REPORT OF PROGRESS

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PROJECT TITLE: Atmospheric Aerosol Microphysics: Formation, Characterization,
and Interaction

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Abstract

This project conducts theoretical and computational studies of the physical transformation processes of aerosols which underlie their atmospheric formation, interaction, transport, and removal and derives results that contribute to improved capabilities for modelling aerosol physical and chemical evolution in support of the environmental component of the National Energy Strategy. The subject of study is submicrometer aerosol particles with primary focus upon the ultrafine fraction. This report summarizes technical progress during the first two and one-half years of the project.

Results of calculations of equilibrium vapor pressures over adhering pairs of 50, 100, and 200 nm particles are reported showing substantial depression of equilibrium vapor pressure relative to isolated spheres. Calculations are given of collective, long-range intermolecular energies for irregular particles to be used for growth rate calculations for realistic particles. Molecular dynamic simulations of thermal collisions of small clusters with each other and with single atoms are presented as a function of cluster size in the range from 1 to 8 atoms. Calculations of aerosol condensation in which vapor depletion and heating effects are taken into account for atmospheric cloud nucleation modelling are reported.

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PART I. PROJECT PERFORMANCE REPORT FOR

1 SEPTEMBER 1991 – 28 FEBRUARY 1994

A. Research Objectives of Original Propopsal

Aerosols are of broad importance in atmospheric chemistry and in the environmental impacts of the National Energy Strategy. They originate both as local, primary emissions and as secondary emissions formed within the atmosphere from gases arising from both natural and energy-related sources. To evaluate and anticipate the environmental roles and impacts of atmospheric aerosols, descriptions of the physical processes underlying aerosol formation, evolution, and interactions are required. In support of these information needs, this project conducts theoretical and computational studies of the physics of aerosol particle formation, evolution, and interaction.

The research developments originally encompassed by this project are divided into complementary tasks reflecting the division of its efforts. In Task I, calculational procedures are being developed for collision rates of ultrafine particles for application to the description of irregular particle (also called *fractal aggregates*) formation from noncoalescing primary particles. This work will facilitate subsequent studies of the condensational properties of irregular ultrafine particles as well as future investigations of their morphological stability under atmospheric conditions. In Task II, the equilibrium vapor pressures over adhering pairs of 0.1 – 0.4 μm particles have been calculated. In both tasks, the overall objective is to investigate the role of realistic aerosol shape as a factor in atmospheric condensational processes both at subsaturation and supersaturation conditions.

B. Associated Research Activities

Task I of the current project, as originally written, was intended solely to lay the foundation for subsequent research on the condensational properties of aggregates of primary particles in the 1 – 5 nm range of radii. Important aspects of this subsequent research are now in progress under support from NASA which began 1 September 1992. During the first year of the current DOE project, the Principle Investigator won a NASA Innovative Research Program research grant entitled Atmospheric Condensational Properties of Ultrafine Chain and Fractal Aerosol Particles whose objectives were to carry forward the research initiated in the current DOE project. Thus, progress is being made toward the long-range objectives of this project as outlined in the original proposal; because of the close, complementary nature of the two projects, references will be made to work conducted under NASA sponsorship in this progress report though results from the NASA project will not be reported here.

The flexibility afforded the research program at Texas A&M by the NASA project has made possible the assumption of support for additional research related to the objectives of this project but which did not appear in the original proposal. These research projects were initiated under other, prior support and are drawing to completion under DOE sponsorship. Specifically, these two projects are (1) molecular dynamics studies of small cluster collisions to examine the onset of "aerosol" behavior in thermal collisions and (2) computational studies of aerosol condensation kinetics including effects of water vapor depletion in coupled aerosol systems of importance in cloud condensation.

C. Results and Progress to Date

C.1. Equilibrium vapor pressure over adhering, macroscopic spheres.

