-process/*s*-process separations in the FUN inclusions point to dust/gas separations rather than to supernova injection (Clayton 1978b, c, 1979a) is consistent with the results to be presented here. To critics it must be acknowledged, however, that the following explanation is neither unique, necessary, nor deductive; it is postulated as a useful new line of thought relating modern theories of the interstellar medium to meteorites. Sputtering of interstellar dust within a dynamic multiphase interstellar medium excited by supernova shock waves will be implicated. The basic astrophysical picture has been outlined by Cox and Smith (1974), Smith (1977), and McKee and Ostriker (1977). The pervasiveness of sputtering in such a picture has been demonstrated by quantitative models of Dwek and Scalo (1979, 1980) and by Draine and Salpeter (1979b). Their results will be adopted as a guide without repeating their calculations, so the serious reader will need some familiarity with these works.

## II. THE CHEMICAL PROBLEM

The salient facts of the chemical problem are illustrated in Figures 1 and 2, which have been adapted from data of Clayton and Mayeda (1977), Yeh and Epstein (1978), Clayton, Mayeda, and Epstein (1978a, b), and of Wasserburg, Lee, and Papanastassiou (1977) in simplified form in order to spotlight only those aspects of their findings that are essential to the connections sought. The coordin-

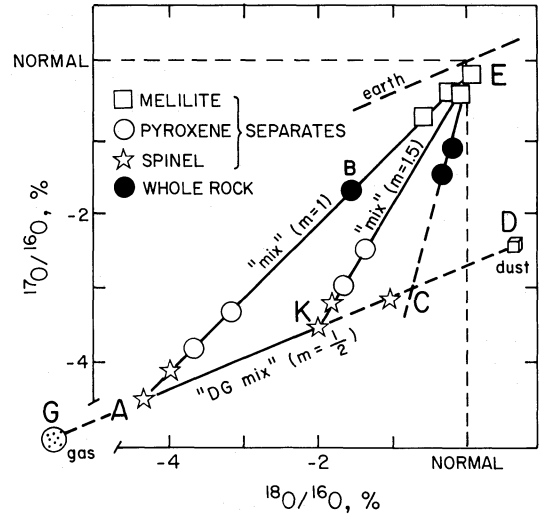


FIG. 1.—The three-isotope plot for oxygen. Terrestrial samples populate a fractionation line with slope  $m = \frac{1}{2}$  passing slightly above E. Mineral separates (spinel, pyroxene, melilite) from Allende Ca-Al-rich inclusions define three mixing lines passing from almost normal oxygen at E through normal inclusions to A, through isotopically anomalous inclusions EK1-4 to K, and through isotopically anomalous inclusion C1 to C. Mineral separates are schematically shown from data of Clayton and Mayeda (1977). Their paper must be consulted for the precision of the data (near  $\pm 0.01\%$ ) and the distribution of mineral separates. The oxygen in a recently discovered FUN inclusion (Lee *et al.* 1980) is not shown, but defines a fourth mixing line between E and another point on GD. The most  $^{16}\text{O}$ -rich minerals are the spinels of these inclusions, which populate points down to the line AC, but apparently not further. The  $m = \frac{1}{2}$  slope of AC is interpreted by us as a mixing line between isotopically heavy sputtered dust and the complementary isotopically light gas. Heavy elements are isotopically anomalous in the inclusions EK1-4 (EK) and C1 (EC). The point B is a special Rb-rich aggregate.

ates are isotopic ratios in the common three-isotope elements (O, Mg, Si) that dominate the fractionation patterns. These coordinates (a so-called three-isotope plot) have useful mathematical features in that (1) mixtures of two distinct isotopic components, each represented by a point, generate compositions lying along the straight line connecting those two points, and (2) small amounts of mass-dependent isotopic fractionation by physical processes generate new samples lying on approximately straight lines of slope  $m = \frac{1}{2}$  passing through the initial sample. Earthly samples lie along such a dashed line labeled "earth" slightly above point E, whose name is a mnemonic for "exchange oxygen." The minerals within most Allende inclusions lie along the line EA having slope  $m = 1$  (Clayton *et al.* 1977), interpreted as a mixture between E and  $^{16}\text{O}$ -rich matter at A (or, in principle, at a point beneath A on the extension of EA). The name A is a mnemonic for "Allende common inclusions." The  $^{16}\text{O}$ -richness of the mineral separates increases along the sequence melilite, pyroxene, spinel. The inclusions falling on the line EA do not seem to have unidentified nuclear anomalies, except in the element titanium, at the level set by current experimental precision.

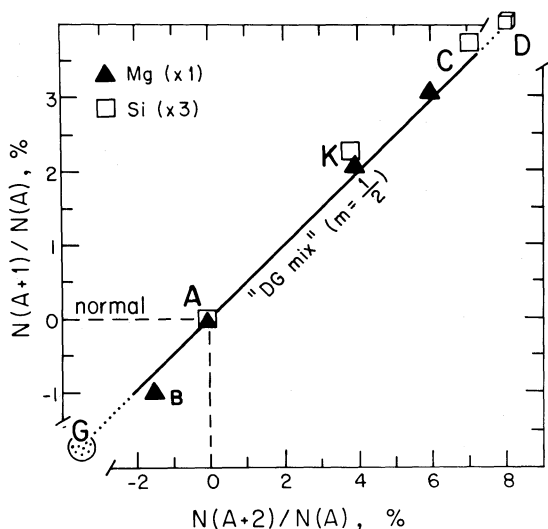


FIG. 2.—Three-isotope plots for Mg and Si in normal inclusions (A), EK1–4 (K), and C1 (C) approximately define a line of slope  $m = \frac{1}{2}$  as in Fig. 1. That nearly linear relation is interpreted as a mixture of heavily sputtered dust (D) and its complementary isotopically light gas (G). Smaller deviations from the linear relation (not obvious in this drawing) reflect a small nuclear difference between dust and gas deriving from differential condensation of the isotopes in SUNOCONS. The isotopic lightness of the Rb-rich aggregate B identifies it as gas-rich Mg. The data is from Wasserburg *et al.* (1977), Yeh and Epstein (1978), and Clayton *et al.* (1978a). For Mg,  $A = 24$ ; and for Si,  $A = 28$ . The Si fractionations have been multiplied by the factor 3 for better visibility.

The same suite of mineral separates from inclusion EK1–4 lies along the line EK having slope  $m = 1.5$ . This line is presumably also generated by a mixture of exchange oxygen at E with anomalous oxygen at K (or its extension). The heavy elements in these separates do have nuclear (N) isotopic anomalies. The other inclusion, C1, having UN anomalies in heavy elements has not been so thoroughly measured in mineral separates, but its bulk compositions for two samples lie along EC. We follow Clayton and Mayeda (1977) in assuming that mineral separates from C1 would fall along EC if they could be separated and measured. One spinel from C1 has been studied (R. N. Clayton, personal communication) and found to lie just to the left of point C as shown. A third FUN inclusion named HAL was discovered after the submission of this paper to have oxygen compositions along a line EH (not shown) where point H falls near the point D (Lee, Mayeda, and Clayton 1980).

The isotopic composition for Mg (Wasserburg, Lee, and Papanastassiou 1977) is, unlike that of oxygen, uniform within minerals of a given inclusion. Each inclusion therefore plots as a point instead of a line in the three-isotope Mg plot. The points AKC in Figure 2 locate these three inclusions for Mg. The element Si is similar to Mg in having smaller fractionation (Yeh and Epstein 1978; Clayton, Mayeda, and Epstein 1978a). Its isotopic excesses have been multiplied by three before plotting in Figure 2. These correlations, taken together with the fact that several heavier elements are otherwise isotopically normal in the many inclusions of type A but

abnormal in K and C, constitute the isotopic enigma. The very existence of aggregates rich in Ca, Al, and other very refractory elements is the chemical enigma.

