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COMPREHENSIVE REPORT OF ACTIVITIES

Report Period: March, 1987 – August, 1989

for

AEROSOL MICROPHYSICS OF INDOOR RADON: DE-FG05-8-7ER60550

Grant Period: March, 1987 – February, 1990

Summary

The objective of Aerosol Microphysics of Indoor Radon is to develop the basis for modeling indoor radon decay products' activity-size distributions for use in dose calculations.

During the first 21/2 years of this project, the theory of electrical diffusion charging has been extended to aerosol particles under 10 nm diameter for the first time and shown to explain experimental data. Collision rates of pairs of icosahedral and dodecahedral molecular clusters corresponding to the smallest high-mobility aerosol particles carrying radon daughters have been calculated based upon exact long-range interaction energy calculations and approximate collision methods. A generalization of the collision rate formula for spherical particles interacting via long-range potentials has been derived and will be useful for practical calculations involving size distributions of particles carrying radon daughters. Separate reviews of ultrafine aerosol collision rates and radon daughter aerosol charge have been conducted and have indicated directions for work in those areas. Collaborative measurements are being made of the electrical mobility distribution of cluster ions containing ^{218}Po formed in the presence of common atmospheric trace gases. Calculations comparing background aerosol charging with radon daughter aerosol size distribution and of the attachment of high diffusivity particles to cylinders in a flow stream are also in progress.

Original and Revised Project Tasks and Research Accomplishments:

TASKS

Aerosol Microphysics of Indoor Radon was originally written as a project of theoretical and computational studies of the physical processes related to indoor radon decays. Its long-term objective was to permit modeling and prediction of the activity-sized distribution of indoor aerosols for use in dose calculations. In part, it was to support experimental and modeling research programs at other laboratories. Additionally, its purpose was to provide improved theoretical descriptions of aerosol processes for utilization in indoor radon models that would be more comprehensive than possible with existing theory. To achieve these overall objectives, the project was structured as three overlapping tasks:

Task 1 was to be concerned with computational or theoretical needs in aerosol measurement methodology and with applications of the theory of aerosol diffusion charging. Its general function was to support or to assist the interpretation of laboratory measurements conducted elsewhere. As part of this Task, a critical

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review of aerosol diffusion charging and related processes was to be performed; in addition, calculations of neutral and charged high-diffusivity (i.e. "unattached fraction") aerosol capture by cylinders in aerosol flow streams were to be made.

Task 2 was written to be supportive of kinetic modeling (to be performed elsewhere) of the formation and evolution processes which determine the radon daughter activity-size distribution. The activities of this Task were to be computations of the aerosol collision rate densities for spherical particles in the 1-1000 nm diameter range that would constitute the collision kernals for use in computations of aerosol coagulation that are central to aerosol kinetic modeling.

Task 3 was intended to address questions in the physics of cluster formation and evolution by establishing the basis for calculations of collision rate densities of "particles" spanning the range from small clusters to ostensibly "bulk-like" particles of 1-2 nm. While this work would be of immediate use in modeling efforts, work of a longer-range nature was to be undertaken which would involve the application of Lifshitz-van der Waals theory and its derivation from discrete, intermolecular interactions to questions relating to nucleation theory.

To the greatest extent, the foregoing division of tasks has been followed in this project. Specific deviations, whose content will be elaborated in the following discussion of research accomplishments, are in two task areas. Task 1 involves a greater part of the efforts of this project than originally planned. In response to experimental observations at the Radon Measurements Intercomparison Workshops at the University of Illinois in 1988 and 1989, we have undertaken a computational modeling study of the relationship of ultrafine room aerosol electrical charge to the diffusivity distribution of the high-diffusivity fraction of radon daughters. In addition, a major undertaking not originally proposed is the measurement of the electrical mobility distribution of ^{218}Po cluster ions as a function of atmospheric trace gas composition. This addition was made in response both to the need for the information and to the abilities of the student working on the project. In Task 2, the routine calculation of aerosol collision rates was not undertaken because the heterogeneous phase kinetic modeling project which was originally intended to use these results never materialized.

RESEARCH ACCOMPLISHMENTS

Electrical charge carried by radon daughter atoms has for many years been a subject of considerable interest for its possible roles in deposition to aerosol particles, room surfaces, and in the respiratory system as well as for ion-induced nucleation. There is also a literature reporting results from measurement and removal systems for radon progeny which utilize charge. In all cases, there has been an implicit assumption that the radon progeny carry a level of charge which is different from that for the comparable room aerosol. Consequently, a recent finding of Chu and Hopke (Radiation Protection Dosimetry 24(1/4) 207-219, 1988) is of particular interest. Their work showed that the electron affinity of freshly-formed ^{218}Po (or more likely its oxide) from radon decays is sufficiently high that it should be rapidly neutralized by electron scavenging from common atmospheric trace gases. It implies that radon daughter charge under ordinary atmospheric conditions would be attributable to interactions of the neutral polonium-containing molecule in the air. In the invited review of radon daughter charge which we presented at the Fourth International Symposium on the Natural Radiation Environment in 1987, we pointed out that the evidence in the literature provided no contradiction to the Chu-Hopke result and that observed radon daughter charge levels were consistent with the charge level for the normal atmospheric aerosol. See the attached paper from this presentation for further discussion of charging and radon daughter deposition.

