Formation of Dust Particles in Cool Stellar Outflows

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Abstract.
The formation of grains in circumstellar outflows is reviewed. Special emphasis is laid on the distinction between the condensation of seed particles and the further growth of these particles serving as nucleation centers for solid material built up by abundant elements. Depending on whether the gas phase is oxygen- or carbon-rich fundamentally different dust grains will form. As it turns out, the predictions of dust formation theory seem to be in good agreement with experimental findings from laboratory investigations of presolar grains extracted from meteorites. We also briefly survey the state of modeling dust producing red giants on the asymptotic giant branch (AGB).

INTRODUCTION

From a theoreticians point of view, the complex of cosmic dust comprises three fundamental problems: Formation — Processing — Evolution. In conventional astronomical investigations details of the composition and size distribution of interstellar grains can only be inferred through physical models or spectra from laboratory analogs. With the advent of advanced preparation techniques and microanalytical tools such as transmission electron microscopy it has become possible to isolate and investigate individual grains extracted from meteoritic material (see the contribution of Zinner to this proceedings). Measurement of their isotopic ratios clearly establishes the presolar, i.e. *stellar*, origin of these grains [1]. Thus, microanalytical studies of meteoritic material provide important clues regarding the nature of interstellar dust.

Nevertheless, one has to be aware of the fact that the grains found in the interstellar medium (ISM) and — even more so — within the solar system have undergone some degree of processing which may have modified their original nature (see below). Thus, the cosmic dust connection is unclear and an "*ab initio* theory" of dust formation is needed. This is all the more true, as dust formation in stellar objects plays a crucial role in the cosmic cycle of matter: In setting the initial conditions it has a decisive influence on the further evolution of the dust component.

So, as a first step, the formation of primary condensates has to be clarified: What are the physical and chemical constraints for solid matter to form in space? By means of which mechanisms can dust formation be described microscopically? As we will see, circumstellar envelopes can be regarded as an ideal laboratory for the formation of solid material in space. This formation of grains will be the topic of our contribution. We will only review dust formation in cool circumstellar outflows and refer to the contribution of Wooden in this proceedings for details on dust grains in supernova ejecta.

Once formed, existing grains may be processed in a variety of ways, both destructive and non-destructive. Interstellar shock waves, for one example, lead to physical and chemical sputtering, even complete vaporization (see the contribution of Jones to this proceedings). Furthermore, intense UV-fields and cosmic rays may change the chemical structure of the solid material [2–4]. On the other hand, grains in quiescent molecular clouds are coated by ice-mantles and, given sufficient time, tend to coagulate forming larger aggregates. Due to the volatile nature of such ice-mantles and the aggregates being prone to shattering even by moderately strong shock waves these processes can be classified as processing rather than formation.

Evolution of the dust component comes to a temporary halt after the grains have been incorporated in meteoritic and cometic material or within other solar system bodies provided that the body is not large enough to experience significant radioactive heating as is known to have occured on the parent bodies of several classes of meteorites.

DUST FORMATION

Judging from the abundance of solid matter in the ISM (1% by mass), the formation of dust particles should be a very common phenonemon in astrophysical situations as long as the temperature is low enough for matter to assume a solid state. It turns out, however, that moderate temperatures are only a necessary condition for the occurrence of solids. The actual *formation* of grains usually involves reaction energy barriers that require a sufficiently large temperature to be overcome at all. Furthermore, the reaction sequence that builds up solid matter from atomic or molecular constituents has to take place in a finite time interval, thus necessitating densities higher than those of the ISM.

From the above discussion it should be obvious that efficient dust formation, in contrast to the occurrence of dust itself, is not that common a phenomenon after all, but is only possible in a limited area of the p-T-plane.

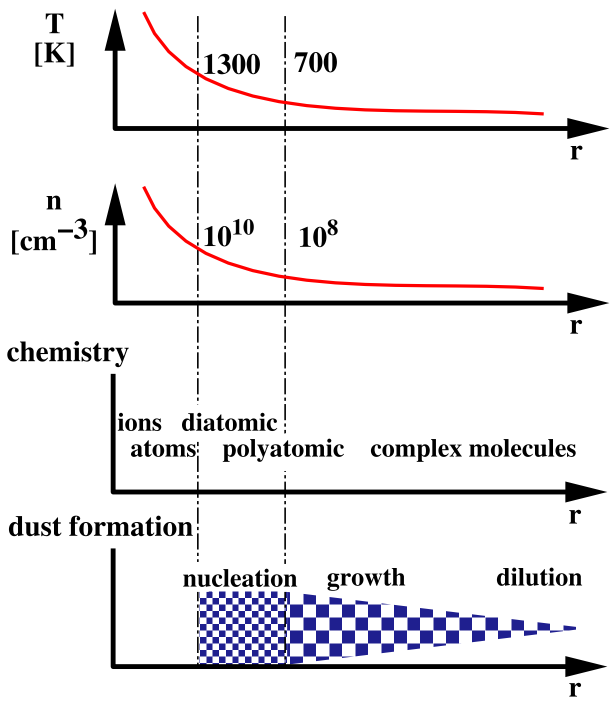

FIGURE 1. Schematical radial course of temperature and density of an expanding stellar outflow showing the evolution of a complex chemistry and dust formation (adopted from Sedlmayr 1994 [13]).

Physical conditions in cool circumstellar envelopes

Circumstellar envelopes around cool late-type stars are unique with regard to the prevailing physical conditions. There are few other places in the universe where all of the conditions for efficient dust formation previously mentioned are fulfilled. Figure 1 schematically shows the radial course of temperature and density in an expanding stellar outflow as well as the resulting stepwise gas–solid transition of matter from molecules to grains.

Generally, the concept of a single temperature for all components of a tenuous plasma is not a good assumption. This is due to the fact that, although radiation is the main energy pool and can be regarded as acting as a heat bath, molecules, small clusters, and grains of different composition all assume a different radiative equilibrium temperature because of their different spectral absorption and radiative emission efficiency. Furthermore, one has to distinguish between the kinetic temperature and the internal vibrational or lattice temperature, respectively, since the former determines the collision frequency while the latter is relevant to the chemical reactivity [5]. A realistic description of dust formation has to account for these facts.

Last not least, stellar envelopes are places of pronounced non-equilibrium effects with regard to both chemistry (see next subsection) and thermodynamics. The latter means that shock heating or adiabatic cooling due to expansion may lead to a temporary departure of the kinetic gas temperature from the radiative equilibrium temperature depending on the efficiency of radiative heating and cooling [6]. Given the extreme temperature dependence of the dust formation efficiency such thermal non-equilibrium effects may easily rule out the formation of dust completely or make it possible in the first place [7].