The equilibrium vapor pressure over a spherical particle is generally taken to be determined by the balance between surface energies and vapor condensation factors. Over a small spherical surface, the well-known elevation of the equilibrium vapor pressure indicates that condensation at a given vapor pressure cannot occur unless the wetting angle is sufficiently small on that surface, i.e. unless the particle is sufficiently hydrophilic. However, there is another possibility for even hydrophobic particles that has not received attention in the aerosol literature and that pertains to many realistic aerosols such as from combustion and elsewhere. In this case, adhering spheres are in question and capillary condensation can be expected to result in formation of a meniscal ring of water in the interstitial region between the spheres. This meniscus is concave for water-particle wetting angles under ninety degrees and therefore can result in a depression of the vapor pressure according to the Kelvin equation. The theory for the vapor pressure of water over adhering spheres has been developed and calculations performed under this project. This work is described in the attached Master' Degree thesis entitled *Equilibrium Vapor Pressure and Capillary Ring Formation of Adhering Spherical Aerosol Particles* by Yoan Crouzet. Relevant parts of this work have been written for publication in the accompanying manuscript, "Calculations of the equilibrium vapor pressure of water over adhering 50-200 nm spheres" by Y. Crouzet and W. H. Marlow which has recently been submitted to *Aerosol Science and Technology*. *Preprint - Removed for separate processing*

The approaches and results of the studies of water condensation on adhering spheres are currently being generalized to include condensation on adhering spheres with sulfuric acid deposits as a model for realistic atmospheric aerosols. Results have

been developed for a range of electrolyte-surface contact angles and several fixed masses of sulfuric acid distributed either uniformly over the surfaces of the two particles or initially concentrated in the meniscal rings encircling the contact points of the two spheres. These configurations are adopted to model possible surface distributions of sulfuric acid and resultant wetting behavior for realistic aerosols. At present, these studies are drawing to a close under the support of the NASA program discussed above and will be written for submission to the literature. The other component of this NASA project which builds upon work initiated under the current DOE project is studies of condensational thermodynamics of aggregates of spherical particles in the 1 – 5 nm range of radii taking into account the fractal dimensions of the aggregate particles. This work is in progress but as yet has no results.

C.2. Ultrafine particle aggregation

Ultrafine particles in the range of radii below 5 nm first enter or are produced in the atmosphere as the result of a number of primary and secondary processes. Generally speaking, if the compositions and sizes of these particles preclude their rapid condensational growth, they will grow by coagulation with other comparable particles or they will be captured by (or equivalently, coagulate with) larger particles that are also present. Since the collision frequency for an irregular particle is significantly larger than for a sphere of the same mass, the atmospheric longevities of ultrafine particles as independent particles are, to a large degree, determined by their coagulation with other ultrafine particles and the shape or form that the coagulated particles assume: if the coagulating particles coalesce to form spheres, then their rate for capturing additional ultrafine particles will be lower than if the particles do not coalesce but form chains and more complicated aggregate structures. Also, for an irregular particle, transport behavior including thermal diffusivities, aerodynamic properties,

and possibly condensational kinetics resemble spherical particles of much larger masses. Thus, in accounting for the fate of ultrafine particles, the first step is to account for the partitioning between collisional growth among ultrafine particles and capture by larger particles and the implications this has for subsequent atmospheric chemical and physical interactions including removal processes such as rainout. A second, equally important need for accounting for the evolution of ultrafine particles is their interaction properties with the atmospheric gases including water vapor. If these particles are captured by 100 nm or larger aerosols such as haze particles, then they likely will play little if any direct role in cloud formation. Similarly, if the particles coalesce to form ultrafine spheres, then they will not likely participate in cloud formation either. However, if they form chains and more complex aggregates, then their condensational properties will differ entirely, being dominated by capillary condensation (discussed above for macroscopic spheres) and they may become either haze particles or cloud condensation nuclei.

Prior DOE/OHER support for Aerosol Microphysics projects at Brookhaven National Laboratory and at Texas A&M University resulted in the development of the theory of collision rates for ultrafine, spherical aerosol particles. This theory and computational results include the roles of particle size and chemical composition via the effects of the long-range intermolecular potential between particles. While the interaction potentials were originally based upon the Lifshitz theory of the van der Waals force for continuous, macroscopic matter, the work was subsequently extended to the collisions of highly symmetric, discrete molecular clusters by the utilization of the discrete, iterated dipole interaction energy that had previously not been applied in research on mesoscopic physics questions such as are posed by ultrafine aerosols.

