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Vapor-phase synthesis of nanoparticles

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Abstract

An overview of methods for preparing nanoparticles in the vapor phase is given, and recent advances are reviewed. Developments in instrumentation for monitoring vapor-phase synthesis of nanoparticles and in modeling these processes are also included. The most important developments relate to improved control and understanding of nanoparticle aggregation and coalescence during synthesis, and to methods for producing multi-component nanoparticles.

Keywords: Nanoparticle; Nanocrystal; Aerosol

1. Introduction

Nanoparticles are viewed by many as fundamental building blocks of nanotechnology. They are the starting point for many ‘bottom-up’ approaches for preparing nanostructured materials and devices. As such, their synthesis is an important component of rapidly growing research efforts in nanoscale science and engineering. Nanoparticles of a wide range of materials can be prepared by a variety of methods. This review focuses on methods for preparing nanoparticles in the vapor phase. It provides broad but shallow coverage of this field. An attempt to describe advances in a wide range of synthesis methods has been made, at the expense of detailed coverage of any particular technique. More detailed coverage of flame synthesis of nanoparticles is given by Kammler et al. [1]. Gas-phase synthesis of nanoparticles for electronics-related applications was reviewed by Kruis et al. [2]. Hahn [3] presented a useful overview of gas-phase synthesis of nanocrystalline materials.

Synthesis of nanoparticles in the liquid phase is not covered here. Reviews in this area have been presented by Grieve et al. [4], Trindade et al. [5], and Murray et al. [6], among others. The production of fullerenes, carbon nanotubes, and related materials is also outside the scope of this review. Finally, we do not attempt to discuss applications of nanoparticles produced in the gas phase, though these clearly provide the ultimate driving force for the advances in synthesis that are covered here.

In vapor-phase synthesis of nanoparticles, conditions are created where the vapor phase mixture is thermodynamically unstable relative to formation of the solid material to be prepared in nanoparticulate form. This includes usual situation of a supersaturated vapor. It also includes what we might call ‘chemical supersaturation’ in which it is thermodynamically favorable for the vapor-phase molecules to react chemically to form a condensed phase. If the degree of supersaturation is sufficient, and the reaction/condensation kinetics permit, particles will nucleate homogeneously. Once nucleation occurs, remaining supersaturation can be relieved by condensation or reaction of the vapor-phase molecules on the resulting particles, and particle growth will occur rather than further nucleation. Therefore, to prepare small particles, one wants to create a high degree of supersaturation, thereby inducing a high nucleation density, and then immediately quench the system, either by removing the source of supersaturation or slowing the kinetics, so that the particles do not grow. In most cases, this happens rapidly (milliseconds to seconds) in a relatively uncontrolled fashion, and lends itself to continuous or quasi-continuous operation. This contrasts with many colloidal syntheses of nanoparticles that are carried out in discrete

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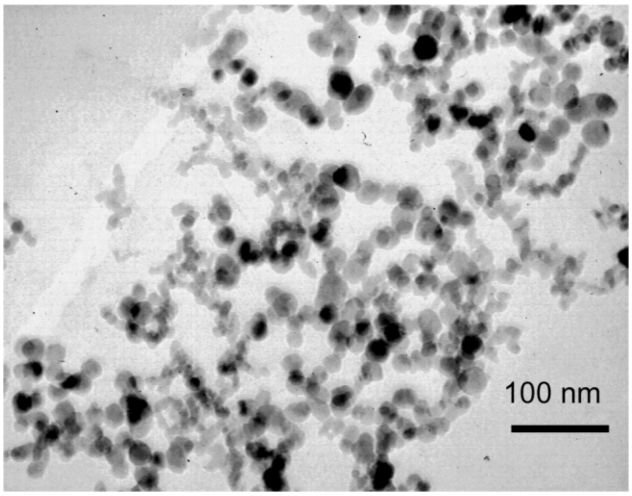

Fig. 1. TEM image of agglomerated nanoparticles typical of those produced in many vapor-phase processes. These particular particles are silicon produced by laser pyrolysis of silane, but the degree of polydispersity and agglomeration is typical of many vapor-phase processes in which no special efforts have been made to avoid agglomeration or narrow the size distribution of the primary particles.

batches under well-controlled conditions with batch times of hours to days.