The circumstellar radiation field is rather complex with regard to both its frequency and angular dependence. Molecule formation in the cool outflow is responsible for a wealth of absorption lines in the visual, infrared and radio regions of the spectrum. Furthermore, these lines become continously doppler-shifted in the accelerating outflow. Concerning the angular dependence there are contributions from the stellar photosphere and from the illuminated dust shell surrounding the star.

Gas phase chemistry

In order to approach the problem of dust formation in stellar atmospheres it is essential to investigate the chemistry of the gas phase. This is due to the fact that molecules are both precursors for the seed particles as well as the main contributors to further growth of existing grains.

The decisive quantities for the chemistry, apart from the state variables temperature and density, are the element abundances ϵ_i. They are determined by the elemental composition of the molecular cloud the star originally formed from, the history of the nuclear processes within the star and the efficiency of possible dredge-ups, that transport nuclear processed material to the atmosphere (see the contribution of Lattanzio to this proceedings).

Understanding stellar nucleosynthesis is also the key to the determination of isotopic ratios of elements, which can be regarded as a signature or fingerprint of an astrophysical object. Since the chemical reaction efficiencies, and hence dust formation, are insensitive to the different masses of isotopes, these ratios are directly reflected in the isotopic composition of the grains formed. A corresponding laboratory analysis of meteorites and interplanetary dust particles may thus help to identify the sources of origin of presolar materials and, keeping in mind possible chemical processing of the material in the ISM and the solar nebula, provide important clues concerning the details of dust formation (see the contribution of Zinner to this proceedings).

Investigating the chemistry in circumstellar outflows is difficult, since it unfortunately turns out that there are a multitude of possible species and reactions (cf. Figure 2) which generally will give rise to a complex chemical rate network. The situation sometimes is less complicated in the dense regions close to the star. Here, the chemistry evolves in a quasi-static way and a kinetic equilibrium approach is sufficient [8]. If shocks and chromospheric UV-radiation fields are absent, detailed balancing of the kinetic reactions can

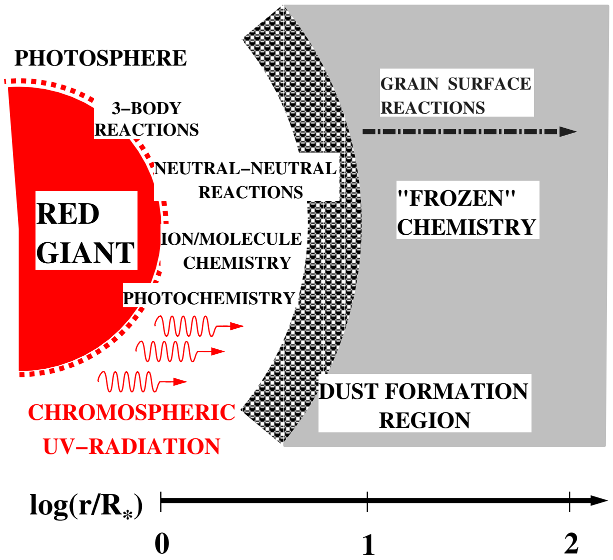

FIGURE 2. Chemical processes in the inner part of the circumstellar shells around red giants (adopted from Patzer el al. 1996 [11]).

be assumed, and thus chemical equilibrium may be established [9].

In those stars where no strong UV-fields are present, a considerable simplification arises from the fact that some molecules with exceptionally high bond energies may have already formed at high temperatures early in the outflow. For element compositions $\epsilon_{\rm O} > \epsilon_{\rm C}$ ($\epsilon_{\rm O} < \epsilon_{\rm C}$) all carbon (oxygen) is consumed or blocked in the CO molecule, effectively rendering one of the two most abundant metals chemically inert under equilibrium conditions.

Nevertheless, any realistic modeling the chemistry of circumstellar outflows still remains a difficult task [10]. Figure 3 shows the results of chemical non-equilibrium calculations for the envelope of a typical C-star [11]. Obviously, there are pronounced deviations from the corresponding chemical equilibrium concentrations. Furthermore, the concentrations of some species "freeze", when the temperature and density in the expanding flow have fallen below some critical level, i.e. when the chemical reaction timescales have become larger than the hydrodynamical ones. Similar calculations have been performed for oxygen-rich M-stars [12].

Seed nucleation

From a microscopic point of view, grain formation from the gas phase can be conceived as a complex network of chemical reactions leading under favourable conditions to more and more complex molecules or clusters which finally grow to macroscopic particles. It is impossible to perform this approach in its full generality by means of either todays analytical or computational methods. It has, however, turned out to be a good compromise to treat dust formation as

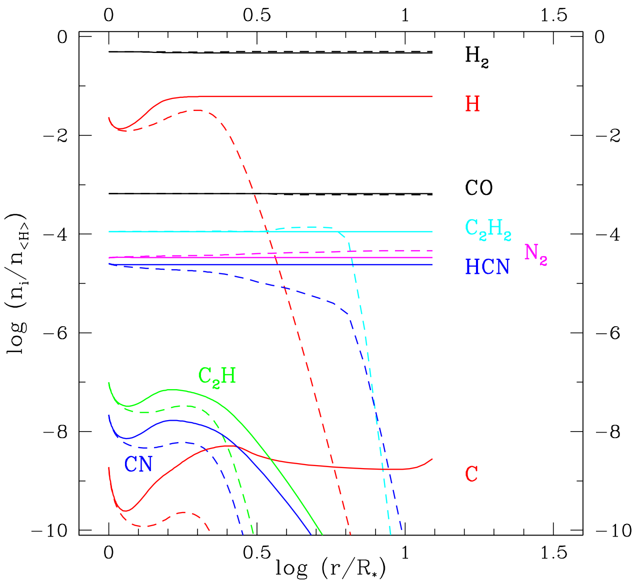

FIGURE 3. Molecule concentrations in the inner part of a C-star shell as resulting from non-equilibrium calculations (dashed lines are the equilibrium concentrations) (adopted from Patzer et al. 1996 [11]).

a two-step process of initial seed nucleation and subsequent growth of these seeds to macroscopic particles [13].

This division is guided by nature: Small clusters behave similar to molecules, they have, for example, discrete energy levels. Beyond a size N_ℓ of the order of $10^2 \ldots 10^3$ atoms, however, clusters tend to develop a continuous band of energy states. A localized energy input, e. g. by photon absorption, exothermic reactions or due to impacting particles, is quickly dissipated within the many degrees of freedom of the complex aggregate. Such properties are characteristic of macroscopic particles. Consequently, clusters of sufficient size are usually characterized by bulk material parameters rather than by their detailed internal structure at an atomic level.