In the current project reported here, the theory and computational methods originated under the previous projects have been refined and extended for computa-

tions of ultrafine particle aggregation. To describe the elements required for the calculation, the following diagram is useful with

r_p = primary particle radius

s_{pa} = center of mass separation of particle and aggregate

Let E be the long-range intermolecular interaction potential between particle and aggregate as a function of orientation and separation. Calculations of the rates and locations of attachment of particles to aggregates require complete accounting for E from almost contact to sufficiently large separations that the energy becomes negligible. Before E can be calculated with confidence, three open questions must be answered: (1) the ranges of usefulness of the Lifshitz theory of the van der Waals interaction must be evaluated for spherical particles approaching molecular cluster dimensions; (2) approximations of the retardation of the long-range interaction energies between spheres must be evaluated; and (3) the importance for E of many-body interactions between spheres in the aggregate must be evaluated.

C.2.1. computation of interaction energies for quasispherical clusters and particles

Calculations of the long-range intermolecular force for $r_p > 1$ nm spheres are usually based upon a continuum description of matter which accounts for many-body interactions in the condensed state in terms of the frequency-dependent dielectric

constants of the interacting materials. The attractive energy calculated on this basis is the Lifshitz-van der Waals energy and labelled E^L in the following. However, below some size of particle, the description of the interaction energy in terms of uniform, dielectric spheres becomes questionable. Earlier Aerosol Microphysics projects have calculated the interaction energies between icosahedral and dodecahedral clusters of carbon tetrachloride molecules as a simple model of organic clusters. Those calculations were based upon an approach originally developed by Langbein solely to demonstrate that the Lifshitz energy could be developed as a particular limiting case for the interaction energy of a system of discrete, coupled dipoles. In Langbein's picture, each molecule of the system for which the interaction energy is being calculated is represented by its frequency-dependent dipole polarizability. The molecules interact via the dipole-induced dipole interaction with all other molecules to all orders of coupling among the molecules in a complex, many-body calculation. Since this approach treats each molecule according to its location and properties, it is termed the "discrete" interaction energy and is labelled E^D in the following discussion.

In the current project, interaction energies have been computed according to Lifshitz theory for small particles of the same mass, and corresponding radii and dielectric properties, as the icosahedral and dodecahedral clusters cited above. These energies have been compared with the energies computed upon the basis of discrete coupling of the all molecules in the two particles, E^D . In addition, the Lifshitz-Hamaker approximation, E^{LH} , to the energy E^L , has been computed for the sake of evaluating this approximation which is widely used in colloid science. Figures 1 – 3 are graphs of the relative difference of E^L or E^{LH} and E^D vs scaled separation: The solid lines correspond to $\frac{E^L - E^D}{E^D}$ vs. d_{cm}/d_{av} and the dashed lined correspond to $\frac{E^{LH} - E^D}{E^D}$ vs. d_{cm}/d_{av} where d_{cm} is center of mass separation of the particles and d_{av} is an average diameter for the discrete cluster:

Figure 1. Relative error graphs of interaction energies for pair for 13-molecule CCl_4 clusters and particles

Figure 2. Relative error graphs of interaction energies for pair for 33-molecule CCl_4 clusters and particles

Figure 3. Relative error graphs of interaction energies for pair of 55-molecule CCl_4 clusters and particles

These graphs indicate that for large surface-to-surface separations of the particles, E^L converges to E^D within the accuracy of the approximation used to compute E^L while E^{LH} underestimates the exact energy, E^D , by 10 – 15%. For small separations, both approximations overestimate the attractive interaction energy as is well known for the macroscopic interaction. As has been pointed out in earlier Aerosol Microphysics projects, this inaccuracy is not so important for collision rate calculations due to the depth of the potential well at separations where the continuum models differ greatly from the discrete energy. Improved representation of the interaction energies for ultra-fine particles at short range is currently under development under the NASA program.