Once particles form in the gas phase, they coagulate at a rate that is proportional to the square of their number concentration and that is only weakly dependent on particle size. At sufficiently high temperature, particles coalesce (sinter) faster than they coagulate, and spherical particles are produced. At lower temperatures, where coalescence is negligibly slow, loose agglomerates with quite open structures are formed. At intermediate conditions, partially sintered non-spherical particles are produced. If individual, non-agglomerated nanoparticles are desired, control of coagulation and coalescence is crucial. In contrast to the liquid phase, where a dispersion of nanoparticles can be stabilized indefinitely by capping the particles with appropriate ligands, nanoparticles in the gas phase will always agglomerate. So, by non-agglomerated nanoparticles, we usually mean particles agglomerated loosely enough that they can be redispersed without Herculean effort, as compared to hard (partially sintered) agglomerates that cannot be fully redispersed. Fig. 1 illustrates typical degrees of agglomeration and polydispersity obtained in gas phase processes when no special efforts have been made to control agglomeration or narrow the particle size distribution. In the cases of carbon black, fumed silica, and pigmentary titania, such particles are produced commercially in huge quantities (millions of metric tons per year). In some applications, such as catalysis, agglomerates with an open structure like those in Fig. 1 are desired. However, in many potential applications non-agglomerated spherical nanoparticles of uniform size are needed. Thus, many of the advances reviewed here relate to understanding and controlling particle aggregation and coalescence to produce non-agglomerated particles of desired size and narrow size distribution.

2. Recent examples of and advances in particle synthesis techniques

In this section, recent examples of and advances in gas-phase methods for preparing nanoparticles are reviewed. One useful way of classifying such methods is by the phase of the precursor and the source of energy used to achieve a state of supersaturation. This section is structured around such a classification.

2.1. Methods using solid precursors

One general class of methods of achieving the supersaturation necessary to induce homogeneous nucleation is to vaporize the material into a background gas and then cool the gas.

2.1.1. Inert gas condensation

Perhaps the most straightforward method of achieving supersaturation is to heat a solid to evaporate it into a background gas, then mix the vapor with a cold gas to reduce the temperature. This method is well suited for production of metal nanoparticles, since many metals evaporate at reasonable rates at attainable temperatures. By including a reactive gas, such as oxygen, in the cold gas stream, oxides or other compounds of the evaporated material can be prepared. Wegner et al. [7•] presented a detailed, systematic modeling and experimental study of this method, as applied to preparation of bismuth nanoparticles, including both visualization and computational fluid dynamics simulation of the flow fields in their reactor. They clearly showed that they could control the particle size distribution by controlling the flow field and the mixing of the cold gas with the hot gas carrying the evaporated metal. Other advances in this method have been in preparing composite nanoparticles and in controlling the morphology of single-component nanoparticles by controlled sintering after particle formation. Maisels et al. [8•] prepared composite nanoparticles of PbS with Ag by separate evaporation/condensation of the two materials followed by coagulation of oppositely charged PbS and Ag particles selected by size and charge. Ohno [9] prepared Si/In, Ge/In, Al/In and Al/Pb composite nanoparticles by condensation of In or Pb onto Si, Ge, or Al particles prepared by inert gas condensation and brought directly into a second condensation reactor. Nakaso et al. [10•] conducted a detailed experimental and theoretical study of the in-flight sintering (coalescence) of gold nanoparticle agglomerates after evaporation/condensation synthesis. They were able to differentiate between different sintering mechanisms, and extracted rate constants for the restructuring process from their experiments. Similarly, Nanda et al. [11] studied the in-flight sintering of PbS nanoparticles. They were able to tune the band-gap of these semiconductor nanoparticles by changing the particle size and morphology.

