Section VI

Interstellar Dust Models

THE CORE-MANTLE MODEL OF INTERSTELLAR GRAINS AND THE COSMIC DUST CONNECTION

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1. INTRODUCTION

Historically there have been two different types of grain modelling: One of these basically uses particle populations which evolve essentially by coagulation (e.g., the MRN model: Mathis, Rumpl and Nordsieck, 1977); the other considers the physical and chemical evolution of the particles with a particular emphasis on changes not only in sizes but also in chemical and morphological structure (e.g. Greenberg, 1978; Williams, 1989). The model of Oort and van de Hulst (1946) was the first to consider that grains must evolve in interstellar space by treating both growth and destruction in clouds. The chemical properties had already been derived by van de Hulst (1946) and then later described as the dirty ice model which consisted of the saturated molecules H_2O, CH_4 and NH_3 with trace constituents of other atoms and molecules resulting from surface reactions of atoms on the grains. How such grains could nucleate was left as an unsolved problem but the fact that, once formed, there did not seem to be any reason why they should not grow until they exhausted the condensable atoms in the gas led to the suggestion that a limiting destructive mechanism must be provided. This was assumed to be by grain-grain collisions within clouds moving at relative speeds of 10 $\mathrm{km\,s^{-1}}$. We thus had the first dynamical theory leading to a steady state distribution of grain sizes. This model provided for me the starting point of the core-mantle model of grains. The observations of the 60's and henceforth clearly showed the existence of other types of small particles, which have been invoked to explain the 2200 Å hump (Stecher and Donn, 1965), the far ultraviolet (FUV) extinction (Greenberg and Chlewicki, 1983), and now certain infrared emission features. These other components notwithstanding, grains still account for the major fraction of the solid particle mass in space.

In this summary I shall attempt to provide some key observational and theoretical arguments leading to the modern core-mantle model. The major evolutionary aspects are: (1) the concept of cyclic processing within alternately diffuse and molecular clouds, and (2) the photochemical evolution of accreted frozen mantles on the grains. The problem of nucleation was essentially solved with the observation of the 10 μm excess emission for cool evolved stars which provided evidence for the ejecta of a substantial number of small silicate particles into space (Humphreys *et al.*, 1972).

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Although observations of interstellar dust abound, there are severe constraints on the regions in which all of these characteristics are seen; e.g. where infrared observations are best, the visual and ultraviolet are difficult if not impossible. The FUV extinction has never been observed in the core of a molecular cloud. Where might we observe all interstellar dust properties? If the theory of comets as aggregates of interstellar dust is correct, the answer is "In the nuclei of comets." Several observations suggesting that this is a reasonable answer are given in the last section where I show that, already, based on comet dust and IDP properties, we may derive some of the core-mantle interstellar dust properties or vice versa. The cosmic dust connection is alive and kicking. I should point out here that, in a different context (meteorites), Cameron wrote a paper "Interstellar Dust in Museums" (Cameron, 1973). If we extend this idea to some of the low density IDP's, this is not far from reality (Greenberg, 1989; Brownlee, 1978).

2. CYCLIC EVOLUTION OF DUST

Recent studies of the observations of so-called diffuse cloud dust (dust not in molecular clouds) in the ultraviolet have revealed the fact that there are three populations of dust (Greenberg and Chlewicki, 1983; and Chlewicki, 1985). There are elongated "large" grains of $\sim 0.12\ \mu m$ in mean radius which provide the major blocking of starlight in the visual. The polarization of starlight in the visual which is associated with extinction implies that these particles are elongated by at least a factor three (Greenberg, 1968). There are also very small carbonaceous particles of $< 0.01\ \mu m$ in radius which produce a strong absorption feature at about 220 nm. In addition there is an independent population of small particles required for the far ultraviolet. Perhaps these are small ($<0.01\ \mu m$) silicates or perhaps PAH's.