C.2.2. retardation of the long-range intermolecular potential between ultrafine particles

Retardation of the long-range intermolecular potential between two particles can be understood by referring to the calculation of Casimir and Polder of the interaction energy of two molecules. They showed that as molecular separation, s , increases, the familiar s^{-6} dependence of the London-van der Waals energy transforms into an s^{-7} dependence for large distances. For molecular interactions, this accelerated decrease of the energy with distance is of no practical consequence because at values of s for which this retardation effect is significant, the magnitude of the interaction energy is already too weak to affect physical processes. As demonstrated in 1980 under an earlier Aerosol Microphysics project, this is not true for aerosol collision rates. Retardation has a significant effect on these rates for particles larger than several nanometers (specific size depending upon composition) and indeed retardation alone has been used to explain discrepancies between data and its interpretation which neglected retardation of the van der Waals interaction. Considering the ranges of particle and aggregate dimensions that are involved in treating ultrafine particle growth by aggregation, the effect of retardation will clearly be a factor.

In the 1980 treatment of retardation of the intermolecular force between aerosol particles, a "point-dipole" approximation was introduced for computational convenience but with only qualitative justification. This approximation treated the retardation effect as if the magnitude of the retardation between two spheres were the same as for two point dipoles located at the centers of the interacting spheres. Under the current project, this question has been reexamined due to its clear importance in aggregation. The approach adopted here is based upon the premise that the most important effects of retardation should be shown in the molecule-to-molecule retardation between interacting pairs of molecules in the two interacting particles. Thus, the geometrical distribution of mass across the extension of the two spheres will be treated explicitly, unlike

the point-dipole approximation. In the calculations performed here, a continuum picture of the interacting particles is adopted and the retardation effect is calculated by integrating dipole densities over the two spheres in a manner that is similar to the sum-over-pair interaction approach to computing interaction energies. In Figures 4 – 6, the ratio of the retarded to nonretarded energies for CCl_4 spheres are computed in the point-dipole and the integrated dipole density approximations.

Figure 4. Ratio of retarded to nonretarded interaction energies for two 5 nm radii spheres. Dotted line for point-dipole retardation and dashed line for integrated dipole density retardation.

Figure 5. Ratio of retarded to nonretarded interaction energies for two 50 nm radii spheres. Dotted line for point-dipole retardation and dashed line for integrated dipole density retardation.

Figure 6. Ratio of retarded to nonretarded interaction energies for 5 and 50 nm radii spheres. Dotted line for point-dipole retardation and dashed line for integrated dipole density retardation.

These retardation calculations indicate that the point-dipole approximation is sufficiently accurate to permit its continued use in collision calculations

C.2.3. calculation of the collective, long-range intermolecular forces between complex particles

At the basis of calculations of E^L and of E^D and their close correspondence with each other is the fact that all molecules comprising the particles are collectively coupled to each other. For small clusters, the "iterated-dipole" interaction energies of the clusters, i.e. E^D , can be calculated exactly, as alluded to above. However, for particles of the dimensions of interest here for understanding ultrafine aggregation phenomena, this is not possible. Conversely, E^L provides an adequate accounting of the energy between spheres, but in general the Lifshitz theory of the van der Waals interaction has only been solved for a limited number of idealized geometries and therefore cannot *a priori* be expected to portray accurately the interaction energies between a particle and an aggregate of particles. However, the question can be asked of how important the many-body interactions are if the coupling between constituent elements of the system is carried out to sufficiently high order? Specifically, if E , between a particle and an aggregate could be expressed as a simple sum over E^L values between each constituent particle of the aggregate and the single particle, then a computationally tractable long-range energy would be available for aggregation calculations as well as for accurately portraying the interactions of fluids on surfaces as indicated in the final sections of the attached Master's Degree thesis entitled *Equilibrium Vapor Pressure and Capillary Ring Formation of Adhering Spherical Aerosol Particles* by Yoan Crouzet.

To determine if the above interaction picture is useful, the exact, iterated-dipole energies among three 13-molecule CCl_4 clusters were computed. The clusters were arranged so that two are in contact and the third is some distance separated from

them. For various separation distances, the exact, completely-coupled energies were calculated. Next, only the energy resulting from coupling the single cluster separately to each of the contacting clusters was calculated. In this manner, the effect of the coupling between the contacting clusters on E was computed. These calculations were performed for two orientations of the contacting clusters relative to the isolated cluster: in the first, the centers of the three clusters lay on a line; in the second, the contacting pair of clusters were rotated by ninety degrees about their contact point relative to the first orientation so that the three clusters were in a "T" configuration. The results of those calculations, along with the energy between the sphere and paired spheres calculated according to Lifshitz theory, are given in Figures 7 and 8.

