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Chemical and Physicochemical Properties of Submicron Aerosol Agglomerates

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Abstract

This is the final report of a three-year, Laboratory Directed Research and Development (LDRD) project at Los Alamos National Laboratory. The formation of nanometer-sized aerosol particles in a premixed methane flame from both solid-phase aerosol precursors and gas-phase precursors was investigated. Techniques were developed to determine the distribution of the individual chemical species as a function of agglomerate size by using inductively coupled plasma atomic emission spectroscopy (ICP-AES). To determine the distribution of chemical species both from particle to particle and within the particles on a nanometer scale, we used the analytical electron microscopy techniques of energy dispersive x-ray spectrometry (EDS) and electron energy loss spectrometry (EELS) coupled with transmission electron microscopy (TEM). The observed distribution of individual chemical species as a function of agglomerate size was linked to the material properties of the solid-phase precursors. For aerosol formed from gas-phase precursors by gas-to-particle conversion, the distribution of species on a nanometer scale was found to correspond to the equilibrium phase distribution expected from equilibrium for the system at the flame temperatures.

Background and Research Objectives

Nanometer-sized aerosol particles and their agglomerates are an aerosol byproduct of high-thermal-energy processes, including waste incineration, coal combustion, explosions, and metallurgical processes. They also may be intentionally generated for use in applications such as advanced ceramic materials, superconducting powders, and ultraviolet (UV) screening films. Additionally, nanoparticle aerosols have been implicated as potential contributors to observed excess mortality related to air pollution and to untoward pulmonary effects in laboratory animals. The particles may be multicomponent, with individual chemical/elemental species distributed with respect to particle size and within individual particles. The goal of this research is to examine systematically the

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chemical and transport characteristics of agglomerates of nanometer aerosol particles, including

- the effect of adding a second chemical species on the size distribution of agglomerates generated from solid-phase aerosol precursors,
- the distribution of individual chemical species with particle size for binary aerosols, and
- the arrangements of individual chemical species in the aerosol agglomerates (particle-to-particle variation).

Particles form in gases when chemical reactions produce condensable molecules. The condensable molecules then form critical (stable) nuclei that may be solid or liquid, depending on the thermodynamic properties of the substance and the system's time-temperature history. The stable nuclei may continue to grow by vapor condensation. When particle concentrations are very high and the gas is depleted of condensable vapor, particle growth is controlled by collisions among stable nuclei and subsequent coalescence (sintering).

We have a fairly good understanding of aerosol collision and coagulation processes for spherical particles (Fuchs, 1964; Friedlander, 1977). However, it is much more difficult to predict rates of coalescence of nanometer particles. Koch and Friedlander (1991) considered the case of collisions among nanometer particles that collide and coalesce in a cooling gas. As the gas temperature decreases, the rate of coalescence decreases. After a certain cooling time, which depends on the material properties of the particles, the average particle diameter approaches a limiting value, changing little over time scales of practical interest. These primary particles continue to collide, forming fractal-like agglomerates.

Ultrafine or nanometer-sized particle (nanoparticle) air pollution has been implicated as a possible contributor to increased morbidity and mortality from acute and chronic pulmonary illness (Dockery et al., 1993; Seaton et al., 1995; Bates, 1996; Sunyer et al., 1996). Studies on laboratory animals indicate exposures to nanoparticles (Warheit et al., 1990; Oberdorster et al., 1992; Ferin et al., 1992; and Oberdorster et al., 1995) and their agglomerates (Oberdorster et al., 1996) can produce pulmonary inflammation, breakdown the pulmonary vascular epithelia barrier, and cause pulmonary fibrotic reactions. The inhalation hazard potential of nanoparticle aerosols results from obstacles nanoparticles present to clearance from pulmonary regions of the lung. This potential is related to the physicochemical character of the nanoparticle materials.