How the large grains form and evolve is a complex physical and chemical story. Basically, they start with silicate particles forming in the atmospheres of cool stars which, after being ejected into space and cooling down to 10–15 K, provide nucleation cores for the growth of mantles of ices. These mantles result from accretion of gas phase atoms and molecules of the abundant atomic species, oxygen, carbon, nitrogen and sulfur along with hydrogen (Greenberg, 1982) which may also undergo subsequent surface reactions. Since the silicate cores provide an extra polarization at $10\ \mu m$ – associated with their $Si - O$ stretch infrared absorption – they must be non-spherical and aligned. The non-sphericity is apparently not a result of crystallinity because the shape of the $10\ \mu m$ absorption is characteristic of amorphous silicates. It is probably due to small (more or less spherical) particles stuck together: the elongation of two connected equal-sized particles is obviously 2:1. Although the accretion of the gases is random, it does not, as expected, make the particles more spherical. Rather, because the particles must be spinning suprathermally in order to produce the required alignment (Greenberg and Chlewicki, 1987), the extra centrifugal force causes the atoms and molecules which hit and stick, to tend to slide outward from the center of rotation; i.e., the surface diffusion is slightly enhanced by the centrifugal energy difference. This differential sticking leads to enhanced non-sphericity. The observation of an extra degree of linear polarization at $3.07\ \mu m$ shows that the solid H_2O in interstellar mantles is also elongated, thus confirming this effect. In addition to H_2O, many other simple molecules – and even ionic-species – like CO, H_2S, CH_3OH, OCS, OCN^-, NH_4^+, H_2CO etc. have been observed in interstellar dust. These ices are always being photoprocessed by ultraviolet from either distant stars or by UV created by cosmic rays or arising from local hot stars and/or stellar winds. The results of such photoprocessing is not only a change in the basic composition of the ices, but also the production of complex organic refractory residues which have been studied in the Leiden laboratory and compared with astronomical observations (Greenberg, 1982*b*; Agarwal *et al.*, 1985; Schutte, 1988).

Laboratory-produced residues have been successful in recreating the essential shape of the 3.4 μm feature which consists of contributions from the $C-H$ stretch in CH_2 and CH_3 groups in complex organic molecules (Greenberg, 1984; Schutte and Greenberg, 1986). Although the precise shape of the interstellar 3.4 μm feature (or features) has not yet been matched there are several good reasons for this: first, no laboratory residue has yet been subjected to the complete radiation history of an interstellar grain because of present laboratory limitations; second, line of sight effects by grains with varying histories create averages of spectra of various complex organic residues (Greenberg, 1984).

The spectral correspondence between the laboratory residue and the galactic center is supported quantitatively by the measured strength of the laboratory 3.4 μm feature. The major component of the dust in low density (diffuse) clouds is the organic refractory material.

The evolutionary picture of dust starts with a small silicate core captured within a molecular cloud gradually building up an inner mantle of organic refractory material which has been produced by photoprocessing of the volatile ices. Within the dense clouds critical densities lead to star formation and subsequent ejection of some of the cloud material back into the surrounding space. Much of this material, finding itself in a very tenuous low density environment, expands to the diffuse cloud phase. During the intermediate stage of passage from high to low density, the volatile grain mantles are heavily photoprocessed producing the major part of the organic refractory material. It is important to note that without their organic refractory mantles, the silicate cores could not survive. The rate of destruction of pure silicate grains leads to a maximum lifetime of $\tau_{Sil} \sim 4 \times 10^8$ yr, which converts to a galactic average mass loss rate of $d\rho_{Sil}/dt = 5 \times 10^{-43}$ g-cm^{-3}-s^{-1} which is 100 times lower (!) than the destruction rate. On the other hand, the mean production rate of the organic refractory of $d\rho_{O.R.}/dt \sim 10^{-41}$ g-cm^{-3}-s^{-1} is adequate to replenish the mantle material lost in the diffuse cloud phase even if the organic refractory component is somewhat less tough than the silicates (Greenberg, 1982). Therefore, silicate core-organic refractory mantle grains survive the diffuse cloud phase to reenter the molecular cloud phase, having lost a fraction of the protective organic refractory "shield" which permits the silicate cores to survive.