Figure 7. Interaction energy as function of center of mass separation between contacting pair of CCl_4 clusters and a third cluster in the line orientation. Dotted line corresponds to simultaneous coupling among all 39 molecules; dashed line corresponds to coupling separately of isolated cluster to each contacting cluster; solid line corresponds to sum of corresponding Lifshitz energies between single particle and each of the paired particles.

Figure 8. Interaction energy as function of center of mass separation between contacting pair of CCl_4 clusters and a third cluster in T orientation. Dotted line corresponds to simultaneous coupling among all 39 molecules; dashed line corresponds to coupling separately of isolated cluster to each contacting cluster; solid line corresponds to sum of corresponding Lifshitz energies between single particle and each of the paired particles.

In both Figures 7 and 8, the discrete energies due to completely coupling and partially coupling the clusters agree quite closely indicating that little error will be made by calculating E as a sum over individual E^L values as suggested above.

C.2.4. aggregation kinetics and morphology

Trajectory calculations for the collisions of single ultrafine particles with aggregates of particles currently are in progress utilizing the results for interaction potentials discussed above. In these calculations, the operational assumption is that the single particles become permanently attached to the aggregate at the point of first contact. In addition, the transport calculations for collisions must also account for transition

regime effects as the greatest dimension of the aggregate becomes comparable with the mean free path of the gas. This will be done by extending the Fuchs "dividing sphere" approach as an envelop of the appropriate dimensions about the aggregate.

C.3. Molecular dynamics simulation of small cluster growth.

Homogeneous nucleation is the formation of new, condensed-phase material within the gas phase. While the classical theory of nucleation has proved to be useful in providing a format for the qualitative understanding of nucleation phenomena, improvements to the theory have been largely qualitative and empirical without addressing underlying assumptions, approximations, or extensions to enhance its usefulness. At the basis of the questions surrounding the subject is the incompleteness of understanding of cluster formation, growth, and transformation processes in general, regardless of whether or not critical nucleation is involved. Since clusters are broadly present in the atmosphere and undoubtedly play many important roles in atmospheric chemistry, a broad-scale need exists for the development of methods for treating gas-phase cluster processes, in general. As the overall field establishes reliable and quantitative techniques, there is the prospect that these techniques may be helpful in addressing questions which currently are not amenable to treatment utilizing what is known including questions involving the treatment of nucleation .

One of the fundamental questions in cluster kinetics is collisional growth processes. For low vapor pressure species for which the "critical nucleus" is very small (e.g. under 10 atoms), and arguably is the monomer, collisions among small clusters will certainly occur. The question may be posed of whether such clusters upon collision will form stable new clusters as in aerosol collisions or if they will fragment or undergo evaporation. In addition, the question of how the paths for cluster growth are partitioned between monomer accretion (i.e. condensation) and coagulation with simi-

lar-sized clusters is open. To address these issues, an understanding must be developed of what can constitute an adequate representation for the interatomic potentials of the atoms involved. These questions are being addressed for the collisions of nickel clusters under this project; the expectation is that the development of a well-grounded picture for simple atomic clusters will guide the subsequent development of a picture for molecular clusters. Two quite distinct atomic potentials are being used to determine the sensitivity of the results to the form of the potential. One potential is a conventional, two-body Lennard-Jones type while the other is a form involving all nearest neighbors and is therefore is a many-body potential though less complex than used in the calculations described above for aggregation. Throughout these simulations, "importance sampling" of the molecular dynamic trajectories has been used resulting in error bars in the range of 10% of the values displayed. Selected results of the calculations are given in Figures 9 and 10.

Figure 9. Sticking efficiency vs. size in the collisions of identical Ni clusters with many-body potential.

Cluster-monomer collisions for Ni at $T = 1200$ K

Figure 10. Sticking efficiency vs. size in the collisions of Ni clusters with Ni atoms with many-body potential.