These findings suggest that health and environmental protection will be an important factor in the profitability of developing nanoparticle technologies. Protection will depend on the development of materials that balance process requirements with the hazard potential. Cost savings may be realized by using materials that meet minimum process requirements and have lower hazard potential than process-optimal materials. Nanoparticle formation kinetics will be crucial in the nano-engineering of candidate materials. A clear understanding will be needed of the effects nanoparticle formation conditions have on the product specifications important to the process requirements and to hazard potential.

Importance to LANL's Science and Technology Base and National R&D Needs

A number of ongoing activities at Los Alamos may produce multicomponent aerosols that consist of agglomerates of nanometer particles. These activities include explosive testing, laser machining, and hazardous waste incineration. Control of fugitive nanoparticle aerosol emissions will require an understanding of nanoparticle aerosol transport. This transport is important in predicting collection efficiencies of available process gas- and air-cleaning systems, optimization of system performance, and in the development of control strategies.

Nanometer aerosol agglomerates are also important in the development of domestic commercial technology in the areas of catalysis, advanced nanoparticle materials, particle control technology, and hazardous waste incineration. Understanding of formation kinetics and transport properties of nanoparticle aerosols is necessary for determining their hazard potential. Formation kinetics determines physical and physicochemical properties of the aerosol that have been related to pulmonary effects of inhalation exposure observed in laboratory animals. Inhalability and pulmonary deposition of the nanoparticle aerosols depend on their transport properties. The hazard potential of the nanoparticle aerosols is important in establishing safe workplace exposure levels and setting environmental emission limits.

To facilitate cooperation between the University of California-Los Angeles (UCLA) and Los Alamos National Laboratory (LANL), we organized a workshop on nanoparticle formation. This workshop took place March 30, 1995, and included talks by the authors of this report.

Scientific Approach and Accomplishments

We distinguish among three types of particles in order of increasing size:

- critical (stable) nuclei, which may range in size from molecules to molecular clusters up to a nanometer;
- primary particles of the order of one to a few hundred nanometers, which result from collision and coalescence of stable nuclei; and
- agglomerates of primary particles, which may range from a few nanometers up to a micron or larger.

Within the agglomerates, the various chemical components can be arranged in different ways, such as

- single-component primary particles, which form independently then agglomerate later,
- multicomponent primary particles with the chemical components uniformly distributed through each particle or
- single-species particles with the existence of a second species as a coating.

Combinations of these morphological arrangements within the same aerosol are also possible.

To study primary particle and agglomerate formation, the UCLA Aerosol Technology Laboratory has developed several different particle generators, two of which are used in this project. The first, the honeycomb flat-flame aerosol generator, provides an approximately one-dimensional configuration in which primary particles are formed then agglomerate as the aerosols pass up the chimney. The premixed methane/oxygen/nitrogen flame is stabilized on a stainless-steel honeycomb grid, allowing the passage of solid-phase aerosol precursors. In later work with gas-phase precursors, a second premixed methane/oxygen/nitrogen flat-flame generator was assembled, consisting of a porous plug of sintered bronze pellets.

Characterizing Binary Aerosol Formed by Passing Micron-Sized Particles Through a Flat-Flame

Binary aerosol mixtures of zinc oxide with magnesium oxide, as well as copper oxide with magnesium oxide, were generated from aqueous solutions of metal salts (zinc nitrate/magnesium acetate and copper nitrate/magnesium nitrate); these were atomized,

dried, and passed through the flat-flame burner. The aerosol agglomerates were classified with respect to aerodynamic diameter using a Hering low-pressure impactor. The bulk metallic compositions in each particle-size range were determined using inductively coupled plasma atomic emission spectrometry (ICP-AES). The resulting mass distributions are shown for both systems in Fig. 1. The distributions were bimodal for the Zn and Mg in the ZnO/MgO system. In the CuO/MgO system, the Cu was concentrated in the smallest size ranges ($d_p < 0.5$ micron) and the Mg distribution was bimodal. Experiments were also conducted to determine the mass distributions of single-component aerosol, which in turn enabled us to determine the effect of adding a second species on the distribution of the first. The single-component mass distributions can be found in Ehrman et al. (1996).