When investigating seed nucleation as a first step towards the formation of grains the first question one has to ask is, which elements are likely constituents of the dust or in other words which solid materials can be expected to form from the gas phase? As has been argued in the last section, some elements may be partially or completely blocked in molecules with high bond energies such as CO. This rules out the effective participation of the these elements in chemical reactions, and therefore also in those reactions important for dust formation. Application of a second criterion, namely the stability of a possible dust material at high-temperatures, further reduces the number of candidates for primary condensates. Last but not least, the element abundance has to be

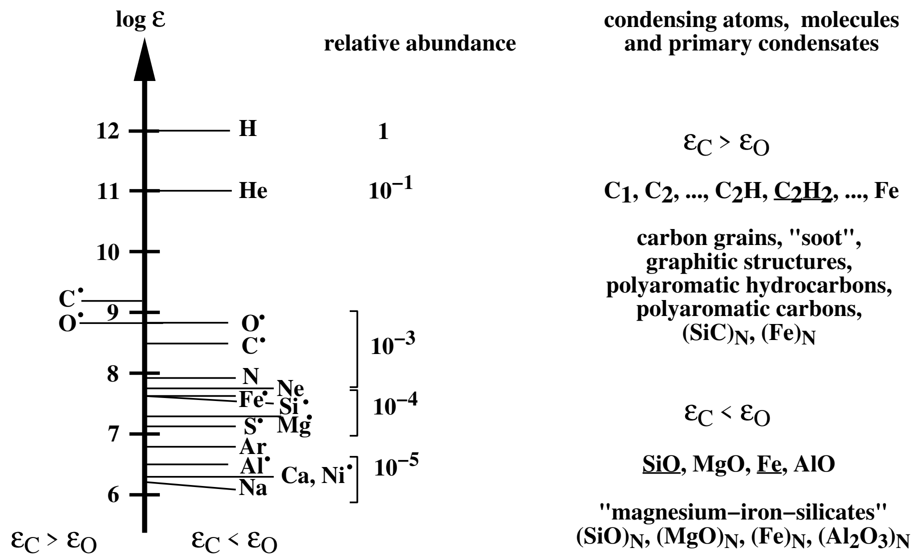

FIGURE 4. Abundance hierarchy of the elements. • means that this element is a constituent of high-temperature condensates. The leading molecules in the grain formation sequence are underlined (adopted from Gail & Sedlmayr 1987 [14]).

considered as well (cf. Figure 4). Aluminium, for example, is a factor of ten less abundant than silicon, magnesium of iron and hence cannot make up the bulk of the dust, although it is conceivable that Aluminium compounds form initial seed particles.

The nucleation of seed particles accounts for only a small fraction of the total dust mass produced. Nevertheless a detailed description and thorough understanding of the processes involved is necessary, since the formation rate of nucleation centers determines the *total number of grains to emerge.* There are several different methods to treat the nucleation process, each having its own specific advantages and drawbacks. Here, we will describe the three methods which are presently in use: classical nucleation theory, chemical rate network and chemical pathway approach.

Chemical rate network

This approach follows rather naturally as an extension of treating the non-equilibrium chemistry of the gas phase. Straightforward as it seems to be, the method raises some serious practical problems. When the network is extended to include mesoscopic clusters of sizes up to N_ℓ the number of "nodes" and reactions that have to be considered increases dramatically. Furthermore, the necessary input data, e.g. reaction rates and cross sections, are not known except for the simpler molecules.

Ultimately, the use of this approach is limited to some specific scenarios where a theoretical analysis aimed at a reduction of the network can be performed. This is done by applying criteria such as the abundances of element constituents, the stability of molecules or clusters in the temperature and pressure range considered or the activation energies of the reactions involved.

Hitherto, the chemical network approach has only been applied to the formation of polycyclic aromatic hydrocarbons (PAHs) in simple models of carbon-rich stellar envelopes [15,16]. However, in order to include aromatics beyond two-ring compounds ($N_\ell \sim 20$), the authors were forced to lump similar compounds into one hypothetical species and to group similar reactions into "reaction classes" which were assigned a single rate coefficient. The most important result of these investigations is that efficient PAH production is limited to a small temperature interval of 800 K to 1000 K. It was also concluded [16,17] that the predicted PAH yield is significantly increased if a proper distinction between gas kinetic temperature and vibrational temperature of the PAHs is made.

Chemical pathway approach

As a consequence of the practical problems resulting from the "pure" chemical network approach, a simplified but more feasible method was proposed [18]. Its basic idea is that knowledge of the most frequently passed reaction channel through the network is sufficient for the determination of the formation rate of seed particles.

In order to select a certain pathway for a given temperature and total pressure independent of other parts of the chemical network, it is essential that the pathway is isolated in the sense that all relevant reactions proceed along that pathway. The reactions along the chain must be much more important than reactions of chain molecules with other molecules. In this case, the concentrations of the chain molecules are determined by chain processes only.

The overall efficiency of this chain is then determined by the *slowest* step, defining the *critical reaction* and the corresponding *critical cluster* of size N_*. The nucleation rate J_* can then be approximated by the product of the concentration of the critical cluster $f(N_*)$ and the rate of the critical reaction $1/\tau(N_*)$

$$J_* = \frac{1}{\tau(N_*)} f(N_*). \tag{1}$$

This nucleation rate is the stationary flux through the reaction chain, determined by the "bottle neck" of the slowest reaction step.