The mean star production rate of 12 $M_\odot$-yr^{-1} implies an interstellar medium turnover time of $\sim 5 \times 10^9$ yr so that this is the absolute maximum lifetime of a dust particle no matter how resistant to destruction. If we use a mean molecular cloud-diffuse cloud period of 2×10^8 yr (10^8 years in each), then a typical grain anywhere in space will have undergone at least 20 cycles so that, for example, the typical diffuse cloud dust particle age is $> 10^9$ yr and consists of a mix of particles which have undergone a wide variety of photoprocessing. Note that the organic refractory mantles are subjected to the highest photoprocessing rates in the diffuse cloud phase – higher by factors of 10,000 or so than in the molecular cloud. This would imply that the organic refractory mantle on a grain is not a homogeneous substance, but rather, layered like the rings in a tree trunk. Sequential organic mantle formation (in the molecular cloud phase) and intense photoprocessing (in the diffuse cloud phase) would lead to a structure in which the innermost layers have been the most irradiated and the outmost layer in the most recent molecular cloud phase is first generation organic refractory surrounded by lightly photoprocessed ices. Because of this kind of layering, and the fact that the grains are of various ages, leads one to expect average homogeneity of diffuse cloud grains both in size and structure which is observed as a uniformity in the visual extinction curve and a rather structureless 3.4 μm feature. In other words, diffuse cloud grains represent a steady-state average of grains of a multiplicity of chemical and physical histories. Since further photoprocessing of organics leads to a greater and greater depletion of O, N and H, the innermost layers are the most "carbonized" and the most nonvolatile. This property will be reflected in the emission characteristics of heated comet dust relative to the absorption of interstellar dust due to the loss of the more volatile organic components after the dust leaves the comet surface.

We arrive at a picture of the "large" grains as being multiple layered particles with a mean silicate core radius of 0.05 μm (elongated by a factor of about 3). In diffuse clouds there is a mean mantle thickness of $\sim$ 0.05 μm (total radius about 0.1 μm) and an elongation of 3 or 4 to 1. In molecular clouds there is an additional mantle (outer mantle) of more volatile species dominated by H_2O but generally containing CO, NH_3, H_2CO etc. (see Table 1). This outer mantle may grow to a thickness of 0.01 in only 10^5 years in a cloud with hydrogen density $n_H = 10^3$ cm^{-3} and, in $< 10^6$ years, it would deplete all of the available gas (excluding H and He) in a cloud of density $n_H = 10^4$ cm^{-3} (Greenberg, 1985). There are normally desorption mechanisms provided by ultraviolet processes which lead to grain explosions (d'Hendecourt *et al.*, 1985; Greenberg, 1976) thus inhibiting such complete grain growth. In very dense clouds such processes are turned off by lack of ultraviolet radiation and the grains then may deplete all of the remaining gas. Such a situation would prevail in the latest stage of the molecular cloud corresponding to the earliest stages of the protosolar system and comet formation.

Graphite was the first candidate suggested for the 220 nm hump (Stecher and Donn, 1965). One of the basic difficulties with this material has been the fact that the shape and position of the hump depends strongly on the size and shape of the particles, whereas the observed hump is highly uniform in structure (Savage and Mathis, 1979). Another problem has been the fact that, although the hump and the visual extinction are remarkably well correlated, the sources of these two dust components are independent in the sense that the visual extinction particles are basically created in the interstellar medium while the graphite is (presumed to be) formed in stellar atmosphere (Czyzak and Santiago, 1973).

There is then a problem of replenishing the graphite at a rate sufficient to counter its erosion and destruction rate (Mathis *et al.*, 1977; Greenberg, 1986).