Figure 9 indicates that in the collisions of identical clusters, those smaller than four atoms become attached in less than half of the collision events while larger clusters' sticking efficiencies increase with increasing cluster size until the range of larger than 7 atoms where efficiencies become essentially equivalent to aerosol collisions. Other sets of similar simulations show that the Lennard-Jones clusters throughout the range of sizes predict lower sticking efficiencies which means that choice of potential is important throughout this range for coagulational growth of clusters.

Figure 10 displays another type of result which requires further examination. Here, monomer sticking is studied and shows efficiencies similar to the efficiencies in the previous graph. However, what is particularly interesting here is that sticking effi-

ciency grows to unity throughout this range despite the fact that all clusters are thermodynamically stable, i. e. the clusters are by any accounting post-critical clusters. This behavior violates an underlying assumption of classical nucleation theory, though in practice it is not relevant because of the assumption of large numbers of monomers in the critical cluster. This behaviour will be examined from a dynamical perspective to elucidate the energy accommodation processes which are presumably responsible. In addition to the sticking efficiency question, other results of these simulations have shown that when Lennard-Jones potentials were used, the sticking efficiencies at the lower end of the range of masses displayed in Figure 10 disagreed with the efficiencies plotted in the figure while they converged to similar values to those shown for the larger clusters. Thus, these studies seem to show that for condensation processes, the usefulness of a particular potential is dependent upon the size of the cluster in question.

C.4. Computations of condensational kinetics of ultrafine aerosols

Optical and radiative properties of clouds depend on their droplet size spectra and microphysical properties which, in turn, are determined by the size distribution and composition of aerosol particles that serve as cloud condensation nuclei (CCN). The number of such CCN activated to form cloud droplets controls the sizes of the droplets which influence the properties of the cloud. The activation of CCN to cloud droplets can be computed from Kohler theory in terms of vapor supersaturation.

The general theory of heterogeneous condensation of water vapor correlates size and properties of the nucleating particles with vapor supersaturation. However, most studies of cloud nucleation do not consider the significant effects of vapor depletion or temperature changes during the droplet growth processes. This explains the fact that most authors report experimental droplet growth rates smaller than theoretic-

cally predicted. In many cases, agreement between modelling and experiment is achieved by choosing incomplete mass and thermal accommodation on droplet surfaces. Most analytical and numerical studies fail to provide accurate predictions of rapid condensation of volatile compounds like water at high supersaturations.

In studies conducted under this project, a comprehensive treatment of the dynamic process of simultaneous growth of distributions of ultrafine aerosol particles has been addressed. The condensation kinetics are computed over a range of supersaturations by solving the coupled nonstationary heat and mass transfer equations simultaneously, thereby accounting for nonisothermal effects due to latent heat of condensation. The mutual interaction of the growing droplets is modelled by vapor depletion and temperature increase of the ambient background gas. Noncontinuum effects are included by computing the fluxes as a function of the Knudsen number of the particle and the model is capable of simulating rapid condensation of volatile species when the initial transient period after condensation begins is not short.

Computations are in progress of aerosol condensational including the effects of latent heat of condensation and vapor depletion on droplet activation and growth. The model will be extended to include inhomogeneity in aerosol particle distribution and composition. Such a model will be relevant in computing the activational spectrum of CCN as a function of varying supersaturation.

In current practice in cloud modelling, the concentration of CCN at a single supersaturation (the effective supersaturation of the cloud) is considered in computing the cloud droplet spectrum. Such approaches are unable to explain the spread in the observed cloud droplet spectrum which is related to both the spread in activational spectrum of the CCN and to nonuniformity in local humidity in a cloud. Means to compute an activational spectrum as a function of varying supersaturation in a cloud is vital to global climate predictions and these calculations are in progress under this project.

I.D. Projected Progress by End of Initial Project Period

The research conducted under the support of this project forms the basis for one completed Master of Science and three forthcoming Doctor of Philosophy Degrees. The latter should be complete or nearing completion as the original grant period for this project ends and within a few months following their completion, approximately seven additional papers describing the work will be submitted to the peer-reviewed literature.

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