Depending on the physical and chemical conditions, the concentrations of all species along the pathway can be determined by means of chemical or kinetic equilibrium calculations. If the flow of clusters up to the critical size cannot

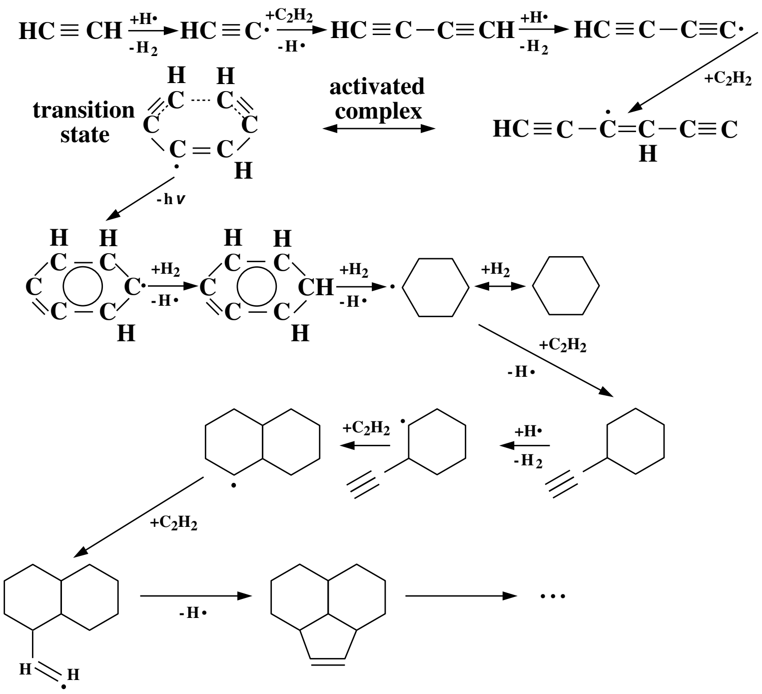

FIGURE 5. Chemical pathway to PAHs in a C-star envelope. The aromatic molecules larger than benzyne are represented by their skeleton of localized C–C bonds only (i.e. hexagons correspond to aromatic rings, while double and triple lines represent double or triple C–C bonds, respectively), dots mark free radical sites (adopted from Goeres 1993 [18]).

be regarded as a stationary process,a corresponding set of rate equations has to be solved. There are, however, many fewer species and reactions to be considered than in the full network approach.

The chemical pathway approach has been applied to the formation of dust nuclei via PAH precursors in the envelopes of cool C-stars [19,18,20]. It turns out that both the most efficient pathway and the critical cluster are not unique but depend on the position in the p-T-plane. An exemplary pathway at $T = 800$ K and $p = 10^{-9}$ bar including the important first ring closure is presented in Figure 5 up to acenaphthalene. Further growth takes place through the addition of acetylene at the boundary of the PAH leading to larger molecules which usually are composed of hexagons and pentagons. Although adding hexagons to the PAH is preferable with regard to maximizing the bond energy, pentagons minimize the entropy loss.

The built-in pentagons may also be the key to the transitions from flat PAH-platelets to three-dimensional structures [21]. This is due to the fact that the presence of pentagons in the usual planar honeycomb lattice causes stress and introduces a curvature (cf. the Buckminsterfullerene C_{60}). Another possibility proposed for the 3D-transition is that of coagulation leading either to the stacking of several planar PAHs similar to a tiny graphitic crystal [22] or to out-of-plane clustering [23].

Concepts of modified classical nucleation theory

The original classical nucleation theory was developed to describe of the nucleation of monomers in low supersaturation, low nucleation flux systems, e.g. the formation of droplets in supersaturated vapour [24]. Therefore, the classical approach assumes that the thermodynamic quantities of small clusters such as their Gibbs free energy of formation can be approximated in terms of macroscopic parameters such as the surface energy of the bulk solid [25,26]. The surface tension concept was introduced in order to determine the equilibrium cluster size distribution.

A fundamental restriction is that clusters are described by a single quantity, usually the number of constituting monomers N. Consequently, classical nucleation theory can only be applied to the formation of homogeneous clusters, e.g. pure C_N or $(SiO)_N$. Also, no distinction is made between the various isomers of an N-mer. For this special case, however, classical nucleation theory yields a formula analogous to Eq. (1) where the equilibrium abundance of the critical cluster is now determined from thermodynamic arguments and the size of the critical cluster is given by the minimum of the equilibrium cluster distribution function. Regarding the further growth of critical clusters up to sizes of the order of N_ℓ it is assumed that growth processes are sufficiently fast for stationarity to establish so that the formation rate of clusters of size N_ℓ is equal to that of the critical cluster.

Keeping in mind the basic restriction to homogeneous clusters and the neglect of isomeric variety, several important modifications and extensions of the original theory have been made. First, the description of cluster growth is not limited to the mere addition of impinging i-mers of the same type, which often are not present in the gas phase at all, but also allows for heteromolecular chemical reactions leading to a net deposit of monomers on the cluster [27,9]. Secondly, the surface tension concept could in principal be abandoned in favor of the actual size dependent values if these data were availabe, for example, from laboratory experiments. In a recent paper, a kinetic nucleation theory for chemical non-equilibrium in the gas phase and thermal non-equilibrium between the gas and the clusters was presented [28]. Thus, the critical assumptions of the conventional theory namely the existence of an equilibrium precondensation cluster distribution and the equality of kinetic gas temperature and cluster vibrational temperature have been abandoned. These assumptions have been criticized in the past with regard to their validity in astrophysical environments [29].

Due to its simplifying concepts modified classical nucleation theory has the advantage of being easier to use and of requiring less data than described approaches previously. The various applications include the formation of pure carbon grains in C-stars [30,31,27] as well as the condensation of $(SiO)_N$, $(MgS)_N$, $(Fe)_N$ [32] and recently $(MgO)_N$ clusters [33] in oxygen-rich M-star envelopes. Few investigations [34,9,35] deal with astrophysical condensates, the nominal monomer of which is absent from the gas phase, e.g. $(Al_2O_3)_N$ and $(MgSiO_3)_N$.

Grain growth

Further growth of the seed particles is the second step in dust formation. While the nucleation rate provides the *number of grains* per hydrogen particle, the growth rate determines the *fraction of material actually condensed* into grains, and the *size distribution* of dust particles. An important point to note is that the elemental composition of the seed material does not neccessarily have to be the same as the composition of the bulk of the grain mass.

On the contrary, it seems likely that high-temperature condensates of rare elements (e.g. titanium or aluminium compounds) could serve as heterogeneous nucleation seeds for other materials at a point further out in the cooling stellar outflow. The mantle material will usually be built from more abundant elements (cf. Figure 4). This view is supported by the microanalysis of presolar grains extracted from meteorites in which inclusions of TiC crystals were found [36,1]. The sticking efficiencies for different substrate and adsorbate materials, however, are probably reduced [37], so that building the first layer of a new grain mantle material may be difficult. A further investigation of this important aspect seems to be necessary.

Carbon grains

There is still some debate regarding the growth efficiency, and the possible composition and structure of carbonaceous grains produced in carbon-rich circumstellar envelopes (CSE) of cool late-type stars. Suggestions include (see e.g. [38,39] and references therein): (hydrogenated) amorphous carbon, graphite, PAHs, soot, diamond-like carbon, fulleranes, coal and other kerogen-like materials. Several of the proposed materials differ only in terms of the relative proportion of the various possible bond types of carbon atoms. Although these parameters generally do not determine the complex, probably non-crystalline structure of a carbonaceous grain, the dominant hybridisation state of its constituting atoms can nevertheless serve as a decisive characteristic. Three important configurations can be considered: sp^2 and sp^3 hybridised carbon, and also carbon atoms which are part of an aromatic ring structure[1].