A purely interstellar process leading to the presence of a carbon component follows from ultraviolet photoprocessing of grain mantle ices (van der Zwet *et al.*, 1987; Greenberg *et al.*, 1987). The assumption here is that some of the organic refractory mantles are either broken off by some grain destruction process, such as shocks, or ejected by grain explosions, and appear in space initially as very small carbonaceous particles which are small enough to produce the hump (Greenberg, 1979; van de Hulst, 1957).

TABLE 1: MOLECULES DIRECTLY OBSERVED IN INTERSTELLAR GRAINS AND/OR STRONGLY INFERRED FROM LABORATORY SPECTRA AND THEORIES OF GRAIN MANTLE EVOLUTION.

Molecule	Comment*	
H_2O	O	M2
CO	O	M2
H_2S	O	M2
NH_3	O	M2
H_2CO	O	M2
$(H_2CO)_n$	I	M2
OCN^-	O	M2(M1)
NH_4^+	O	M2
CH_3OH	O	M2
OCS	O	M2
CO_2	I	M2
CH_4	I	M2
S_2	I	M2
complex organic	O	M1
"silicate"	O	C,B
"carbonaceous"	(O,I)	B

*: O = observed, M1 = inner mantle,
M2 =outer mantle,B = small bare,
I = inferred, C = core

Other forms of small carbon particles (or perhaps large molecules) similar to polycyclic aromatic hydrocarbons (PAHs) are suggested by their contribution to certain interstellar infrared emission features (see the section on the infrared emission features in this book, and references therein). These particles appear to use up on the order of 5% of the cosmically available carbon, and may finally wind up either as a part of the organic refractory component or imbedded in the volatile mantles. It is not clear what happens to them in the molecular cloud phase. Grain modelling of the far ultraviolet part of the extinction curve has been successfully performed using very small silicate particles but PAH's have some advantages.

The optical properties of interstellar dust are a very important ingredient, not only in theories of extinction and polarization in space, but also in theories of how they act as aggregates. Observationally, it is seen that the diffuse cloud grains absorb about 60% of the incident (visible) radiation (albedo $= \alpha = 0.6$) and that they scatter about 80% of the radiation in the forward direction (asymmetry factor $g =< \cos\theta_{sc} >= 0.8$) (Savage and Mathis, 1979). The laboratory-created organic refractory mantles are strongly absorbing in the ultraviolet and experiments, supplemented by theory, lead one to expect that strongly photoprocessed organics have a complex index of refraction in the visual of $m_o/m_{si} \sim 1{:}5$ (Chlewicki and Greenberg, 1989) which implies a dark material when small particles are aggregated. The first generation laboratory organic residues start out yellow (UV absorption) and become darker (brown) with radiation. Theoretical calculations of core-mantle particles with such mantle optical properties have been shown to match the observed extinction and polarization as well as the albedo of interstellar dust (Greenberg and Chlewicki, 1987).

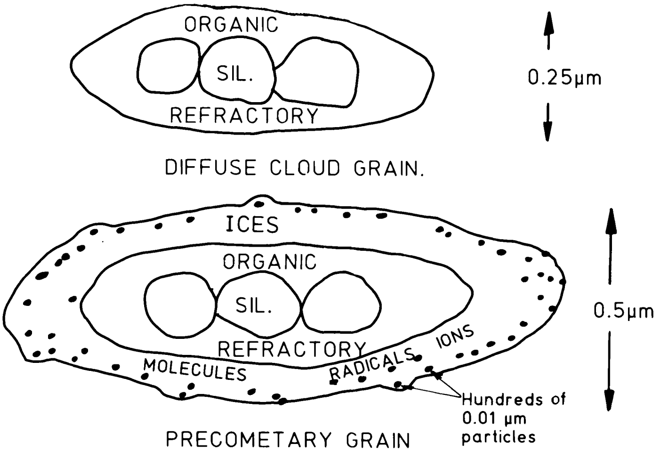

Fig. 1. Interstellar grains as core-mantle structures.