Clayton and Mayeda (1977) and Wasserburg, Lee, and Papanastassiou (1977) both emphasized another significant aspect of the almost straight lines AKC. They have slope  $m = \frac{1}{2}$ , as if they represent different samples of the same source that have been subjected to differing degrees of mass-dependent isotopic fractionation. They postulated that some process imposes such fractionation on solar system samples, leading to otherwise similar aggregates for the parents of these inclusions. They proposed that subsequent reaction with solar system oxygen gas at isotopic composition E was the mixing process that generated the three mixing lines AE, KE, and CE. They argue that no process other than mass fractionation could accidentally cause O, Mg, and Si lines AKC to lie along a slope  $m = \frac{1}{2}$ . Furthermore, the chords AC:AK stand in the ratio  $\frac{3}{2}$  for both O and Mg, slightly greater for Si.

These arguments implicating mass-dependent isotopic fractionation are convincing. What is missing is identification of the reason for the mass fractionations being so large and for their being accompanied by isotopic anomalies of an unidentified nuclear type within heavier elements. Small UN anomalies exist in both Mg and Si as well, as discussed in the papers on which Figure 2 is based. We will return to these after arguments about the isotopic mass fractionation process, which is our major concern. Its explanation is the key to what follows.

### III. THE PHYSICAL BASIS

The UN anomalies in the heavier elements seem unlikely to be the result of the mass fractionation process. They are, more likely, the result of a mixing process. Our arguments (Clayton 1978b, c, 1979a) that *s*-process/*r*-process polarizations in the FUN inclusions are the end result of initial gas/dust separations strengthen this conclusion. Because it is uneconomical to require two processes for the differences among these inclusions, we take it that the lines AKC must also be mixing lines. We regard this criterion of logical simplicity as a clue to what has occurred. How then shall it be arranged that mixing lines have slope  $m = \frac{1}{2}$ , so as to emulate mass fractionation? This can result naturally if the two end members of the mixing line are components that have previously been prepared by mass fractionation. This is no play on words by the discoverers of the FUN anomalies, because we maintain that the fundamental physical process is a mixing process rather than a mass fractionation of normal material. In what follows we present a new argument which shows that isotopic mass fractionation exists between dust and gas in the interstellar medium, even for refractory elements, and that its reaccumulation should lead to FUN accumulates.

Supernova explosions maintain a constant barrage of shock waves in the interstellar medium (Cox and Smith 1974). These connect via hot tunnels through the denser portions of the ISM (Smith 1977). McKee and Ostriker (1977) have used the physics of this to construct a three

phase model of the ISM in which most of the volume consists of a hot interstellar medium (HIM) maintained by this barrage. Spitzer (1976), Jura (1976), Barlow and Silk (1977), Shull (1977, 1978), Barlow (1978*a, b*), Cowie (1978), Draine and Salpeter (1979*b*), and Dwek and Scalo (1979, 1980) have discussed the associated destruction of interstellar dust. The mixture of gas and dust is heated and compressed when impacted by the supernova shock wave. Barlow and Silk (1977) estimated a mean lifetime of  $3 \times 10^9$  years for silicate grains against thermal sputtering, for example, whereas Draine and Salpeter (1979*b*) estimate a shorter life of order  $10^8$  yr. In shock waves a large relative velocity between gas and dust is established by the much greater inertia of the dust. This velocity is comparable to the shock velocity. These are describable by a spectrum of shock velocities ranging from several hundred kilometers  $\text{km s}^{-1}$  to tens of  $\text{km s}^{-1}$ . The grains will be badly sputtered while they move at such velocities relative to the gas. When they have slowed by gas drag to roughly  $20 \text{ km s}^{-1}$ , sputtering loses its efficiency. Most of the sputtering actually occurs in the warm ionized medium on the edge of the HIM.

Dwek and Scalo (1979, 1980) have used this picture to quantitatively evaluate its effect on the depletion of refractory elements from the gas phase. They argue that the sputtering is so effective that it becomes hard to even understand the large depletions of many refractory species. It will be assumed in the present work that their conclusions are, though skillfully arrived at, too extreme. They did not at first take into account the mantle structures expected around refractory cores. Dwek and Scalo (1980), in a more elaborate version of their work, have shown that mantle shielding can greatly increase the depletions. They take sputtering efficiencies for all refractory elements that differ only by the sublimation energy of the compounds, however; therefore, they reveal only minor differences in the sputtering of, say, Ca and Mg. The major physical expectation overlooked by them, however, is that magnesium silicate mantles are likely to surround Ca- and Al-rich cores (e.g., Clayton 1978*a*, 1980*a*). The Ca-Al-rich cores are the first to form in the supernova expansion. Owing to their residence in refractory-mineral grain cores, Ca, Al, and Ti may be largely shielded from this sputtering, whereas most of Mg and Si are returned to the gas by their more efficient sputtering from the magnesium silicate refractory mantles. This chemical description is not as arbitrary as it at first sounds. This writer (Clayton 1978*a* and references therein; Clayton 1980*a*) has repeatedly argued that the different chemical siting within the ISM of the pair Mg and Si from the more refractory Ca and Al is expected and influences their differential depletions from the gas. This difference was deduced prior to the considerations of this paper, and is not ad hoc. It also influences their sputtering rates. Figure 3 illustrates this expectation schematically. The Mg and Si-bearing mantles (characterized as  $\text{MgSiO}_3$ ) grow around the Ca-Al-rich SUNOCONS for two reasons: (1) later SUNOCON condensation of a portion of Mg and Si; (2) interstellar deposition of gaseous Mg and Si onto refractory cores. Later sputtering

returns Mg and Si, but not Ca and Al, to the gas, explaining why the Ca and Al depletions remain so large even though the Mg and Si depletions are smaller and more variable.

What we next emphasize is that this same sputtering will leave the heavier isotopes enriched in the grains. By necessity the light isotopes are enriched in the gas. We may therefore think of the HIM as containing two isotopically distinct pools, dust and gas. These are suggested by points D and G in Figures 1 and 2. From the expectation that 50–90% of Mg and Si are sputtered from the grains (Dwek and Scalo 1979) one can estimate that the isotopic fractionation will also be large—typically 1% per atomic mass unit.

The calculation of the isotopic fractionation can proceed directly from published accounts of astrophysical sputtering rates. Both Barlow (1978*a*) and Draine and Salpeter (1979*a*) have discussed the low-energy sputtering yields relevant to grains in the interstellar medium. The former paper fits a linear formula to the experimental data to derive an empirical “sputtering yield slope,” but contains no discussion which would permit one to estimate the dependence of the sputtering yield on isotopic mass. The latter paper is more useful in that regard, because it provides a semiempirical formula for low-energy sputtering yields which shows explicitly the dependence on the masses of both projectile and target. Near threshold, that sputtering yield can have a large dependence on the mass of the target atoms. As a specific example, consider the differential sputtering of  $^{24}\text{Mg}$  and  $^{26}\text{Mg}$  by 200 eV He ions ( $v = 100 \text{ km s}^{-1}$ ). With an assumed binding energy  $U_0 = 6 \text{ eV}$  for Mg atoms, their equation (31) predicts for the ratio of sputtering yields  $Y(^{24}\text{Mg})/Y(^{26}\text{Mg}) = 1.098$ . This is a large, 10%, isotopic enhancement of this ratio in the atoms returned to the gas. The sputtering skin depth is thereby made progressively more  $^{26}\text{Mg}$ -rich. Eventually a steady state is reached wherein the sputtered skin depth is so  $^{26}\text{Mg}$ -rich that the Mg sputtered away is returned to normal isotopic composition. One then envisions a  $^{26}\text{Mg}$ -rich layer of fixed isotopic composition eating its way into the continuously ablating surface. The isotopic composition of that skin depth must be the inverse of the sputtering yield ratio in order that a divergenceless isotopic flow be established. Thus one expects about a 10% enrichment of the  $^{26}\text{Mg}/^{24}\text{Mg}$  ratio in the skin, with, of course, a roughly 5% increase in the  $^{25}\text{Mg}/^{24}\text{Mg}$  ratio. The skin depth itself is roughly the range of a 200 eV He ion in this example, which, from equation (18) of Draine and Salpeter (1979*a*), is  $R \approx 12 \text{ \AA}$ . A similar result is obtained from the bombardment by H, C, N, O, etc., ions, but at the present state of knowledge this estimate from He ions suffices to establish the expected order of magnitude of the effect.