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1) Note that sometimes the aromatic bonding configuration is refered to as sp^2 by other authors.

Spectral features due to sp^1 bonded carbon are not evident in astronomical spectra [40].

In CSE of C-stars the dangling bonds of carbon atoms on a grain surface are expected to be saturated by hydrogen atoms. Efficient grain growth, i.e. addition of carbon to the surface, however, will involve neutral–radical reactions of hydrocarbon species with the surface atoms. Thus, growth reactions preferably occur at temporary radical sites, where an H-atom has been abstracted (dehydrogenation).

This scenario has interesting analogies to soot formation in fuel-rich flames as described in the terrestrial combustion literature [41–43] and to gas-activated diamond film deposition [44]. Exploiting these analogies relevant aspects of the grain surface chemistry on pre-existing carbonaceous seed particles were described with respect to cosmic conditions [39,45]. In order to determine the reactive fraction of the grain surface, the probability w of a particular carbon surface site $\mathcal{C}$ to have a free radical site ($\mathcal{C}\cdot$) rather than being saturated ($\mathcal{C}$–H) by a hydrogen-atom can be calculated by treating the production of radical sites by the following reaction scheme:

$$\mathrm{r1}: \quad \mathcal{C}{-}\mathrm{H} + \mathrm{H} \rightleftharpoons \mathcal{C}\cdot + \mathrm{H}_2 \tag{2}$$

$$\mathrm{r2}: \quad \mathcal{C}\cdot + \mathrm{H} \rightleftharpoons \mathcal{C}{-}\mathrm{H}. \tag{3}$$

From Fig. 6 it can be seen that for high temperatures and H/H_2 ratios hydrogen is abstracted more easily. For $T \lesssim 1600$ K radical surface sites are predominantely formed at sp^3 surface sites.

After radical sites have been formed, gaseous hydrocarbon species colliding with the grain may bond to these active surface sites (chemisorption). An estimate for the accretion rate $1/\tau_{\mathrm{acc}}$ of carbon onto a grain is given by the product of the number density of the growth species acetylene contributing 2 carbon atoms, the reactive cross-section $w\sigma_{\mathrm{d}}$ of the grain and a suitably defined relative collision velocity v_{rel} averaged over the grain surface

$$\frac{1}{\tau_{\mathrm{acc}}} = 2n_{\mathrm{C_2H_2}} \langle v_{\mathrm{rel}} w \sigma_{\mathrm{d}} \rangle. \tag{4}$$

In the process of chemisorption of C_2H_2 the triple bond between the two carbon atoms is broken. The resulting structure is then incorporated into the lattice by reconstruction or rearrangement (isomerization) processes and forms a new part of the grain surface (cf. Figure 7) as long as the dust temperature $T_{\mathrm{d}} \lesssim 1600$ K.

So far, we have described the growth mechanism for pure carbon grain mantles. The 11.3 μm emission feature, which is observed in the shells of many carbon-rich red giants [47,48] and is attributed to silicon carbide, suggests the formation of C/SiC composite grains. From radiative transfer calculations a fraction by mass of only a few percent is deduced for solid SiC [49,50]. The question whether SiC is incorporated as impurities or carbon condenses onto

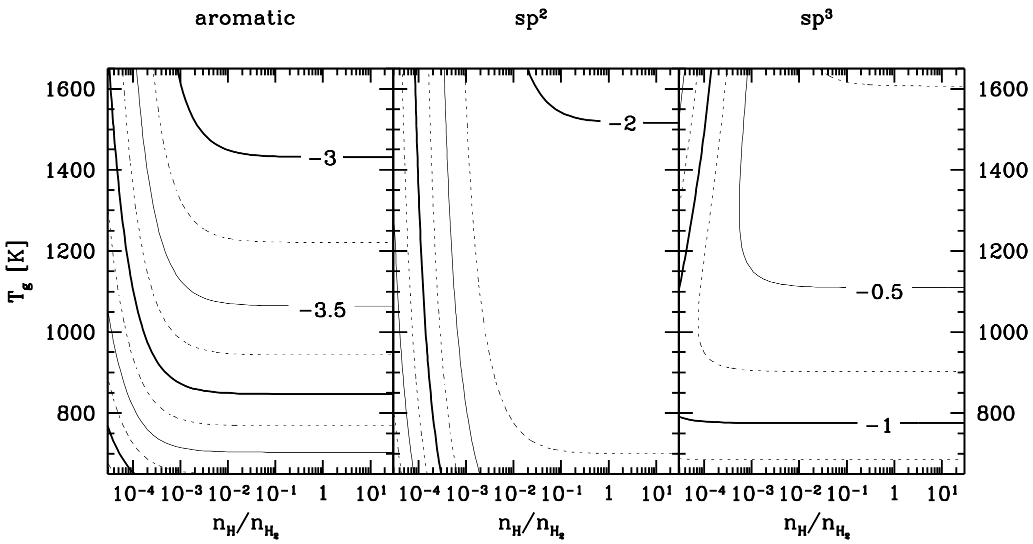

FIGURE 6. Contour plots of the dehydrogenation probability $\log w$ of a carbon surface site for different bonding configurations. Thick lines correspond to $\log w = -1, -2, \ldots$; thin lines to -0.5,-1.5, ...; dashed lines to -0.25,-0.75, ...; in the upper right corner $\log w \rightarrow 0$. (adopted from Krüger et al. 1996 [45]).

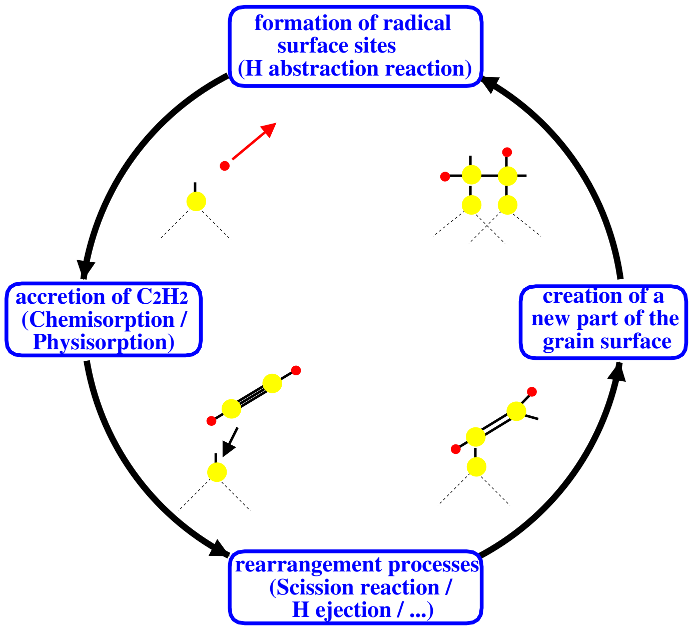

FIGURE 7. Schematic growth cycle of carbon grains (adopted from Patzer 1997 [46]).