Comparison with the mass distribution of the atomized precursor particles indicated that the large mode consists of the material that reacted in the solid state to form the non-volatile oxide. Transmission electron microscope (TEM) images showed the large mode to consist of large ($d_p > 0.5$ micron) particles. TEM images of the small mode revealed agglomerates of nanoparticles ($d_p = 5$ to 20 nm) that nucleated homogeneously from the vapor phase. This mode accounted for approximately one-third of the mass of the final aerosol but nearly all of the estimated surface area.

The structure and chemical speciation on a nanometer scale were studied using JEM 200 CX analytical TEM. Particles were collected for TEM using the critical orifice stage of the Hering low-pressure impactor. Distinct agglomerate structures of varying size were analyzed using energy dispersive x-ray spectrometry (EDS). Analysis of a single primary CuO/MgO particle, $d_p = 20$ nm, revealed that the single particle contains both chemical species and in the same ratio as the larger agglomerate structures. This feature suggests that the two species are nucleated together from the vapor phase.

In a high-temperature process, the metal salt particles may undergo vaporization, breakup, chemical reaction, or some combination of the above (Fig. 2). The bimodal distributions indicate that more than one route to aerosol formation may be occurring at the same time. The partitioning of the individual species amongst the pathways is a result of competition between the processes.

The vapor pressures of the oxide species at the maximum measured flame temperature of 1350 K are low for CuO and ZnO (1×10^{-3} and 2×10^{-2} Torr, respectively). For MgO, the vapor pressure at 1350 K is very low: 6×10^{-9} Torr (Samsonov, 1982). For CuO and ZnO, it is possible that some vaporization and recondensation of the oxide may occur, but it is not likely the main mechanism for formation of the fine aerosol. MgO forms a fine aerosol, and this can only be explained by a process other than vaporization and recondensation of the oxide. The behavior of the metal salts before reaction to the

oxide must be considered. Only qualitative information about the vaporization behavior of the metal salt species was found in the literature. The nature of the bond between the metal cation and the anion appears to affect the decomposition behavior (Stern, 1972). In the case of anhydrous copper and zinc nitrate, the bond between the metal cation and the nitrate anion is covalent. In a high-temperature process, the covalently bonded nitrates are stable enough to melt and possibly vaporize before decomposition to the oxide. Anhydrous magnesium nitrates are ionically bonded. Ionically bonded nitrate salts are not stable enough to vaporize and usually decompose to the oxide in the solid phase (Stern, 1972).

Based upon the limited information about the behavior of nitrate salts in a high-temperature process, the morphological information in the TEM images, and the mass distribution of the product oxide aerosol, mechanisms for the formation of the particles can be suggested. Magnesium acetate alone appears to follow the vaporization path. Zinc nitrate is bimodal in both cases (Ehrman et al., 1996). During heating, some zinc nitrate may be reacting to zinc oxide in the solid phase, trapping some magnesium acetate in the particle and resulting in the bimodal distribution for magnesium acetate in combination with zinc nitrate. Copper nitrate alone also appears to follow the vaporization path (Ehrman et al., 1996). In combination with magnesium nitrate, the distribution is still unimodal. It may be possible that magnesium nitrate reacted to the oxide later in the process, when compared to zinc nitrate, thereby allowing more of the copper nitrate to diffuse to the surface of the precursor particles and evaporate.

It is apparent that multicomponent aqueous solutions of metal salts exhibit highly complex behavior as aerosol precursors. The combination of impactor and ICP-AES analysis to determine the bulk chemical composition as a function of size in a mixed aerosol with information from TEM images was useful for determining which processes may be occurring during aerosol formation. However, because of the paucity of information about the behavior of the metal salt compounds in high-temperature processes in the literature, it is difficult to predict which routes to aerosol formation will occur. In the next phase of the research, we focused on the gas-to-particle route, and on using analytical TEM techniques to investigate the arrangement of the chemical species in the product aerosol on a-nanometer scale.