If the particles have radius  $a$  greater than the skin depth, the fraction contained in the isotopically enriched skin is approximately  $3R/a$ , leading to a bulk  $^{26}\text{Mg}$  enrichment

$$^{26}\delta = \left[ \frac{Y(^{24}\text{Mg})}{Y(^{26}\text{Mg})} - 1 \right] \frac{3R}{a} \approx 0.035 \left( \frac{100 \text{ \AA}}{a} \right).$$

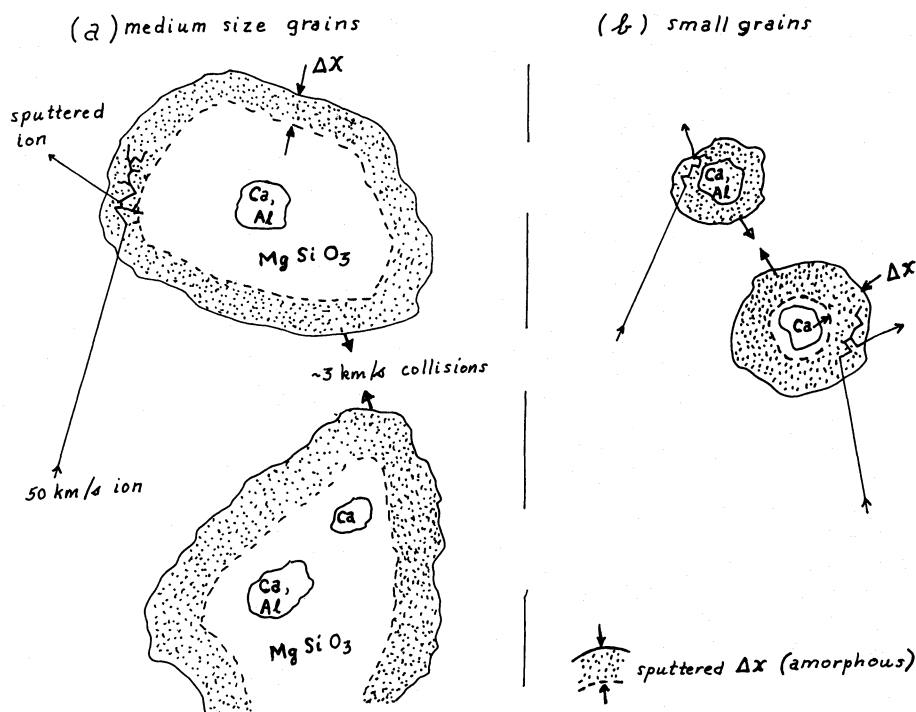


FIG. 3.—Because they are the first supernova condensates, the Ca-Al-rich SUNOCON cores are surrounded by Mg and Si bearing mantles shown here as  $\text{MgSiO}_3$ , which protect Ca and Al from interstellar sputtering. The cores initially carry the daughters of extinct  $^{26}\text{Al}$  (Clayton 1975b, 1977a) and of extinct  $^{41}\text{Ca}$  (Clayton 1975b, 1977c), leaving ghosts in the remainder of Mg and K. The Mg, Si, and O in the mantles is severely sputtered, so that an amorphous sputtered layer  $\Delta X$  progressively enriched in heavier isotopes of those elements is established. Its thickness depends on the historical sequence of sputtering and interstellar deposition, which is cyclic and which can produce a  $\Delta X$  much greater than the stopping length of individual ions. Accumulates of (a) larger grains and (b) smaller grains can differ in the mass fraction of the sputtered component. An important question is whether collisions between such grains throughout galactic history can have disrupted the mantles and fused larger than average grains that are Ca-Al-rich, because the latter can be gathered later by sedimentary processes (Clayton 1980a).

For a particle of size  $a = 100 \text{ \AA}$  the bulk enrichment  $^{26}\delta = 3.5\%$  is quite comparable to the observed enrichments in the FUN inclusions shown in Figure 2. Similar results are obtained for the O and Si in the skin, so that a collection of heavily sputtered grains is a plausible way to obtain a bulk aggregate of many grains with compositions like the FUN inclusions.

We note that a relevant situation exists in the lunar soils, which reveal large isotopic fractionation on their surfaces owing in part to sputtering by ions of the solar wind. (Whether sputtering is actually the dominant cause is controversial.) Haff *et al.* (1977) have summarized a model for sputtering that they believe applicable in such a case; however, a somewhat different physics is needed for sputtering of small grains in space. The small sizes of interstellar grains suggest that the fractionated layer can be a major fraction, even all, of those grains. Lighter isotopes have greater range at a given energy and may also assume higher recoil energies than heavier isotopes. In low-velocity sputtering, which should be ubiquitous in the turbulent HIM, the preferential escape of lighter isotopes seems therefore quite certain.

Low-energy sputtering is also a source of chemical

fractionation. When  $\epsilon \approx \epsilon_0$ , equation (31) of Draine and Salpeter (1979a) becomes

$$Y(E) \approx 2A\epsilon_0(\epsilon - \epsilon_0),$$

where  $\epsilon = \eta EU_0^{-1}$  and, for He ions bombarding heavy targets,

$$\eta = 16M_T(M_T + 4)^{-2}.$$

This also takes the form  $Y \propto (E - E_T)U_0^{-1}$ , where the sputtering threshold is either  $E_T = 4U_0$  or  $E_T = U_0\eta^{-1}$ , as discussed by Draine and Salpeter. Increasing binding energy  $U_0$  drives the low-energy sputtering yield down for more refractory atoms (molecules). These features play a role, but not the only role, in moderating the steady depletion factors from the interstellar gas. The major extra factor in the depletion of the very refractory elements is the layered structure of the refractory component as discussed above and illustrated in Figure 3. Thus it seems quite possible that the element fractionations according to volatility that are evident in meteorites derive from the memory of interstellar processing rather than the postulated hot condensation sequence in the solar system. It requires that sputtering near threshold be

much more common than high-energy sputtering, which is chemically nonselective.

Another problem needing study for chemical and isotopic fractionation is grain-grain collisions. Grains of  $100 \text{ km s}^{-1}$  slow down largely by gas drag (Spitzer 1968) rather than by grain collisions. When grain collisions do occur, however, the chemical effects may be significant. Ca-Al-rich SUNOCON cores may survive the fragmenting collisions while Mg and Si in surrounding mantles can be largely evaporated, also with significant mass fractionation among the isotopes. Such micrometeorite bombardment seems in fact likely to be responsible for isotopic fractionation in the lunar surface (Clayton, Mayeda, and Hurd 1974). We must also bear in mind that turbulent velocities of order  $10 \text{ km s}^{-1}$  may be ubiquitous in the HIM and in the warm phases. Such velocities are below the sputtering threshold but would be quite significant in the chemistry of grain collisions. *A relatively small fraction of Ca and Al may build up by this process to larger accumulates rich in Ca and Al.* Selective agglomeration (Clayton 1980a) of these can be the seeds of the CAIs, as first set forth in much less specific terms in Paper I. A complete dynamic description and history of the ISM will be needed to clarify this problem quantitatively. One cannot easily discount the existence of dust clouds within the HIM, because dynamic diffusion processes may establish such regions (Harrison 1978), even in a turbulent medium. If grain-enhanced regions exist, fast-moving grains driven into them might be expected to encounter grain-grain collisions. Draine and Salpeter (1979b) have shown that those are important in cloud-cloud collisions. Another possibility is in the dense cloud, where the grains may collide with ambient grains, followed by the whole cavity dumping into the HIM for subsequent sputtering. Scalo (1977) showed that turbulence in dense clouds makes grain-grain collisions significant there. When we speak subsequently of mass fractionation, we intend high-velocity collisions to be included. We will not explicitly consider other classes of suprathermal grains (Tarafdar and Wickramasinghe 1977) except to note that sputtering and collisions may affect them similarly.