SiC cores formed earlier in the stellar outflow [51,52] is still a matter of debate. However, laboratory findings [1] and also recent theoretical investigations [53] of this problem seem to indicate that SiC condensation in CSE is not a primary process but a secondary phenonemon taking place at the surface of already existing carbon grains.

Astronomical silicates

Considering the hierarchy of element abundances (cf. Figure 4), silicates and oxides of the heavy elements are expected to make the main contribution to the production of dust in oxygen-rich stellar outflows. However, for most of the thermodynamically stable dust materials (e.g. Al_2O_3, Mg_2SiO_4, Fe_2O_3,...) the corresponding monomer is absent from the gas phase. Consequently, if the stable minerals actually form the growth processes have to be heteromolecular and proceed in a sequence of surface reaction steps resulting in the net deposit of a "monomer" [54]. Unfortunately, data on the relevant surface reaction rates is rare and investigations concerned with dust formation in oxygen-rich environments usually concentrate on the thermodynamic stability of a solid assuming purely geometrical reaction rates, i.e. estimating sticking efficiencies to be approximately unity.

The condensation of a solid from the gas phase requires a supersaturated vapour. In other words, the actual concentration of the monomer in the gas phase has to be larger than its concentration corresponding to phase equilibrium, i.e. the vapour pressure of the solid material. In this case, the adsorption rate of dust forming gas particles onto stable surfaces is larger than the evaporation rate. When the nominal monomer is absent from the gas phase, the procedure is essentially the same. *Generalized supersaturation ratios* S_i for each species i contributing to the growth can be defined in an analogous manner [55] and are in principal given by the quotient of the actual number density n_i and an equilibrium density $\mathring{n}_i$ above the condensate refering to a hypothetical state of phase equilibrium. Such calculations to determine $\mathring{n}_i$ have been carried out [55] and were recently extended [56].

A curve of stability in the p-T-plane separates the regions where dust growth dominates or grains tend to evaporate, respectively (cf. Figure 8). On the curve of stability itself, the S_i equal unity and all reactions are completely balanced by their reverse reactions, i.e. detailed balancing prevails. As can be seen, a clear thermodynamic condensation sequence develops (cf. also [54] and references therein) where Aluminium compounds are the first potential dust materials to become stable followed by Magnesiumsilicates and finally Ironoxides. It should be kept in mind here, that whether or not this condensation sequence will actually be followed in a stellar outflow will depend both on kinetic considerations and the availability of nucleation seeds.

A straightforward method to determine the *rate* of dust growth once a solid material becomes stable consists of identifying the reactant that has the least collisional frequency onto a target grain, e.g. Aluminium in the case of solid Al_2O_3 formation, as the *key species* [34]. Each collision with this key species can be regarded as leading to the addition of one complete monomer of the

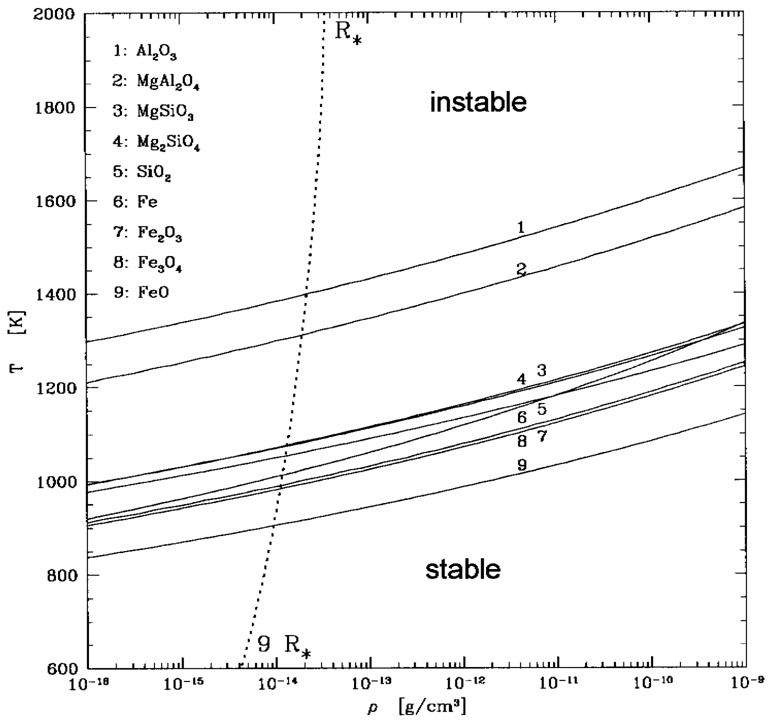

FIGURE 8. Equilibrium curves of stability for various dust materials. The dashed lines represents the trajectory of a representative M-star outflow (adopted from Breuer 1996 [56]).

final condensate to the grain. However, this approach makes no assertion about the reverse process, i.e. evaporation, and thus overestimates the net growth rate close to the curve of stability when the vapour is only marginally supersaturated.

Knowledge of the generalized supersaturation ratios S_i allows for a more detailed treatment [55]. For each growth species i a *net* deposition rate

$$\frac{1}{\tau_{\mathrm{net},i}} = \alpha_i n_i v_{\mathrm{rel}_i} \sigma_{\mathrm{d}} (1 - S_i^{-1})$$

can be calculated, where α_i is the sticking efficiency usually set equal to unity. Adding up these rates, net element fluxes onto the surface can be calculated. If all element fluxes relevant for the grain material under consideration are positive, growth of an existing nucleus is possible. In order to determine the total growth rate the element fluxes have to be reduced to values consistent with the stoichiometric composition of the compound [55].

Under the conditions in circumstellar environments a variety of solid materials of different composition can generally be stable and the material actually condensing on the grain does not neccessarily have to be one of stoichiometric composition. Possibly, some kind of mixture of materials or *composite* material will form, resulting in "dirty" grains. From laboratory physics, for example, a variety of silicates incorporating other metals like iron or magnesium are known which can certainly coexist in a grain [57,58]. The composition that forms only has to be compatible with certain boundary conditions so that the stoichiometric relations of the composite solid do not leave the settings given by the composition of the pure materials [55].