A schematic representation of grains in the various regions of space is shown in Fig. 1. In the final stage of cloud condensation we may expect that all remaining (condensable) molecules will have accreted onto the dust. In addition, the very small ($< 0.01\ \mu m$) particles will have been collected and trapped within the outer volatile icy mantle. An alternate route for the very small particles is that they may themselves accrete icy mantles in the dense cloud after the ultraviolet has been "turned off" so that some may accrete as small protuberances on the outer parts of the large grains enclosed in their own small mantles.

3. DUST AGGREGATION AND MORPHOLOGY

Our focus here will be on establishing a relationship between the chemical and morphological structure of presolar interstellar dust and comet dust, interplanetary dust, and meteorites.

In our solar system all of the planets and satellites have incorporated into their bodies at least the most refractory components of the interstellar dust which existed in the presolar nebula. Comets appear likely to have preserved their original composition best, including their volatiles, not only because the volatile molecule S_2 may be traced back to the photochemical evolution of the interstellar dust (Grim and Greenberg, 1987), but also because of the observed CH_4/H_2O ratio (Larson *et al.*, 1988). As a first approximation, therefore, we consider a comet nucleus as if its chemical composition and morphological structure were directly related to interstellar dust. Table 2 shows the relative fractions of the various chemical constituents

TABLE 2

MASS FRACTIONS OF THE COMET NUCLEUS COMPONENTS BASED ON PRESOLAR INTERSTELLAR DUST

COMPONENT		MASS FRACTION
Silicates + Metal Oxides	DUST	0.14 + 0.06[a]
Carbonaceous		0.06[a]
Organic Refractory		0.19
H_2O	GAS	0.37[b]
$CO + CO_2$		0.05[c]
Other Metals and Residuals (NH_3, CH_4, HCO, OCN^-, S_2, CH_3OH, ...)		0.13

[a] Very small particles $a \lesssim 0.01\ \mu m$

[b] Based on 70% of H_2O in volatile grain mantles in molecular clouds

[c] Based on $CO/H_2O \lesssim 15\%$ in molecular clouds plus effect of photoprocessing to produce CO_2

which have been obtained by an extrapolation from the molecular cloud dust phase (Greenberg, 1982*b*, 1983).

In forming the nucleus we assume that, first, clumps of grains form, and then clumps of clumps and so on, until finally we reach the size of the comet nucleus. If we should start with the interstellar dust tightly packed and then remove all the volatiles (along with the trapped super small particles), the resulting mean density of the remaining core-organic refractory grains skeleton is about 0.5 g-cm^{-3} (Greenberg, 1986). It is, however, observed that meteors (which are what is left after the original cometary volatiles have evaporated) have a characteristic density much lower than this, often being even less than 0.1 g-cm^{-3}. This leads to a packing factor of 0.2; i.e., a comet of about 80% empty space! A model of such an open aggregate of 100 typical precometary grains is shown in Fig. 2a.

4. COMET DUST

4.1. SIZE AND FLUFFINESS

The "unexpected" submicron sized particles detected by the Vega and Giotto impact systems extended down to the limit of 10^{-17} g. The interstellar core-mantle grains (without volatiles) have an average mass of $\sim 0.65 \times 10^{-13}$ g and the bare (0.01 μm) particles have a mass of $\sim 10^{-17}$ g.

The spatial and temporal distribution of the masses and flux of dust particles measured by the dust counter and mass analyzer (DUCMA) on Vega 1/2 showed, among other things, that the lowest masses were the first particles encountered at the fringes of the coma. One of the explanations for this phenomenon by Simpson *et al.* (1986) is that some of the dust particles are "comprised of much smaller particles"