If this description is approximately true, the points A, K, and C of Figures 1 and 2 are quite easily generated by different mixtures of D and G, with K and C being more dust-rich than point A. The relevance to other isotopic anomalies follows directly. Gas/dust separations should generate isotopic anomalies in virtually all heavy elements owing to the differential siting of different isotopic patterns in different chemical forms (Clayton 1978a and references therein). Most straightforward are the differential sitings of *s*-process isotopes and *r*-process isotopes in dust and gas, first pointed out in 1975 in the preprint of Clayton and Ward (1978). But also significant may be different forms of dust (Clayton 1978a), so that correlations between chemical form and size can be included in the differential sputtering and reaccumulation phases generating samples on the DG line. The unknown nuclear anomalies in inclusions EK and EC (Lee, Papanastassiou, and Wasserburg 1978) may be of the latter form. It seems likely that the refractory Ca minerals in grain cores

will be less sputtered than will the mantles, so that Ca will be expected to remain much more depleted and less fractionated, as observed in inclusions EK and EC. But different Ca-bearing SUNOCONS (Clayton 1978a) may be differentially affected by the sputtering, leading also to nuclear Ca anomalies even though the mass fractionation is small for Ca. The key to such possibilities is the expectation that different chemical forms of interstellar dust have different isotopic compositions (Clayton 1978a).

#### a) Variations Based on Grain-Size Separates

Before proceeding with the dust (D)-gas mixtures of Figures 1 and 2, it may be noted that other physical models based on the same astrophysical context can also be advanced. Consider grain-size separates, idealized for clarity into two populations: grains and small grains. If they are equally sputtered, the surface thickness  $\Delta X$  of isotopically fractionated material constitutes a larger fraction of the mass of the small grains than of the grains. Figure 3 illustrates this schematically. The point D then represents the small grains. The mixing line AKC could then be different mixes of grains and small grains instead of different mixes of dust and gas. The associated nuclear anomalies can also follow naturally, because different classes of grains are expected to differ isotopically (Clayton 1978a). For example, it could easily be true that *r*-process SUNOCONS are on the average smaller grains than STARDUST (Clayton 1978a) carrying the *s*-process condensed component, or vice versa. Similarly, the Ca-bearing SUNOCONS from a Si-burning supernova shell should be expected to be on average of different size (and, of course, different chemical constitution) from the Ca-bearing SUNOCONS from the C-burning shell. Furthermore, the different chemical carriers may sputter differentially, as mentioned earlier. In consideration of these and other alternatives the reader will appreciate that what is being advanced is a new astrophysical context that may establish a connection of significance for astronomy rather than a firm fix on the specific details. It is the entire picture of the interstellar FUN correlation that is new, not its specific components.

The foregoing variation based on grain size imagines collecting dust before the gas has been accreted onto that dust—a sequence difficult but not impossible to achieve. So versatile are astrophysical conditions, however, that, even if the gas is first accreted, a fractionation according to grain-size separates should remain. Clayton (1980a) has recently outlined how this occurs and how it is that in this case it is the largest, rather than the smallest, grains that are progressively enriched in the heavier isotopes. This common expectation is of such significance to astrophysical chemistry that its main features must be discussed here. When material is sputtered away from grains, and if *the same material* is subsequently reaccreted by those grains, no isotopic fractionation results. That null result follows because both processes are proportional to the surface areas of the grain population. However, if *extra material* from the gas is added during the accretion process, an isotopic fractionation will in this

case be obtained, specifically because the grain sizes grow during reaccretion differently than they were reduced during the sputtering. Clayton (1980a) presents the following illustrative numerical model: a size distribution  $dN/da_0 \propto a_0^{-3}$  of the first Al SUNOCONS depletes half of the accompanying MgO in proportion to surface area, following which half of the MgO shell thickness is sputtered away, leaving the residues enriched by 2% in heavy Mg isotopes. When all gaseous Mg is then accreted, the final fractionations range from +0.5% in the largest grains to -0.1% in the smallest. The importance of this example calculation is neither its detailed assumptions nor its numerical results, but rather the general expectation of the migration of isotopic patterns in grain-size carriers under conditions of grain sputtering followed by redeposition.

The situation is even more dramatic when one relinquishes the assumed homogeneity of the grain population. For example, Mg can be sputtered away from grains as in Figure 3 during interstellar shocks but later accreted by a *separate population* of numerous smaller grains (e.g., graphite). A severe polarization of fractionated isotopic patterns then coexists between the two grain populations. There is no need in the exposition of principles to postulate which astrophysical condition has been most relevant.

The same principles apply to chemical fractionation. After more volatile elements have been sputtered away from grains in intercloud media, they will be reaccreted in cooler phases by grains in proportion to their numbers and surface areas. This process acts cyclically during repeated interstellar phase transitions, mapping volatility patterns onto the grain-size pattern (Clayton 1980a). The grain sizes (and/or densities) can be later sorted by dynamic forces, leading to macroscopic chemically fractionated accumulates.

#### b) Macroscopic Accumulates

Both the severe chemical and isotopic fractionation between gas and dust and the nuclear anomalies in various types of dust occur on microscopic scales. For an astrophysical model of the anomalous inclusions of meteorites, it is necessary to preserve these anomalies at some subsequent time in bulk accumulates that provide the parents of the inclusions. Studied inclusions from meteorites, primarily Allende, are millimeter to centimeter in size. Gas-dust separations are one of the most plausible physical ways to achieve macroscopic differences. Accumulation of distinct grain-size separates accomplishes this in a simple way. One requires accumulation of dust and gas into macroscopic accumulates at various times and places in the ISM and ultimately after sedimentation to a solar disk. It is in these macroscopic accumulates and the dust/gas separations that characterized their accumulation that the isotopic patterns are preserved. The microscopic chemistry within each accumulate will be changed later when the actual inclusions are mineralogically set in the solar system. But the bulk isotopic signatures may be preserved in the different inclusions.

It is probable that these accumulations occur *before* the parents of the inclusions (accumulates at A, K, C) become part of a hot solar disk. It may be there that they exchange oxygen with a reservoir at or near point E of Figure 1. It seems simplest to assume that E characterizes gaseous oxygen in the heated solar accretion disk (but see Clayton 1980b). Its oxygen will be expected to differ from that of the dust aggregates. This exchange with E in the solar system generates the mixing lines EA, EK, and EC in Figure 1 in the same way already proposed by Clayton and Mayeda (1977). We introduce no extra features of this oxygen exchange except for a specific suggestion of the identity of the oxygen pools being exchanged. The initial dust aggregates are more  $^{16}\text{O}$ -rich than the gas because of their SUNOCON component (Clayton 1977b, 1979b). The fact that spinels within the final inclusions are the most  $^{16}\text{O}$ -rich minerals after the exchange is not explained by us or by Clayton and Mayeda (1977), but we adopt their suggestions as reasonable for these mixing lines. The thrust of the present work is the interstellar dynamics involved in the preparation of the samples before the oxygen mixing. There are no analogous mixing lines for Mg and Si in Figure 2. This may be easily understood on the present picture, because by the time the solar accretion disk had formed, there remained no more Mg and Si in the gaseous phase. Gaseous Mg and Si will have been accreted onto the grains much earlier, after the HIM condensed to enter a cooler phase. The solar oxygen at E is much different from that at G that had been sputtered away from the grains. The sputtered oxygen atoms G must be mixed with the remainder of gaseous oxygen to produce E.