2.1.2. Pulsed laser ablation

Rather than simply evaporating a material to produce supersaturated vapor, one can use a pulsed laser to vaporize a plume of material that is tightly confined, both spatially and temporally. This method can generally only produce small amounts of nanoparticles. However, laser ablation can vaporize materials that cannot readily be evaporated. Marine et al. [12] presented a recent analysis of this method, in which they also reviewed its development. Nakata et al. [13•] used a combination of laser-spectroscopic imaging techniques to image the plume of Si atoms and clusters formed during synthesis of Si nanoparticles. They investigated the dependence of the particle formation dynamics on the background gas, and found that it was substantial. Some other recent examples include the preparation of magnetic oxide nanoparticles by Shinde et al. [14], titania nanoparticles by Harano et al. [15], and hydrogenated-silicon nanoparticles by Makimura et al. [16].

2.1.3. Spark discharge generation

Another means of vaporizing metals is to charge electrodes made of the metal to be vaporized in the presence of an inert background gas until the breakdown voltage is reached. The arc (spark) formed across the electrodes then vaporizes a small amount of metal. This produces very small amounts of nanoparticles, but does so relatively reproducibly. Weber et al. [17•] recently used this method to prepare well-characterized nickel nanoparticles for studies of their catalytic activity in the absence of any support material. By preparing the nanoparticles as a dilute aerosol they were able to carry out reactions on the freshly prepared particles while they were still suspended. Metal-oxides or other compounds can be prepared by using oxygen or another reactive background gas. Rather than having the background gas present continuously, it can be pulsed between the electrodes at the same time that the arc is initiated, as in the pulsed arc molecular beam deposition system described by Rexer et al. [18].

2.1.4. Ion sputtering

A final means of vaporizing a solid is via sputtering with a beam of inert gas ions. Urban et al. [19] recently demonstrated formation of nanoparticles of a dozen different metals using magnetron sputtering of metal targets. They formed collimated beams of the nanoparticles and deposited them as nanostructured films on silicon substrates. This process must be carried out at relatively low pressures (~1 mTorr), which makes further processing of the nanoparticles in aerosol form difficult.

2.2. Methods using liquid or vapor precursors

An alternate means of achieving the supersaturation required to induce homogeneous nucleation of particles is chemical reaction. Chemical precursors are heated and/or mixed to induce gas-phase reactions that produce a state of supersaturation in the gas phase.

2.2.1. Chemical vapor synthesis

In this approach, vapor phase precursors are brought into a hot-wall reactor under conditions that favor nucleation of particles in the vapor phase rather than deposition of a film on the wall. It is called chemical vapor synthesis or chemical vapor condensation in analogy to the chemical vapor deposition (CVD) processes used to deposit thin solid films on surfaces. This method has tremendous flexibility in producing a wide range of materials and can take advantage of the huge database of precursor chemistries that have been developed for CVD processes. The precursors can be solid, liquid or gas at ambient conditions, but are delivered to the reactor as a vapor (from a bubbler or sublimation source, as necessary).

There are many good examples of the application of this method in the recent literature. Ostraat et al. [20•] have demonstrated a two-stage reactor for producing oxide-coated silicon nanoparticles that have been incorporated into high-density nonvolatile memory devices. By reducing the silane precursor composition to as low as 10 parts per billion, they were able to produce non-agglomerated single-crystalline spherical particles with mean diameter below 8 nm. This is one of relatively few examples of a working microelectronic device in which vapor-phase synthesized nanoparticles perform an active function. In other recent examples of this approach, Magnusson et al. [21] produced tungsten nanoparticles by decomposition of tungsten hexacarbonyl and Nasibulin et al. [22] produced copper and copper oxide nanoparticles from copper acetylacetonate.