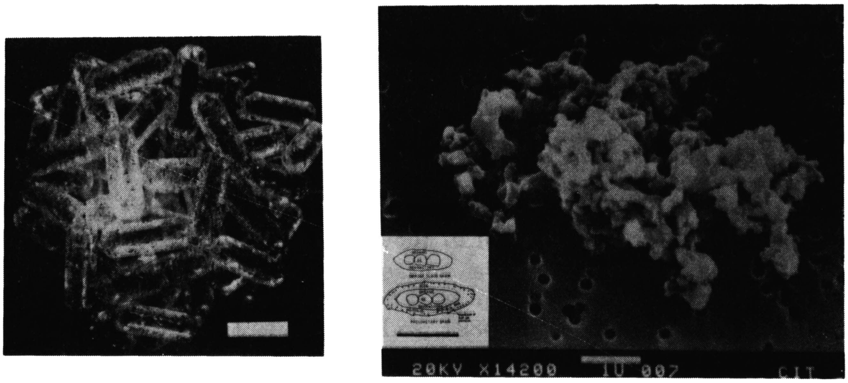

Fig. 2. **a**: A piece of a fluffy comet: Model of an aggregate of 100 average interstellar dust particles. Each particle which consists of a silicate core, an organic refractory inner mantle, and an outer mantle of predominantly water ice in which are embedded the numerous very small (< 0.01 μm) particles responsible for the ultraviolet 216 nm absorption and the far ultraviolet extinction (See Fig. 1). Each particle as represented corresponds to an interstellar grain 1/2 μm thick and 1.5 μm long. The mean mantle thickness corresponds, in reality, to a size distribution of thicknesses starting from zero. The packing factor of the particles is about 0.2 (80% empty space) and leads to a mean mass density of 0.28 g-cm^{-3}, and an aggregate diameter of 5 μm. **b**: A highly porous chondritic IDP. Note that the bird's nest particle (Fig. 2a), the IDP (Fig. 2b), and average interstellar core-mantle particle (Fig. 2b insert), are equally scaled to 1 μm.

from which pieces are shed which appear at great distances as the material which binds them sublimes; i.e., the initial dust is crumbly and resembles the bird's nest model (Fig. 2a without volatiles).

4.2. CHEMICAL COMPOSITION

The dust impact mass analyzers on Vega 1/2 (PUMA) and on Giotto (PIA) showed a predominance of the light elements H, C, O, N (organics) relative to the heavier elements, Si, Mg, Fe (minerals) in the dust (Kissel *et al.*, 1986*a*, 1986*b*).

Kissel and Krueger (K+K) (1987) have derived a molecular analysis of the comet dust and, in particular, its organic component. Masses between 2 x 10^{-15} and 10^{-11} g were measured, with the masses of most of the particles estimated to be in the range 10^{-12}–10^{-13} g with a "systematic error within an order of magnitude." The typical total relative atomic abundances in their molecules (of the organic refractory) show a significant lack of oxygen just as is predicted by the interstellar dust model. A four-fold enhancement of carbon was predicted relative to oxygen

(Greenberg, 1982*b*, 1983). The ratio of organics to silicate mass deduced by K+K is $m_{OR}/m_{Sil} = 0.5$ which, not surprisingly, is less than that in the interstellar dust because of the expected evaporation of the less refractory organics at solar system temperatures.

4.3. SIZE PLUS CHEMICAL COMPOSITION: THE 3.4 AND 10 μm EXCESS EMISSION

The 3.4 μm and 10 μm excess emission in comet dust provides evidence, not only for the basic chemical ingredients as given in the mass spectra, but also for the morphological structure (Greenberg, Zhao and Hage, 1989). It turns out that pure silicates, no matter how small, do not achieve high enough temperatures to produce the observed 10 μm emission. At, for example 1.11 AU, the required temperature needed to keep the total mass of the emitting particles at all reasonable is $T > 430$ K. Absorbing organic refractory mantles such as those on interstellar silicate cores are absolutely required to raise the compound grain temperatures high enough to make the 10 μm peak observable. Furthermore, the $T > 430$ K temperature constraint leads to a highly probable silicate core radius ~ 0.05 μm and a mantle thickness > 0.02 μm i.e., an organic to silicate mass ratio $m_{OR}/m_{Sil} = 0.9$ which, within the uncertainties, is like that deduced from comet dust mass spectra. If only such small particles ($m < 10^{-13}$ g) could have produced the 10 μm (and 3.4 μm) emissions, their fluxes would have been more than 10,000 times higher than observed. It is only by considering them to be in fluffy aggregates that the integrated fluxes come into reasonable resemblance to the particle impact detector data (McDonnell *et al.*, 1987) – although still by a factor of about 25 higher for masses $< 10^{-9}$ g.