Figure 4 gives a rough schematic of the overall scenario. The HIM probably endures about  $10^7$  years against phase condensation into the warm phases (WIM and WNM of McKee and Ostriker 1977), which must themselves live longer as evidenced by their greater mass ( $\tau \propto M$ ). Gaseous Mg and Si are depleted onto the Ca-Al-rich grain accumulates either during denser cooler filaments or clouds within the HIM or later during the residence in the warm medium, which may be stable for as long as  $10^9$  years before evaporation into the HIM or further condensation into a cool cloud. This depletion of gas mixes most DG accumulates to a point near A, but allows some accumulates having accelerated accumulation histories to mix only as far as point K and C. These dust-rich accumulates may be mapped onto collections of large grains (Clayton 1980a), but it is not immediately necessary to specify how the dust-rich accumulate comes about. In these dust-rich accumulates not only will O, Mg, and Si appear heavily fractionated, but isotopic anomalies derivable from dust/gas enhancements will of necessity be admixed as well. These accumulates provide the bulk parents of the CAIs. Their detailed mineralogy will still be altered by the exchange with solar oxygen in the mildly hot solar accretion disk. One must not expect that the actual CAI mineralogy is established by astrophysical history. There will no doubt appear several ways of improving both the astrophysics and the chemistry of this scenario, and we offer Figure 4 not as a



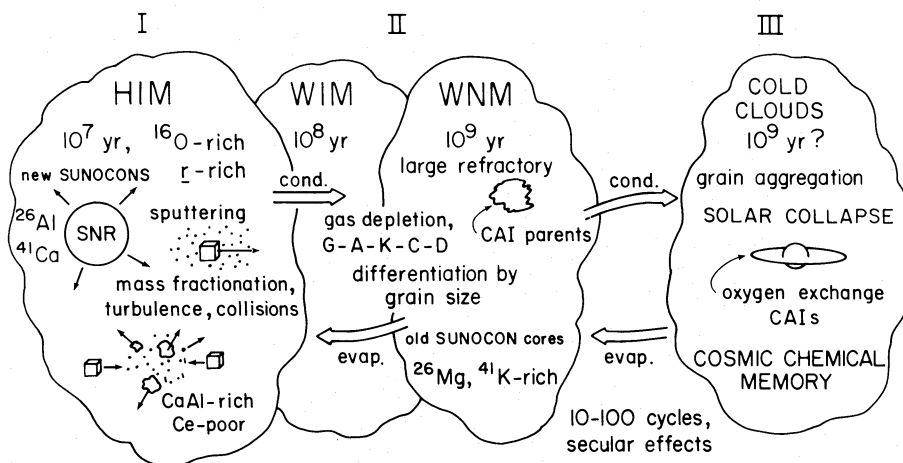


FIG. 4.—A scenario for generations of CAIs in the three-phase ISM is schematically illustrated. Supernova remnants (SNR) expand into a connected HIM, where sputtering and turbulent collisions generate the dust-gas isotopic fractionations along with Ca-Al-rich refractory dust. Most of the sputtering actually occurs in what was the WIM on the borders of the HIM, but the subsequent hot shocked gas is here regarded as joining the HIM. The HIM turnover time is taken as  $10^7$  years against condensation onto clouds through the surrounding warm media (WIM and WNM), which are also replenishing the HIM by evaporating elsewhere. The basic model follows McKee and Ostriker (1977). Mixing with isotopically light gas occurs as it is redepleted onto sputtered grains, probably in the transitional warm media, where accumulation into larger units incorporating volatiles and *Fremdlinge* occurs. These parents of the CAI differentially exchange oxygen when heated in the solar system.

final theory, but as the direction in which the final theory may be sought.

The primary chemical problem of accumulation remains that of achieving a macroscopic inclusion that is Ca-Al-rich with respect to Mg-Si by an order of magnitude. One must not underestimate the importance of this problem for astrophysics. The recently discovered existence of CAIs was not predicted by any theory. Only three explanations known to this writer have been proposed, none of which would seem plausible except for the existence of these inclusions, which are so abundant that they contain from half to all of the Ca-Al within the type C3 meteorites! The proposals are:

1. Solar system material was once a hot gas, which upon cooling first condensed Ca and Al (Larimer and Anders 1977; Grossman 1972). These refractory suspensions must then be gathered from the hot gas before it cools sufficiently to condense Mg and Si. The physical implausibility is the requirement of an outer solar system that is so hot that even the dust is heated to  $T > 1500$  K in order to vaporize the Mg thermally. Cameron (1978) has described the energetic difficulty of vaporizing dust in the outer solar system.

2. The existence of Ca-Al-rich SUNOCON cores within interstellar dust has been predicted and offered as an explanation for the vastly greater depletions of these elements from interstellar gas (Clayton 1977a, c, 1978a). The proposal (Clayton 1977a) for CAI origin was that high-speed ( $1-10 \text{ km s}^{-1}$ ) grain-grain collisions could fracture and disperse the mantles in Figure 3 but fuse the Ca-Al-rich cores. The physical problems are those of obtaining core amalgamation, which was merely postulated, and in obtaining enough grain-grain collisions even if core amalgamation is probable during individual collisions.

3. Grains of differing size and core constitution fractionate Mg and Si from Ca-Al during depletion processes proportional to grain surface area (Clayton 1980a). This previously overlooked mechanism is plausible and effective. It works in two basic modes that are important for the present discussion: (a) in the supernova expansion itself a large number of MgO molecules nucleate a month or so following Al condensation and collect the Mg in separate small Al-free MgO grains; (2) later, in the ISM, gaseous Mg is collected by a large number of small particles of another kind (e.g., graphite), so that sputtering of Mg mantles slowly produces a mix of Al-rich grains plus Al-free grains (with respect to Mg). Clayton (1980a) argued that the grains having the largest Al cores remain both the largest and the most Al-rich, and that they can later be accumulated preferentially by physical processes (e.g., sedimentation) that collect the largest grains. Cosmic sieves according to grain size are not implausible.

Although none of these explanations is at present totally plausible, we regard the third as the most promising. It offers for the first time a simple mechanism for preserving gas/dust fractionation patterns *even after the gas has been accreted by the dust*, because gas is preferentially accreted (per unit mass) by the smallest particles. Therefore, a collection of the smallest grains is the most volatile-rich collection. This sorting mechanism can be the key to the several degrees of element fractionation according to volatility as observed in the meteorites—not only for Al/Mg fractionation. Within this proposal, grain-grain ISM collisions can also play an important role if they can in fact fuse Ca-Al-rich cores as advanced in proposal (2). Although high-speed collisions are generally disruptive for grains, it remains to be seen whether collisions at moderate speeds occur in any of the common cyclical phases of the ISM, and whether these moderate-

speed collisions can in fact fuse the refractory-rich parts of these grains. In that case the evolution of the Galaxy could be accompanied by a *secular growth* of the sizes of the Ca-Al-rich cores resulting from  $10^{10}$  years of collisions. They may produce a population of larger-than-average very refractory grains. That is all that is required. Following sputtering and redeposition of Mg and Si onto smaller grains, this population of large Ca-Al-rich grains will become increasingly depleted in Mg and Si. Dynamic processes (e.g., sedimentation) can later sort out and collect these large Ca-Al-rich particles with their *relatively less massive* mantles.

A major new development of relevance for this problem is the possibility that the release of internal chemical energy rapidly and spontaneously heats the parents of the CAI (Clayton 1980*b*). The chemical energy results from the severe sputtering of interstellar dust and from the highly disequilibrated assortment of atoms and molecules that are frozen onto the dust during the cold-cloud state. This chemical energy will be greatest in the mantles coating the SUNOCONS. When the accumulate finds itself warmed to the point that chemical reactions can begin, a thermal runaway may fuse the SUNOCON collection into larger Ca-Al-rich minerals. These minerals would resemble molten minerals, as the CAI minerals do, but they would contain highly un-equilibrated trace chemistry, as the CAI minerals also do. Proof of the latter is most dramatically evident in the work of El Goresy, Nagel, and Ramdohr (1978), who find bizarre small foreign bodies (*Fremdlinge*) enclosed within the Ca-Al-rich minerals, and who also find that in an anorthite crystal ( $\text{CaAl}_2\text{Si}_2\text{O}_8$ ) the trace Mg and Na are very inhomogeneously distributed. The discovery of anticipated chemical energy strengthens the plausibility of building CAI from special interstellar accumulates without need of a hot solar system, even though the details of the postulated chemical energy are completely unknown at the present time. But at least this way of fusing the SUNOCONS seems sounder physically than the high-speed collisions that had to be advanced in Paper I for this purpose.