In the presence of clustering gases, the polonium-containing molecules from radon decays may form charge-stabilized clusters prior to neutralization, participate in ultrafine particle formation as sites of heterogeneous growth, or attach directly to ultrafine (i.e. high diffusivity) particles formed by any of a variety of processes. In all of these cases, the charge on the particle carrying a polonium atom will have resulted from a diffusion charging process in which a cluster ion attaches to a particle in the 1-10 nm diameter range. Until recently, no experimental data was available on the charging rates of particles this small so the methods used to calculate the rates for ionic attachment to these particles relied upon a picture of the ion-particle interaction energy that was appropriate for macroscopic condensed matter (the classical image potential). In the enclosed paper (Marlow and McFarlane), this picture was followed and provided good fits to the experimental data. Following publication, inconsistencies in the rates for the finest particles were discovered and attributed to an algebraic error resulting in an unsatisfactory fit to the data. To provide an improved description of this important ultrafine aerosol process, we have introduced for the first time into aerosol studies the "jellium" model potential to quantitatively describe the interaction energy at long range between a conducting particle and an ion (here modeled as a point charge). The benefit of utilizing this potential, in its linearized approximation, is that it accounts for the response of the particle's conduction electrons to the field of the ion rather than relying upon a macroscopic picture whose validity is unclear for sufficiently small particles. In the limit of large separations or of larger particles, the jellium and image potentials converge rapidly implying that no inconsistency exists between the generally-accepted approach for larger particles and our contribution. See the attached draft of the paper we are preparing for submission for publication describing our results.

The high diffusivity fraction of aerosol particles is the primary focus of this project because of its central role in delivering radon progeny to the critical cells of the respiratory system. As a result of decay path chemistry, low vapor pressure molecules are produced and participate in nucleation or clustering processes to contribute to aerosol formation and evolution. For modeling this dynamic environment, the distinction of critical nucleation from coagulation of small clusters assumes considerable meaning because it determines how the species should be treated. We have discussed the importance of the long-range interactions in cluster coagulation vs. its secondary role in critical nucleation, the reasons for this difference, and used this distinction as the basis for a quasi-thermodynamic definition of an aerosol. See the attached review of long-range forces in aerosol coagulation.

Once thermodynamically stable molecular clusters are formed, their subsequent growth and contribution to the high-diffusivity fraction of radon daughters is by collisions resulting in coagulation. One of the major components of Aerosol

Microphysics of Indoor Radon is the development of practical methods for calculating the collision rates of ultrafine particles spanning the scale from small molecular clusters to the smallest assemblies of condensed matter. Compositions of the particles are incorporated in these computations via the long-range interaction energies of the particles. Progress has been made toward the objective of developing approximate methods for the calculation of collision rates for clusters. A closely-related objective is to specify the minimum size cluster for which a bulk-matter model of the long-range interactions provide an adequate approximation to the full collective intermolecular energy.

In 1980 under an earlier project, Atmospheric Aerosol Microphysics, we published a rigorous formula (J. Chem. Phys.73, 6384-6287, 1980) for the collision rates of spherical, free molecular regime particles interacting via an attractive potential which becomes singular upon contact of the particles. While the assumption of a singular contact potential is adequate for bulk-like particles, it is questionable for clusters where the contact energies may be comparable with the thermal energy kT. Therefore, we have generalized the formula to apply also to intercluster potentials $E(r)$ obeying the conditions $E(r) < 0$ for $r \geq r_{ij}$ and $\lim_{r \to \infty} r^2 E(r) \to 0$ where r is the distance between the centers of the clusters, $r_{ij} = r_i + r_j$, and r_i is the radius of one of the interacting particles. As in the original 1980 paper, the result is expressed in terms of σ, the value of r where the effective potential, $V_{eff}(r)$, assumes its maximum value. As before, $V_{eff}(r) = J^2/r^2 + E(r)$ where J is the angular momemtum of the particle in the center of mass system. The lower limit of integration σ_1 is given by

$\sigma_1 = \sigma_o$ if there is a $\sigma_o \neq r_{ij}$ such that

$$\frac{\sigma_o}{2}\left[\left(\frac{\sigma_o}{r_{ij}}\right)^2 - 1\right]\left.\frac{dE}{dr}\right|_{\sigma_o} + E(r_{ij}) - E(\sigma_o) = 0,$$ which is equivalent to $V_{eff}(r_{ij}) = V_{eff}(\sigma_o)$.