Another key feature of chemical vapor synthesis is that it allows formation of doped or multi-component nanoparticles by use of multiple precursors. Schmechel et al. [23] prepared nanocrystalline europium doped yttria (Y_2O_3:Eu^{3+}) from organometallic yttrium and europium precursors. Senter et al. [24] incorporated erbium into silicon nanoparticles using disilane and an organometallic erbium compound as precursors. Srdić et al. [25] prepared zirconia particles doped with alumina. It is also possible to make composite nanoparticles where one material is encapsulated within another. A particularly promising approach to this is the sodium metal/metal halide chemistry used by Ehrman et al.

[26•]. In this approach, a halide, such as $SiCl_4$, is reacted with sodium vapor in a heated furnace to produce NaCl-encapsulated particles. For example $SiCl_4$ reacts with Na to produce NaCl-encapsulated Si particles. The salt-encapsulation can potentially be used to prevent agglomeration of the particles, and the salt can then be washed away in a post-processing step.

2.2.2. Spray pyrolysis

Rather than delivering the nanoparticle precursors into a hot reactor as a vapor, one can use a nebulizer to directly inject very small droplets of precursor solution. This has been called spray pyrolysis, aerosol decomposition synthesis, droplet-to-particle conversion, etc. Reaction often takes place in solution in the droplets, followed by solvent evaporation. Recent examples of this include preparation of TiO_2 nanoparticles by Ahonen et al. [27] and copper nanoparticles by Kim et al. [28].

2.2.3. Laser pyrolysis/photothermal synthesis

An alternate means of heating the precursors to induce reaction and homogeneous nucleation is absorption of laser energy. Compared to heating the gases in a furnace, this allows highly localized heating and rapid cooling, since only the gas (or a portion of the gas) is heated, and its heat capacity is small. Heating is generally done using an infrared (CO_2) laser, whose energy is either absorbed by one of the precursors or by an inert photosensitizer such as sulfur hexafluoride. The silicon particles shown in Fig. 1 were prepared in our laboratory by laser pyrolysis of silane. Nanoparticles of many materials have been made using this method. A few recent examples are MoS_2 nanoparticles prepared by Borsella et al. [29], SiC nanoparticles produced by Kamlag et al. [30], and Si nanoparticles prepared by Ledoux et al. [31,32]. Ledoux et al. use a pulsed CO_2 laser, thereby shortening the reaction time and allowing preparation of even smaller particles.

2.2.4. Thermal plasma synthesis

Yet another means of providing the energy needed to induce reactions that lead to supersaturation and particle nucleation is to inject the precursors into a thermal plasma. This generally decomposes them fully into atoms, which can then react or condense to form particles when cooled by mixing with cool gas or expansion through a nozzle. Heberlein et al. [33•] have applied these methods to the production of nanoparticles of SiC and TiC for nanophase hard coatings.

2.2.5. Flame synthesis

Rather than supplying energy externally to induce reaction and particle nucleation, one can carry out the particle synthesis within a flame, so that the heat needed is produced in situ by the combustion reactions. This is by far the most commercially successful approach to nanoparticle synthesis—producing millions of metric tons per year of carbon black and metal oxides. However, the coupling of the particle production to the flame chemistry makes this a complex process that is rather difficult to control. It is primarily useful for making oxides, since the flame environment is quite oxidizing. Recent advances are expanding flame synthesis to a wider variety of materials and providing greater control over particle morphology. Janzen and Roth [34•] recently presented a detailed study of flame synthesis of γ-Fe_2O_3 nanoparticles, including comparison of their results to a theoretical model. Lee and Choi [35•] controlled nanoparticle morphology by using a CO_2 laser to re-heat flame-synthesized titania and silica nanoparticle agglomerates, thereby sintering them in situ. Wegner et al. [36] controlled the size and morphology of titania nanoparticles by extracting them from the flame through a critical flow nozzle, quenching particle growth and agglomeration. Kammler et al. [37•] have shown that they can influence flame conditions and primary particle size by application of a DC electric field to the particle synthesis flame.