Lest this discussion seem unfocused, be reminded that the CAIs are doubly implausible. Both the Ca-Al richness and the severe mass fractionation in the FUN inclusions pose severe accumulation problems and cannot be regarded singly. By demonstrating that the major features are contained qualitatively if not quantitatively within accumulations of interstellar dust, we are adding support to our thesis that the CAIs are end products of galactic evolution rather than of ad hoc solar system scenarios. Neither the postulate of a hot gaseous solar system nor that of an admixture from a neighboring supernova seems to confront squarely these two coordinated and overriding problems.

### c) $^{16}\text{O}$ Richness

Another dominant feature of the CAIs is their  $^{16}\text{O}$ -richness, which its discoverers (Clayton, Grossman, and Mayeda 1973) attributed to the survival of some  $^{16}\text{O}$ -rich presolar grains within an otherwise hot and

gaseous nebula. These  $^{16}\text{O}$ -rich presolar grains are not likely to be explained by the bulk  $^{16}\text{O}$ -richness of the HIM because the HIM is too dilute for its gaseous and solid oxygen to equilibrate. It seems more likely that the  $^{16}\text{O}$ -richness is a direct consequence of the SUNOCON richness of the parent accumulates. Previously (Clayton 1975*a*, 1977*b*, 1979*b*) we argued that  $^{16}\text{O}$ -rich SUNOCONS could not escape condensing during the supernova expansion, and that they carried the  $^{16}\text{O}$  excess. That picture survives sputtering if the Ca and Al grain cores were initially SUNOCONS and if primarily the Mg, Si, and volatile mantles participated in the mass fractionation by sputtering, as suggested by Figure 3. It is quite possible for the two-component mixing line DG to actually be three-component and still be linear if the  $^{16}\text{O}$ -rich component is a fixed fraction of the dust that mixed DG. To see this, one need only consider as an example a mixture of normal dust and  $^{16}\text{O}$ -pure dust, noting that if it is then sputtered in such a way as to remove the same fraction of oxygen from both types of dust, the resulting mix lies along an  $m = \frac{1}{2}$  line lying below the terrestrial line (as in Fig. 1). For the main purpose of this paper it is not necessary to exactly identify the roles of the  $^{16}\text{O}$ -rich SUNOCONS, although this is an ultimately interesting question for this model. The grains injected into the HIM may in first approximation be thought of as of two types—grains carried into the HIM from the clouds by their evaporation, and new SUNOCONS injected by the supernovae that energize the HIM. The cores of the old grains moving into the HIM are in part old SUNOCONS. An analysis of their relative fates will eventually be essential, because the collinearity of AKC is by no means guaranteed in this picture, even though it is plausible.

It is also clear that oxygen exchanges by the sputtering process will dilute the pure  $^{16}\text{O}$  that may have been injected within SUNOCONS. As a fast silicate grain slows from 100 to 10  $\text{km s}^{-1}$  it will collide with almost half as much oxygen as the total oxygen content of the grain. Although interesting, analysis of this problem can wait.

## IV. RELATED SIMILARITIES TO METEORITES

The physical idea outlined in § III has a large number of fascinating consequences. Many of these are rich payoffs that strengthen this interpretation. Others remain problems. In what follows, we consider the most exciting of these astrophysical consequences in just enough detail to establish their similarity to features actually observed in the FUN inclusions. This approach is consistent with the ground-breaking nature of this effort. Our intent is not so much to quarrel over the fine details as it is to illustrate the fabric of this interpretation.

### a) Isotopic Differences between HIM and Clouds

The HIM could be  $^{16}\text{O}$ -rich in bulk. Were that the case, the later deposition of oxygen gas on growing grains could generate a  $^{16}\text{O}$ -rich mixing line. The truth or falsity of this particular conjecture hinges on these plausible suppositions: (1) supernovae are sites of nucleosynthesis, and the ejecta goes primarily into the HIM; (2) new

production (of  $^{16}\text{O}$ , for example) is a detectable fraction of the rate of injection by cloud evaporation; (3) the ejecta has a recognizable isotopic signature, which is true only if all isotopes do not originate equally in supernovae ( $^{17}\text{O}$  and  $^{18}\text{O}$  production in red giants in the clouds, for example). None of these suppositions is very secure astrophysically, but simple numerical estimates show that they may be consistent with contemporary dogma.

The HIM might similarly be rich in  $r$ -process isotopes of the heavy elements. Accretion onto grains before the HIM has been remixed into the cold cloud (phase III of Fig. 4) would generate isotopic patterns that are rich in the *average*  $r$ -component, rather than in a bizarre  $r$ -component. Clayton (1978*b*, 1979*a*) showed that the experimental discoveries indicated just such an average excess, although he had to use nucleosynthetic criteria for choosing mass fractionations different from those advocated by the discoverers (McCulloch and Wasserburg 1978; Lugmair, Marti, and Scheinin 1978). For Nd and Sm, the fit is too convincing to be an accident. For Ba the fit is not good, but Clayton (1978*b*) already indicated the direction of the solution that fits into the present scenario. The radioactive progenitor  $^{135}\text{Cs}$  ( $\tau_{1/2} = 2 \times 10^6$  yr) holds up that isobar in the HIM, so that depletion as Cs onto grains within  $10^7$  years may lead to different chemical siting of this isotope of Ba. The negative  $^{135}\text{Ba}$  anomaly in inclusion C1 is especially interesting in this regard, as its interpretation has been a problem of quite different magnitude for advocates of a supernova admixture into the solar system (McCulloch and Wasserburg 1978). Clayton (1978*b*) specifically called for some extensively chemical processing of SUNOCONS between 30 years and  $10^6$  years after their ejection from the supernova, and this severe sputtering may no doubt suffice. Of course, the difference in the Ba anomalies between inclusions K and C (McCulloch and Wasserburg 1978) is not yet explained by a simple DG mixing line; but that can be regarded as a detail for the future, because our purpose here is not to present detailed models of the actual inclusions. This particular conjecture stands, in any case, apart from the main thesis of this paper.

#### b) Extinct Radioactives: $^{129}\text{I}$ Free-Decay Interval

The vast majority of supernova radioactivities will decay in the HIM if the supernova ejecta do reside there for  $10^7$  years. If that is the case, depletion there or in SUNOCONS directly may be needed to understand  $^{26}\text{Al}$  or  $^{107}\text{Pd}$  generated anomalies (Clayton 1979*b* and references therein). HIM residence, followed by warm ionized medium (WIM) residence, can explain why  $^{129}\text{I}$  has such a low level in solar system samples ( $^{129}\text{I}/^{127}\text{I} \approx 10^{-4}$  instead of the  $5 \times 10^{-3}$  value anticipated in the average ISM). Most workers have interpreted the long decay interval ( $10^8$  years) needed before formation of solar samples in other ways (see Clayton 1979*b* for a review of those ideas). *But if  $^{129}\text{I}$  remains gaseous in the HIM and WIM, depleting only in the warm neutral medium (WNM), this new explanation for the free-decay interval may be more plausible.* It eliminates the fresh  $^{129}\text{I}$  by requiring it to wait  $10^8$  years in a hot and dilute physical phase where-

in condensation into solids is, for iodine anyhow, repressed. Barlow and Silk (1977) showed, for example, that ice would be removed by thermal sputtering in  $10^5$  years in a gas having  $n_{\text{H}} = 1 \text{ cm}^{-3}$  and  $T = 10^5 \text{ K}$ . Clayton (1980*a*) shows how eventual depletion in the WNM in proportion to grain surface area leads, after sedimentation, to bulk  $^{129}\text{I}/^{127}\text{I}$  variations much larger than are to be expected for isotopic anomalies in other elements. This analysis allows one to interpret the "age differences" as isotopic anomalies that are understandably large for macroscopic bodies. It offers hope for understanding the apparent temporal irrelevance of I- $^{129}\text{Xe}$  "ages" of meteorites (Jordan, Kirsten, and Richter 1980).