M-stars often are associated with chromospheres giving rise to an ionized chemistry. The free electrons present lead to a negative charge of the grains [59,60]. As a consequence, growth reactions between ionized atoms or molecules and the grains will probably be considerably enhanced. Until now, however, no quantitative description of this ionic growth mechanism seems to be available in the literature.

In any case, grain growth in a stellar wind proceeds along a sequence of changing chemical environments due to the cooling expansion flow of the gas and the consumption of condensable elements. Thus, an onion-shell like grain structure can be expected leading quite naturally to a *depth dependent* composition of the grains where each shell may consist of different pure or even continuously changing composite materials.

In conclusion, it can be said that although significant progress has been made in the understanding of astronomical silicate dust formation some important questions remain to be answered. This cannot be done solely on the grounds of thermodynamical arguments. Firstly, the microscopic structure of the grains, i.e. the detailed crystal structure that is created in the growth process, needs to be clarified. More important, the morphology of the grain surface as well as the sticking and chemical reaction efficiencies of molecules impinging on the surface have to be investigated in much more detail in order to proceed further.

Dynamical description of the dust component

After analyzing the microscopic growth processes of carbon and silicate grains, it turns out that the relevant growth timescales are comparable to the timescales governing the hydrodynamical evolution of the expanding stellar outflow. Therefore, grain growth has to be treated as a time dependent problem, in contrast to cluster nucleation which in most cases is a stationary process.

Usually, the dust component can be characterized by a size distribution function f of one parameter, e.g. the number of monomers N or the grain radius a. In many astrophysical scenarios it is not necessary to have the full information provided by the local size distribution function, but it suffices to use appropriately defined moments

$$K_i = \sum_{N_\ell}^{\infty} N^{i/d} f(N)$$

where $d = 2$ for planar and $d = 3$ for three-dimensional particles. These moments directly relate to the mean grain radius, the total dust mass etc. [9,55]. The time evolution of the dust component is then given by the following coupled system of equations

$$\frac{\mathrm{d}K_0}{\mathrm{d}t} = J_*$$
$$\frac{\mathrm{d}K_i}{\mathrm{d}t} = \frac{i}{d\tau_{\mathrm{net}}} K_{i-1} + N_\ell^{i/d} J_*$$

with τ_{net} being the net growth timescale. Fortunately, it turns out that the grain size spectrum can also be calculated with little effort both for homogeneous and onion-shell like grain structures by a functional derivative of equation for K_0 [61,55].

The main limitation of this rapid moment method is that only those continuous growth or destruction processes can be included in this scheme whose efficiency varies proportional to some integer power of the grain radius a. Furthermore, the treatment of discontinuous processes which eliminate grains from the size distribution function is not possible. Thus, coagulation or fragmentation due to grain-grain collisions cannot be described by means of the moment method. The only way to avoid this inherent limitation is to discretize the grain size distribution function, i.e. to divide the mass spectrum into *bins* and treat each bin as a separate species [62–64].

All of the previously mentioned methods either assume the center of mass of the dust component to be comoving with the gas or even treat the dust-gas mixture as a single component, i.e. under the assumption of negligible net motion of the grains relative to the bulk of the gas (position coupling). For one-dimensional stationary flows a special differential form of a balance or conservation equation for the grain size distribution function was derived [65] that takes explicitly into account possible size-dependent, directed drift velocities $v_{\mathrm{d}}(a)$ of the grains. It also allows to describe changes of the distribution function by both continuous (e.g. size-dependent grain growth or sputtering) and discontinuous processes such as coagulation, (possibly complete) vaporization or shattering [65,66]. The balance equation can be solved by means of a computational multi-component method that disretizes the size distribution on a Lagrange-like grid which *moves* in grain size space according to the size evolution of selected sample grains.

MODELING COSMIC DUST PRODUCTION IN AGB STARS

The extended atmospheres of cool red giants are places of considerable dust formation accompanied by substantial mass loss. In fact, they provide by far the largest contribution to the stellar mass injection rate into the ISM. For a reliable modeling of these objects a self-consistent description of the system is required including a detailed and simultaneous treatment of hydrodynamics, thermodynamics, radiative transfer and chemistry as well as dust nucleation and growth. Two different scenarios and modeling approaches can be distinguished.

Stationary dust driven winds

The upper right region of the HR diagram is the domain of stellar objects showing extremely high mass loss. A necessary condition for their stellar winds to be *driven* by radiation pressure on dust is that efficient grain formation has to occur within the subsonic region of the outflow. A variety of stationary models have been published and the status of these dust driven wind models has recently been surveyed in an extensive review article [67]. Here, we restrict ourselves to the presentation of the size distribution of stardust injected into the ISM as it results from models of typical C-stars in consideration of a size dependent grain drift and a size dependent growth rate [68,69].

Figure 9 depicts plots of $f(a)$ and $a^3 f(a)$ which represent the grain size distribution and the corresponding amount of condensate, respectively. In the upper left panel of Figure 9 the maximum of the curve is approximately the *number* of grains formed. Obviously, in the negligible drift case the total number of grains is much higher (almost a factor of two) than in the case where drift is included, although the position of the maximum does not vary much. In the upper right panel of Figure 9 the area below the curve corresponds to the *total amount of condensed material.* The consideration of drift leads to a significant shift of the distribution of condensed matter towards larger grain radii. The average grain mass is more than twice as large, and the dust-to-gas mass ratio $\dot{M}_{\rm d}/\dot{M}_{\rm g}$ injected into the ISM is increased by about 30% compared to the case when drift is neglected.

Long-period Variables

Most red giants surrounded by a circumstellar dust shell are pulsating variables with pulsation periods of ~ 1 yr. Here, the stationary approach fails and modeling these objects requires a fully time-dependent treatment [70–72]. The resulting models turn out to have a rather complex structure, e.g. multiple dust layers, dust induced or dust accelerated shocks (for a comprehensive overview see [73]) and are able to explain the observed lightcurves and brightness profiles of carbon-rich circumstellar dust shells [74,75].