Otherwise, $\sigma_1 = r_{ij}$. Then, the free molecular collision rate density Φ is

$$\Phi_{ij} = \pi\, v_{av}\, r_{ij}^2 \exp\left\{-\left[\frac{E(\sigma_1)}{kT} + \frac{\sigma_1}{2kT}\left.\frac{dE}{dr}\right|_{\sigma_1}\right]\right\}$$

$$-\frac{\pi v_{av}}{2kT}\int_{\sigma_1}^{\infty} d\sigma \left[\frac{dE}{d\sigma} + \sigma\frac{d^2E}{d\sigma^2}\right]\sigma^2 \exp\left[-\frac{1}{kT}\left(\frac{\sigma}{2}\frac{dE}{d\sigma} + E(\sigma)\right)\right]$$

While stated here for the free-molecular regime, it clearly can also be used in the Generalized Fuchs Formula as in the 1980 paper. This new formula will be published in a paper now in preparation.

Calculation of ultrafine particle collision rates requires coupling the transport with the long-range interaction potential energy of the particles. If the particles contain sufficienty large numbers of molecules to be treated as bulk matter, then Lifshitz's theory of the van der Waals interaction can be used for the potential as we have indicated in a series of papers under our earlier project, Atmospheric Aerosol Microphysics. However, for small clusters, the continuum approximation is meaningless and other methods are required for the potential calculation. We pointed out (J. Phys. Chem 90(11), 2302-2305, 1986) that a derivation by Langbein of Lifshitz theory provides a formulation of the interaction energy between clusters of discrete molecules and that this energy becomes the Lifshitz energy for sufficiently large clusters, assuming no collective electronic states such as conduction bands develop as the number of atoms in the clusters increases. The question naturally arises of the cluster size for which the collision rate is the same whether it is computed via Lifshitz theory or the discrete molecular approach. For the computational modeling of radon daughter behavior, this question is of importance because of the relatively greater ease of use of Lifshitz theory for computations. Thus, if we understand the limitations of this macroscopic theory, then recourse to the microscopic theory will be needed only for a limited range of sizes. In addition, understanding the transition from microscopic to macroscopic potentials will be useful in other areas such as the wetting transition of a liquid on a surface that is needed for understanding aerosol condensational growth.

Under this project, we have conducted a study of the collisions of nearly-spherical clusters and compared the results with bulk-like spherical particles of the same size and composition. The material chosen for study was carbon tetrachloride because its low polarizability and high symmetry allows it to be treated as if it were a sphere for the purposes of constructing the clusters of molecules. Calculations were performed for identical pairs of 13-molecule icosahedrons, 33-molecule dodecahedrons, and 55-molecule icosahedrons in two different orientations. In computing the energy, the multiple induced-dipole interaction is the basis for the attraction and this energy involves the CCl_4 polarizability at all frequencies. This polarizability was computed from spectroscopic data and fit with a sum over classical harmonic oscillators (i.e. Drude model). A pairwise r^{24} interaction was used for the short-range, hard-core repulsion of the molecules. These results were compared with the Lifshitz-Hamaker approximation of the van der Waals interaction between spheres where the frequency-dependent dielectric constant was computed by use of the Clausius-Mosotti equation (as in Langbein's original work) and the density of the spheres were taken to be the densities of the spheres circumscribed about the clusters which they were intended to model. The results of the energy calculations are shown on the following graphs (labelled Fig. V.12).