2.2.6. Flame spray pyrolysis

Rather than injecting vapor precursors into the flame, one can directly spray liquid precursor into it. This process is generally called flame spray pyrolysis. This method allows use of precursors that do not have sufficiently high vapor pressure to be delivered as a vapor. Mädler et al. [38•] presented a very detailed study of this method, as applied to the synthesis of silica particles from hexamethyldisiloxane.

2.2.7. Low-temperature reactive synthesis

For particular materials, it is possible to react vapor-phase precursors directly without external addition of heat, and without significant production of heat. Sarigiannis et al. produced ZnSe nanoparticles [39] from dimethylzinc-trimethylamine and hydrogen selenide by mixing them in a counter-flow jet reactor at room temperature. Apparently the heat of reaction was sufficient to allow crystallization of the particles without substantially increasing the gas temperature. This is an intriguing result, because it is one of few methods reported for vapor-phase preparation of compound semiconductor nanoparticles that are usually produced by colloidal chemistry.

2.3. Advances in instrumentation

Because vapor-phase nanoparticle synthesis often takes place on short timescales, in small regions of a reactor, and in complex mixtures, improvements in methods for characterization of reactor conditions and particle formation are essential to improved understanding and control of particle formation. Thus, a few examples of the current state-of-the art are included here. Arabi-Katbi et al. [40•] used Fourier transform infrared spectroscopy in both emission and transmission modes to simultaneously characterize gas temperature, gas concentrations, particle temperature and particle concentration during flame synthesis of titania. Cho and Choi [41•] combined localized thermophoretic sampling and in situ light scattering measurements to characterize particle concentration, size, and morphology during flame synthesis of silica nanoparticles. Janzen et al. [42] compared particle mass spectrometry with TEM imaging of extracted samples from low-pressure reactors, and saw good agreement for ZnO and Fe_2O_3 nanoparticles, but poorer agreement for GeO_2 nanoparticles.

2.4. Advances in modeling of nanoparticle synthesis

Because in situ characterization and control of many vapor-phase nanoparticle syntheses is difficult, modeling studies can play an important role in the development and improvement of these processes. Several of the studies cited above had significant modeling components. Some additional advances in the modeling of vapor-phase particle synthesis are included here. It is increasingly possible to compute particle nucleation rates based on detailed chemical reaction kinetics in cases where nucleation does not occur by simple condensation of a supersaturated vapor. Girshick et al. [43–45] have done this for several silicon-based systems. There have recently been many important developments in modeling multidimensional particle size distributions, where both particle volume and surface area or some other pair of particle characteristics are explicitly treated. These include sectional methods like those presented by Mühlenweg et al. [46•], Tsantilis et al. [47•], Lee et al. [48], and Jeong and Choi [49], as well as bivariate moment methods like that presented by Rosner and Pyykönen [50•] and Monte Carlo methods like that presented by Rosner and Yu [51]. In a similar vein, Efendiev et al. have developed methodologies for modeling simultaneous coagulation and phase segregation in multi-component particles containing mutually immiscible phases, using both a hybrid sectional-moment approach [52•] and a Monte Carlo-based method [53]. Continuing improvements in simulation methodologies, along with inevitable advances in computing power, are beginning to make possible the coupling of detailed chemical reaction kinetics, multidimensional particle size distributions, and computational fluid dynamics simulations in two or even three dimensions to create models that quantitatively describe the details of particle formation processes and that compare reasonably with experiment.

3. Conclusions

A rather broad but shallow review of recent advances in vapor-phase synthesis of nanoparticles has been presented. New approaches and improved understanding of old approaches for improving control of particle size, morphology, and polydispersity are appearing regularly. The variety of materials that can be prepared as nanoparticles in the vapor phase is rapidly growing, and includes multi-component and doped materials. Advances in instrumentation and modeling are improving our detailed understanding of particle formation processes. In the near future, we can expect these activities to continue at a rapid pace, and to result in more examples of vapor-phase synthesized nanoparticles playing important roles as building blocks of nanotechnology.

Acknowledgments

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