Formation of $\text{SiO}_2/\text{TiO}_2$ Nanocomposite Aerosol in a Premixed Methane Flat Flame

We modified the experimental system to introduce gas-phase precursors into the flame. The burner used for these experiments consisted of a water-cooled porous plug of sintered bronze pellets. This burner produced a very uniform flat flame, $T_{\text{max}} = 1720 \text{ K}$,

sitting approximately 1.5 mm above the burner. Silica/titania ($\text{SiO}_2/\text{TiO}_2$) mixed oxide aerosol was chosen as the system for study because of (1) the many applications of mixed $\text{SiO}_2/\text{TiO}_2$ materials (such as optical fiber cladding, coated paint pigments, and catalysts), (2) the availability of suitable liquid precursors for silica and titania, and (3) the interesting material properties of the $\text{SiO}_2/\text{TiO}_2$ system. Silica is a strong network former, and is amorphous as formed in flame reactors (Ulrich, 1971). Flame-processed titania, in contrast, is crystalline (Pratsinis et al., 1996). In the solid phase, there is no thermodynamically stable mixed phase, although experimentally the solubility of titania in silica glasses has been observed up to 12% by weight of titania (Schultz, 1976). In the liquid phase, for compositions ranging from approximately 20 to 90% by weight titania, the liquids are immiscible (DeVries et al., 1954). Because many different precursors for silica were available, we were able to study the effect of precursor reaction kinetics on the formation of mixed aerosol in the premixed flame. We used the same precursor for TiO_2 , TiCl_4 , but varied the silica precursor using SiBr_4 , SiCl_4 , and hexamethyldisiloxane (HMDS).

Single-component silica and titania aerosol were produced. All the silica samples were found by electron diffraction to be amorphous. By measuring the diameter of 50 particles in a sample from the TEM image and averaging, we determined the primary particle size for the silica samples was 10 nm for silica produced from SiBr_4 , 11 nm for silica from SiCl_4 , and 10 nm for silica from HMDS. The similarity in particle size amongst the silica samples suggests that there are no significant differences in the chemical reaction behavior of the precursors in the flame. The pure titania aerosol was found to be crystalline, anatase by electron diffraction, and the particles were faceted in the TEM images. The average particle size was slightly larger for titania ($d_p = 13$ nm) than for silica formed under the same conditions. The difference in particle size can be attributed to the rapid solid-state sintering of titania particles, as well as the relatively slow sintering of silica particles by a viscous flow mechanism (Xiong et al., 1993).

Mixed $\text{SiO}_2/\text{TiO}_2$ aerosol (1:1 mole ratio Si to Ti) were generated from three sets of precursors: $\text{SiBr}_4/\text{TiCl}_4$, $\text{SiCl}_4/\text{TiCl}_4$, and HMDS/ TiCl_4 . The mixed aerosol consisted of spherical or near-spherical particles. The average primary particle size was approximately 15 nm for $\text{SiBr}_4/\text{TiCl}_4$, 15 nm for $\text{SiCl}_4/\text{TiCl}_4$, and 16 nm for HMDS/ TiCl_4 . Again, the lack of a significant difference in particle size suggests that the chemical reactions are occurring at the same point in the flame. Within the primary particles, regions that diffract strongly, an indication of crystallinity, were visible as dark areas in the TEM images. The crystal structure of the mixed aerosol for all combinations of precursors was determined by

electron diffraction to be anatase.