5. INTERPLANETARY DUST

5.1. ZODIACAL LIGHT

Interplanetary dust has classically been observed via its scattering of sunlight – the zodiacal light. Those which are within 1 AU scatter visible light much more effectively than those which are beyond 1 AU. At the same time, those dust particles which are farther out, are more effective emitters of infrared radiation. This implies a difference in kind as well as number with increasing solar distance (Hong and Kwon, 1988). The most obvious explanation of this phenomenon is that the radial decrease of the albedo of the zodiacal light particles is produced by a decrease in material density, just as the albedo of cometary dust is decreased because of its fluffiness.

With the assumption that most interplanetary particles start out as fluffy low albedo comet dust particles (like that in Fig. 2a), Mukai and Fechtig (1983) proposed a mechanism by which solar heating would lead to a gradual compaction of the initially fluffy dust by evaporation of the volatiles in what they called "Greenberg particles," leading to more compact and higher visual scattering particles like the "Brownlee particles" (Fig. 2b).

5.2. IDP'S

Although the mean density of the chondritic porous IDP's is low, it is much higher than that in the initial cometary dust. But, as has been pointed out by Brownlee (Brownlee, 1988), there is no evidence of a bird's nest structure in the IDP's (Fig. 2b). What we see in Fig. 2b is an aggregate of more or less spherical particles of about 0.1 μm diameter whose infrared signature is that of silicates. When the interstellar dust is scaled like the IDP, we can see how its silicate core segments – which are hidden in the bird's nest model (Fig. 2a) – are like the silicates in the IDP. But where are the organic refractory mantles in the IDP's? In the original (interstellar dust) comet nucleus material the ratio of O.R. mass to silicate mass is given as about 1:5 (Table 2). However, already in the comet dust, the loss of the more volatile organic refractory molecules leads to about 1:2 (K+K). While the organic mantles are not "seen" in the IDP electron micrographs, they become immediately apparent with Raman spectroscopy (Wopenka, 1988). It appears that every silicate particle is covered by some organic mantle. The fact that the mean silicate particle size is like that of the interstellar core pieces and each silicate or clump of silicates has an organic refractory coating is certainly suggestive of the interstellar origin while the bird's nest morphological structure is lost because of the removal, during passage from the comet to the earth, of a further part of the original comet dust organic refractory component.

Additional indications for the cometary to interplanetary dust evolution may be seen in the lower density of meteors whose aphelion distance are beyond 5.4 AU as compared with those which spend more time closer to the sun (Verniani, 1973).

6. CONCLUDING REMARKS

We have to look to future space missions to recover comet material that is much more pristine than that which we can infer from flyby or even rendezvous missions. If the comet nucleus material can be retrieved from its depths and maintained intact cryogenically for laboratory studies, we may hope to study, not only its atomic and molecular composition, but also its morphology. Microprobes developed for IDP studies (Bradley and Brownlee, 1986) will make investigations possible of submicron structures in comet material. If it should turn out that the interstellar dust model is correct, individual grains, whose mean lifetime before becoming part of a comet is about 5 x 10^9 yr, will reveal cosmochemical evolution, not only of the solar system, but dating back a further 5 billion years before the earth's beginning back to the earliest stages of the chemical evolution of the Milky Way. Dramatic differences in isotopic abundances should be exciting ones indeed for studies of our origin.

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