Concerning dust formation in long-period variables we only present the mean size distribution in the outer region of the circumstellar shell of a time-dependent model for the extreme carbon star AFGL 3068 assuming position coupling of dust and gas [76]. The presence of periodic pulsations in these

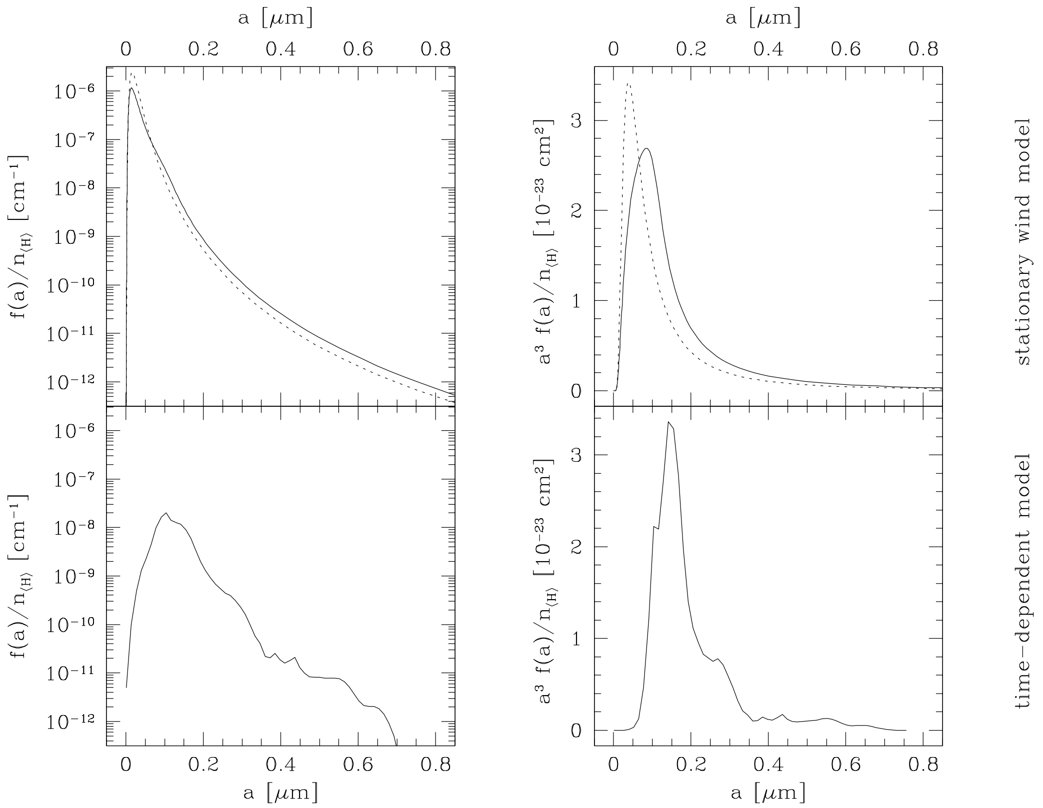

FIGURE 9. Grain size distribution for a stationary model of a typical C-star (after injection into the ISM) (adopted from Krüger & Sedlmayr 1997 [68]) and for a time-dependent model for AFGL 3068 (averaged over the local distribution functions between 30 and 60 R_*) (adopted from Winters et al. 1997 [76]). Dashed lines indicate the results neglecting drift (the time-dependent model was calculated assuming position coupling). **Left panels:** Logarithmic scale. **Right panels:** Weighted by a^3.

models reduces the hydrodynamical timescale to 1 yr or less. Hence, the occurrence of very large grains ($a \gtrsim 1\mu$m) which are an artifact of the stationary models is suppressed (cf. lower left panel of Figure 9). The lower right panel of Figure 9 shows the volume weighted size distribution; the volume weighted particle radius is the relevant quantity determining the extinction coefficient of the grains. The position of the maximum of $a^3 f(a)$ would thus be derived as the dominant particle size from a radiative transfer model based on the small particle limit of Mie theory [76].

CONCLUDING REMARKS

Thanks to modern computing power and refined numerical methods, the modeling of dust forming objects, though not treated in great detail here, has reached an impressive state considering the overall complexity of the problem.

In fact, remaining shortcomings are often due to a lack of suitable microphysical input rather than to macroscopic modeling. Despite the progress achieved in the last decade, the level of sophistication in describing the details of dust formation nowadays lags behind that of the reliable modeling of circumstellar outflows.

Nevertheless, our understanding of the processes leading to cosmic dust condensation has become reasonably thorough, especially with regard to carbonaceous grains. In fact, the results of recent laboratory investigations [1] of graphitic material from the Murchison meteorite are in good agreement with theoretical predictions: It was found that the extracted grains have a core-mantle structure; the nanocrystalline cores are compact masses consisting of randomly orientated sheets of carbon atoms which resemble clusters of PAHs. The absence of layering order as apparent from electron diffraction experiments is attributed to occasional replacements of hexagons by pentagons. Within some of the grains inclusions of crystalline carbides of less abundant metals, such as Ti, Zr and Mo were detected, while SiC occurs in the grain-*mantle*. It seems plausible to identify these metal carbides with the postulated high-temperature condensates. In any case, carbon mantles were added by vapour deposition onto the existing cores, which evidently served as nucleation centers for further growth.

Interestingly, many details of these experimental findings support dust formation theory as it was presented in this contribution, although stardust has undergone some serious processing in the ISM — thermal, mechanical and otherwise — before it was incorporated into meteoritic material.

Unfortunately, finding presolar oxides has been difficult because solar system oxygen-rich minerals formed in such large amounts that it now requires detailed analysis of very many individual grains before one can expect to find just one single presolar oxygen-rich grain. Furthermore, the techniques used to separate carbon-rich grains from oxygen-rich minerals in stony meteorites tend to destroy most oxides. Some refractory oxides survive these procedures, which has allowed the discovery of a few Al_2O_3 grains in meteorites [77]. Therefore, the challenge to develop and improve theoretical methods of the microphysics of grain formation remains, particularly in the field of inorganic compounds.

Regarding the future, the most important problems to be clarified are: What are the first high-temperature seeds and how do they form? What is the efficiency of seed production? What seeds are present in sufficient quantity to allow massive dust condensation? What is the surface morphology of grains and what chemical reactions with gas phase species can occur at the grain surface? Is there a mechanism to describe surface growth processes by means other than the usually adopted thermodynamic approach but avoiding the expenditure of *ab initio* molecular dynamics simulations, e.g. by the application of (semi-)empirical interaction potentials?

Much work addressing these problems is currently under way. A necessary prerequisite for success, however, is that fundamental input data like reaction coefficients, Gibbs free energies of formation and sticking efficiencies for atoms, molecules and clusters involved in cosmic dust formation are made available by laboratory measurements or appropriate quantum mechanical calculations [78].

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