To determine if Ti and Si were both present in the individual particles, energy dispersive X-ray analysis was conducted on single particles using a Phillips EM400 analytical transmission electron microscope. We followed a protocol in which 24 analysis were conducted per sample. The mole fraction of Ti on a metals basis was determined for each analysis. The results are shown as a scatter plot in Fig. 3. Because of the small amount of mass analyzed, the uncertainty associated with this technique is fairly large (approximately 20% relative). However, from the data in Fig. 3, it is reasonable to conclude that the particles contain both species in approximately the same ratio as the starting materials. There were no differences among the samples indicating that the choice of precursor had no effect on particle homogeneity.

To investigate the composition of the crystalline domains, another analytical electron microscopy technique was used: energy electron loss spectrometry (EELS). The microscope was operated under conditions corresponding to a 1- to 3-nm probe diameter. $\text{SiO}_2/\text{TiO}_2$ aerosol from $\text{SiCl}_4/\text{TiCl}_4$ was analyzed. Because we have only a two-dimensional view of the particle subject to analysis, we cannot be certain the probe was gathering information from entirely within a crystalline domain. However, significant differences in composition between the crystalline domains and the rest of the particle were observed. The crystalline regions were strongly enriched in Ti (Ti/Si ratio from 3 to 18); the noncrystalline regions were slightly enriched in Si (Si/Ti range 1.4 to 3).

The processes occurring during multicomponent aerosol formation from gas-phase precursors are the same as for single-component aerosol formation: chemical reaction, nucleation, and aerosol growth. The materials in this study, silica and titania, are refractory materials with low vapor pressures at the temperatures encountered in the flame. Calculations of the critical nucleus size for silica and titania suggest that particle formation is not governed by an activated nucleation process because every molecule can serve as a stable nucleus; particle formation should immediately follow formation of the condensable species by chemical reaction. If there were significant differences in the chemical reaction behavior or the precursors in the flame, chemical segregation (possibly chemically distinct primary particles) may result.

In the reaction zone of the premixed flame, concentrations of radical species such as O, OH, and H are relatively high (Glassman, 1977). Therefore, it is expected that at the high temperatures of the flame, methane oxidation chemistry will be dominated by low-activation-energy radical abstraction reactions. Similar behavior is anticipated for the precursor compounds, with the net result being very fast decomposition of the precursors and little difference from one precursor to the next. The results of the EDS analysis shown

in Fig. 3 support this hypothesis, as well as the uniformity of the particles as observed in the TEM images. We believe that the segregation within the particles is a result of the immiscibility of SiO_2 and TiO_2 in the solid phase. A mechanism for the formation of binary aerosols in a premixed flame is shown in Fig. 4. The chemical reactions to form the condensable species are occurring simultaneously. Because of the low vapor pressure of the oxide species, the oxides condense together to form the aerosol. The individual chemical species can move within the particles by solid-state diffusion.

We suggest that as the residence time increases, the segregated crystalline areas within the particle form and grow by a solid-state diffusion mechanism. This behavior is expected, considering the equilibrium phase distribution (separate silica and titania phases) for the two species at the maximum flame temperature of the system. The driving force for the rearrangement of species by diffusion is the reduction in the free energy of the system by the formation of the thermodynamically favored phases. It is interesting that this phenomenon is occurring on the nanometer scales of the materials formed in these experiments.

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a) Zn and Mg, Mass Distribution
First Trial

b) Cu and Mg, Mass Distribution
First Trial

Fig. 1. Mass distributions as a function of aerodynamic diameter. (a) Mass distributions of Zn and Mg are both bimodal in the ZnO/MgO system. Ratio of Zn to Mg is approximately the same in each size range. (b) Mass distribution of Mg is bimodal for CuO/MgO system, but Cu appears to be concentrated in the $d_p < 0.5$ micron range.

Fig. 2. Possible decomposition paths of metal salt particles in a high-temperature process.

Fig. 3. Results of single-particle, energy dispersive x-ray spectroscopy analysis. Relative uncertainty associated with the analysis is approximately 20%.

Fig. 4. Suggested mechanism for formation of binary aerosol in a premixed flame.