#### c) $^{28}\text{Si}$ Deficiency in HIM Dust

Figure 2 shows not only that the Si is strongly fractionated in the FUN inclusions, but that 1:2 ratio is not exactly maintained. The experimental papers must be consulted to see the large statistical significance of the small deviations from the 1:2 line. Yeh and Epstein (1978) and Clayton, Mayeda, and Epstein (1978*a*) regard this deviation as a UN anomaly. They tentatively speak of it as a  $^{29}\text{Si}$  excess, although they note that it could be equally well a deficiency of  $^{28}\text{Si}$  or  $^{30}\text{Si}$ . All such solutions are possible in the present model, but we here point out that a  $^{28}\text{Si}$  deficiency is probably the most plausible. It is even predicted by the model! Clayton (1977*a*, 1978*a*) has already emphasized that  $^{28}\text{Si}$  should be more gaseous than  $^{29,30}\text{Si}$  because of their greatly differential condensation in SUNOCONS. The  $^{29,30}\text{Si}$  from the carbon shell is easily condensed into SUNOCONS by the more abundant Mg and O there, but the  $^{28}\text{Si}$  from the oxygen burning is too abundant to be condensed into any common mineral. The HIM gas is logically  $^{28}\text{Si}$ -rich, therefore. The condensed mantles in the HIM are therefore expected to be mass fractionated and  $^{28}\text{Si}$ -deficient. Both anomalies are, of course, eliminated in bulk dust by DG mixing to point A, where the gaseous Si has been depleted.

#### d) Fractionated Mg and $^{26}\text{Al}$ Ghosts

Wasserburg, Lee, and Papanastassiou (1977) emphasized that the heavily fractionated inclusions EK1 and C1 show no evidence of *in situ*  $^{26}\text{Al}$  decay—a major puzzle onto which new light now falls. In fact, when normalized to unfractionated  $^{25}\text{Mg}/^{24}\text{Mg}$ , the  $^{26}\text{Mg}$  reveals a fractional deficiency of  $3.5 \times 10^{-3}$  in EK1 and of  $1.8 \times 10^{-3}$  in C1. In the present discussion, the fractionated magnesium is the heavily sputtered Mg remaining in the Mg-rich mantle surrounding Ca-Al-rich cores (Fig. 3). Dwek and Scalo (1979*b*) unintentionally show that these Ca-Al-rich cores are primarily SUNOCONS, as Clayton (1977*a*) emphasized, because the persistently large depletions of Ca and Al from the gas contradict their conclusion that the SUNOCONS are sputtered away. We take it that the SUNOCONS are shielded by the mantles dominating the sputtering exchange; therefore, *both live  $^{26}\text{Al}$  and the excess  $^{26}\text{Mg}$  from its decay remain during the sputtering process in the Al SUNOCONS where they had been since nucleosynthesis.* The fractionated DG

pool therefore has a small negative  $^{26}\text{Mg}$  ghost, a concept introduced in Paper I for this chemical problem. The present work offers the first explanation of why strongly fractionated Mg should be ghostly.

The negative  $^{26}\text{Mg}$  anomalies (taken here to be real rather than a nonlinearity in the mass fractionation process) may be equally well explained, however, by assuming that they represent instead a  $^{24}\text{Mg}$  excess in the gaseous or very small-grained portion of the ISM. Such an excess could arise naturally if the  $^{25,26}\text{Mg}$  condenses into SUNOCONS in the carbon shell expansion more completely than the monoisotopic  $^{24}\text{Mg}$  does in the expansions of the neon and oxygen shells. Morgan (1980) describes the nucleosynthesis of Mg isotopes.

#### e) Other Strange Meteoritic Ca-Al-rich Samples

It is exciting that in C3 meteorites other common Ca-Al-rich minerals having small Mg/Al ratios have been found in the form of individual hibonite crystals ( $\text{CaAl}_{12}\text{O}_{19}$ ). Lorin and Michel-Levy (1978) found quite variable ratios of excess  $^{26}\text{Mg}$  to the Al/Mg abundance ratio, a surprise identifying hibonite crystals as being of great interest. Maccougall and Phinney (1979) have also studied large numbers of hibonite crystals from the C2 Murchison meteorite and find that some are fractionated, some are not, and some have excess  $^{26}\text{Mg}$  while some do not. The tendency again seems to be that excess  $^{26}\text{Mg}$  accompanies unfractionated Mg when it exists at all, whereas fractionated hibonites show no obvious  $^{26}\text{Mg}$  excess. The isotopic resolution is not yet adequate to be sure how such correlations will work in fine detail, however. We await higher resolution studies of these exciting samples. They may constitute the best statistical data on accumulation, because they are very numerous and varied. With increasing precision one may hope to identify families of hibonite crystals that reflect differing ISM histories.

The common assumption has been to assume either that  $^{26}\text{Al}$  was inhomogeneously distributed in the early solar system or that there are large time differences ( $> 10^7$  years) between solidification of the different Al-rich minerals. We have consistently rejected these interpretations in favor of those reflecting the galactic history of differing chemical phases. We would speculate that many Al-rich minerals, especially hibonites, contain no detectable  $^{26}\text{Mg}$  excess for one of these reasons: (1) some accumulates are assembled from dust in which the  $^{26}\text{Mg}/\text{Al}$  correlation has already been destroyed, either by very severe sputtering or by thermal evaporation in stellar atmospheres; or (2) the exothermic chemical heating (Clayton 1980b) leading to the fusion of SUNOCONS into macroscopic minerals was in some cases so severe that the resulting melt allowed ample time for the  $^{26}\text{Mg}$  spike to diffusively join the remainder of the Mg. The first alternative is rather awkward in its requirement of an inhomogeneity, whether spatial (which we have consistently tried to avoid appealing to) or in different grain-size categories, so that sedimentation, for example,

could separate them (Clayton 1980a). One could imagine, for example, that Al condensing during stellar mass loss (STARDUST: no  $^{26}\text{Mg}$ -Al correlation) may be significantly smaller than supernova condensates (SUNOCONS: large  $^{26}\text{Mg}$ -Al correlation). We favor trying to avoid such distinctions by adopting (2). In this view the existence, or should we say *persistence*, of  $^{26}\text{Mg}$  excesses in some minerals but not in others reflects the differences in the degree of isotopic equilibration during the different sudden heating events that fused in different ways, the minerals. The propensity for  $\Delta^{26}\text{Mg}/^{27}\text{Al} \approx 5 \times 10^{-5}$  in anorthite minerals is taken to have, therefore, a meaning similar to that of spinels having  $\Delta^{16}\text{O}/^{16}\text{O} \approx 4\%$  whereas plagioclase tends to have  $\Delta^{16}\text{O} \approx 0$  (Clayton and Mayeda 1977).

A very exciting hibonite dominated inclusion (named HAL for *hibonite Allende*) has been discovered by Lee, Russell, and Wasserburg (1979). It is unique in that even the Ca is strongly mass fractionated, as contrasted to the usual Ca resistance to fractionation. But the Mg is normal, and there is no excess  $^{26}\text{Mg}$  even in extremely high-Al/Mg hibonites. Oxygen is also strongly fractionated (Lee, Mayeda, and Clayton 1980). Can this combination be easily understood? Yes. The Ca will be strongly mass fractionated only if the sputtering and collisions were so severe in that region of the HIM as to destroy much of even the Ca SUNOCONS. In that case the Mg is totally vaporized, according to the mantle structure of Figure 3, so that it is by definition normal upon redeposition. Absence of  $^{26}\text{Mg}$  excess strengthens our conjecture (Clayton 1977a) that  $^{26}\text{Al}$  was not actually alive in the solar system. Since the  $^{26}\text{Al}$  was actually not alive, the daughter  $^{26}\text{Mg}$  excess does not correlate with Al if the hibonite forms from a molten solar object. Of course, any newly created SUNOCONS will have live  $^{26}\text{Al}$ . So it is necessary to distinguish between the sputtering of old grains, including SUNOCONS contained within material that was evaporated from the cold clouds, and the sputtering of newly synthesized SUNOCONS injected from the supernovae energizing the HIM. It seems not at all unreasonable to have live  $^{26}\text{Al}$  in SUNOCONS in some portion of the HIM, and dead  $^{26}\text{Al}$  in SUNOCONS in other portions. The expanding supernova interior lags far behind the fast moving envelope which sputters the older HIM. The new SUNOCONS are sputtered when they overtake the stagnated presputtered HIM. The unknown nuclear anomalies in the fractionated Ca within HAL may come about because the extensive sputtering of the Ca SUNOCONS does not sputter each SUNOCON type equally. Different Ca isotopic patterns reside within different Ca-rich SUNOCONS (Clayton 1978a). A bewildering variety of conditions is expected. We cannot sort them out in this exploratory work, and must be content to have outlined a new approach to this subject—an approach of more far-reaching astrophysical implications than the dynamically suspect concept of a hot gaseous solar system (Larimer and Anders 1977; Grossman 1972) and of the injection of isotopically anomalous matter into it from a neighboring supernova (Cameron and Truran 1977).

### f) Typical Supernovae

Arnett (1978) discusses the problem that the typical supernova is not the main contributor to nucleosynthesis. He finds that O stars are the major producers of new elements, but those are born 30 to 100 times too slowly to account for the observed supernova rate. The maintenance of a violent HIM is apparently accomplished by lower-mass stars, which do some very interesting nucleosynthesis, especially in trace nuclei, but do not eject large masses of C, O, Mg, Si, etc. We have not faced that distinction in this work, preferring to stick to the simplest possible picture in order to set forth new physical ideas. Nonetheless, spatial and temporal variations in isotopic and elemental compositions must characterize the HIM. Our grounds for suppressing such heterogeneities is the belief that they are not the essential factor in most of the isotopic anomalies, which result instead from chemical memory. The thrust of our work rests on the proposition that after a macroscopic accumulation of dust grains has been assembled, their individual variations are less important (microscopic fluctuations) than the systematic differences between chemical phases (Clayton 1978a). Our conclusion has consistently been that emphasis on *spatial inhomogeneities* is a counterproductive point of view until the dominant effects of chemical evolution in an astrophysical context have been analyzed. For this reason we strive to account *chemically* for the very existence of Ca-Al-rich inclusions in the early solar system, recognizing this as the *major* problem, with the FUN correlation and the  $^{16}\text{O}$  richness as the next most significant features. The spatial inhomogeneity models (Cameron and Truran 1977; Reeves 1978; all experimental papers) have enough advocates already, although they have not explained even the existence of the CAIs.

### g) Concluding Remarks

Probably the differences in supernovae are not as significant as the question of what happens to their ejecta. I take the view that the O stars blow out bubbles by winds first and that the supernova shock wave connects these into a common HIM via tunnels (Cox and Smith 1974; Smith 1977), so that the lagging ejecta find themselves overwhelmingly beginning their lives in the HIM. Cameron and Truran (1977) suggested instead direct mixing of a special supernova into the solar system. In his "bing bang" model, Reeves (1978) reasons that supernovae in cold clouds are stopped in those cold clouds, making inhomogeneous patches in an OB association wherein different samples can form. These appeals to the heterogeneity of special events are interesting and may be more relevant than the approach of this paper. If so, the sputtering concepts of this paper can be applied in part to those scenarios. We wish only to be clear in our motive for seeking a more universal and natural evolutionary process consistent with modern ideas on the evolution of the ISM.

The critically skeptical reader can find much to criticize in this paper, because so much of it is based on *ex cathedra* suppositions rather than firm knowledge. We agree with

this criticism, but we conclude that the positive achievements of this model and its potential for vast unifications are sufficient for us to set it forth as worthy of the many studies needed to evaluate it. After all, if we are correct, the dynamic history of the three phases of the ISM can be read in tiny stones in our terrestrial laboratories. This would create a new field of astronomy. It is a cosmic connection not to be overlooked.

In contrast to this hopeful claim, we should mention the competing interpretation popular in the meteoritic community. Clayton, Mayeda, and Epstein (1978b) have advocated the conventional chemical interpretation based on a hot gaseous solar system, and the reader should consult their paper for the clear difference of their viewpoint from ours. They argue that chemical equilibrium between SiO molecules and silicates near 1400 K coupled with equilibrium isotopic fractionation between  $^{28}\text{SiO}$  and  $^{30}\text{SiO}$  molecules has produced the Si isotopic fractionation which they find in "normal" Allende inclusions; i.e., those not having the larger isotopic fractionation of the FUN inclusions. The temperature is chosen in this picture to be near the condensation temperature of silicates from the postulated hot solar gas (e.g., Grossman 1972), whereas we assume that there was no hot solar gas where Allende accumulated. They acknowledge the major weakness of their model, namely, Ca-Al-rich minerals condense fully at higher temperatures, when more than 90% of Si remains gaseous, and therefore can itself bear virtually no isotopic fractionation. They describe this as a *dilemma*. Another weakness is to be found in the inability of this setting to account for the much larger fractionations in the FUN inclusions, which we have discussed in this paper. Clayton, Mayeda, and Epstein (1978b) are therefore forced to acknowledge that some unknown chemical circumstance has produced the large fractionations in the FUN inclusions, but nonetheless suppose that a hot gaseous equilibrium has produced the much smaller fractionations, even though the inclusions are virtually identical petrologically. We suspect instead that the same process is responsible for Si fractionation in all Allende inclusions, but the FUN inclusions are more extreme in recording the effect. Clayton, Mayeda, and Epstein (1978b) conclude with the belief that EK1-4 was well situated so as to be able to absorb isotopic anomalies by accreting anomalous debris from an adjacent supernova, in sharp contrast to our picture of a ubiquitous cosmic chemical memory.

We have shown here and in Clayton (1980a) that severe isotopic fractionation will have been inherited from the interstellar medium unless it was erased by the postulated phase of hot vaporization. Clayton (1980a) described the particularly effective distribution of fractionated isotopic patterns in interstellar grains of differing size. By fractionating gas from dust and small particles from large, routine dynamical effects in cloud collapse will be expected to produce cold macroscopic accumulates having different Si isotopic compositions. An extreme version of this routine phenomenon has probably produced the parents of the FUN inclusions. They, like the non-FUN CAIs, were fused into new mineral forms by a sudden heating

event (Paper I), probably caused by a sudden release of internal chemical energy (Clayton 1980b). Our conjecture can be dramatized by the following *Gedanken* observation: Consider a low-mass star forming in a cold cloud without benefit of a neighboring supernova and surrounded by a preplanetary accretion disk with temperatures less than 1000 K. We argue that Ca-Al-rich aggregates exist there too, and that they also bear isotopic fractionation and isotopic anomalies.

Useful discussions with Don Cox, Eli Dwek, and Jim Ray helped with the early development of these ideas (Clayton 1979c). F. Begemann, T. Kirsten, and H. Wänke

helped me with criticisms of the later ideas of fractionation according to grain-size separates and of SUNOCON fusion by prompt exothermic chemistry. I thank also A. El Goresy for discussing the relevance of *Fremdlinge* and of the nonuniform distribution of Mg in Ca-Al-rich minerals. Four epochs of anonymous referees were sometimes helpful, but sometimes not, in the lengthy struggle of this paper toward publishability in their eyes. This research was supported by NASA grant NSG-7361, by the Max-Planck Institut für Kernphysik, Heidelberg, by the Fulbright Commission during the 1979–1980 Fellowship in Heidelberg, and by Rice University.

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