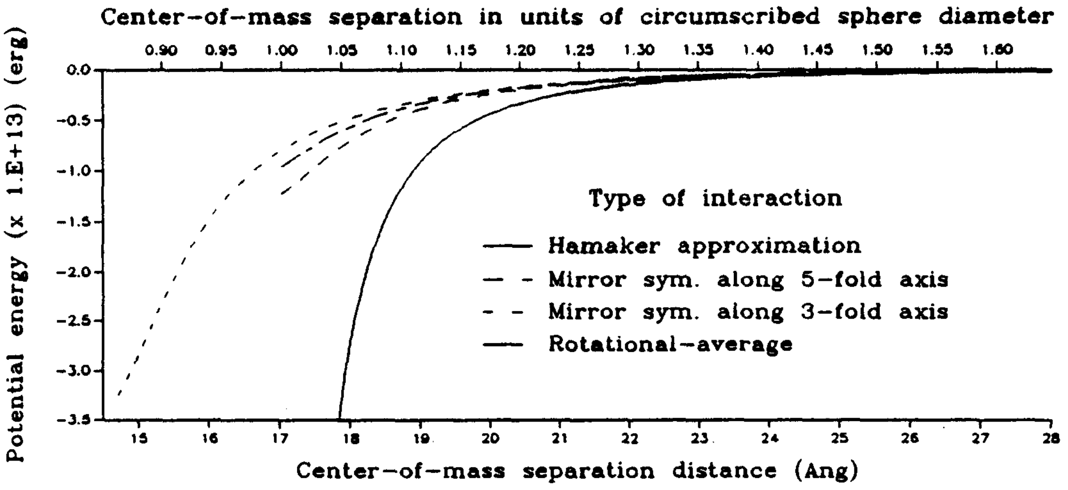

Fig. V.12.a : Interaction potential between 13 molecule icosahedrons

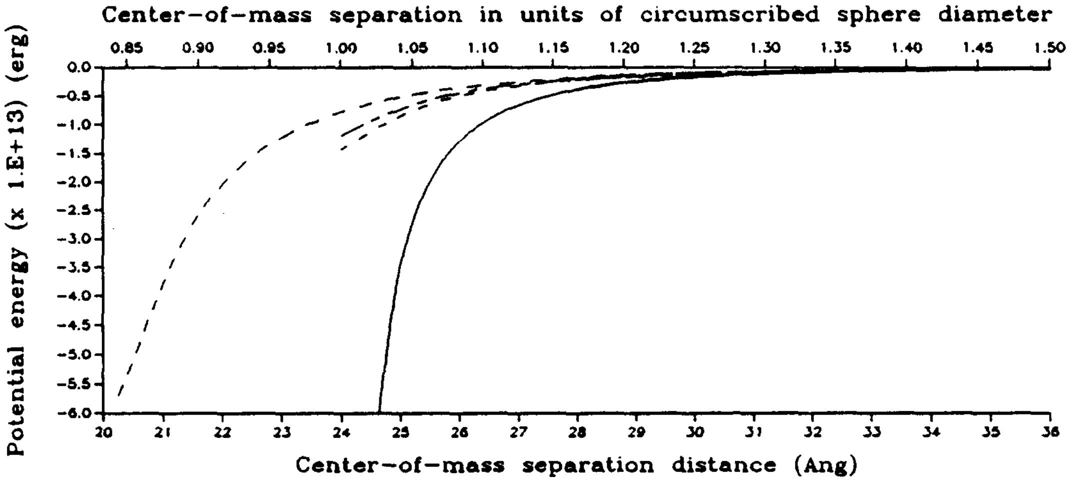

Fig. V.12.b : Interaction potential between 33 molecule dodecahedrons

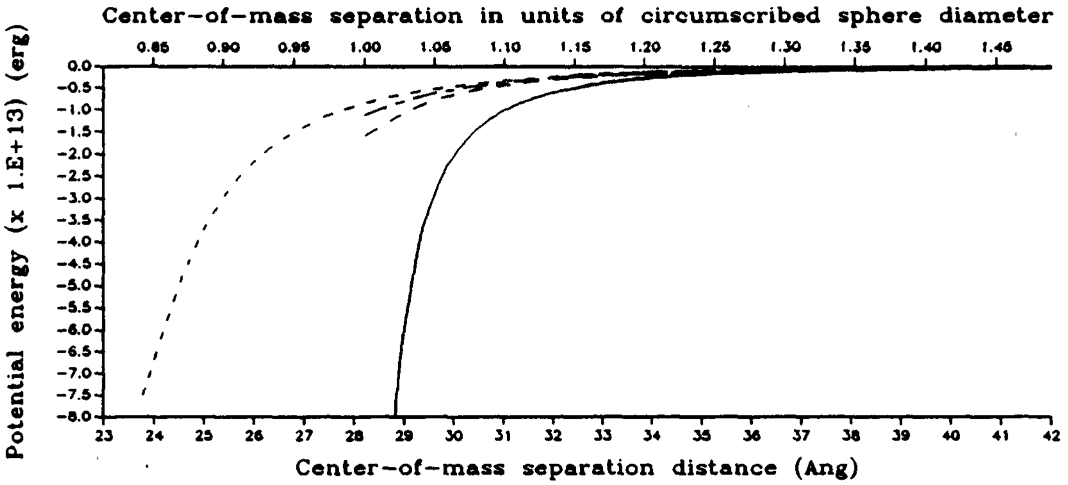

Fig. V.12.c : Interaction potential between 55 molecule icosahedrons

Here, "Hamaker" means the approximate form of Lifshitz theory used and the mirror symmetry designations indicate the relative orientations of the approaching clusters. In these graphs, the lines are terminated in such a way as not to show the short-range repulsion. The important factor here is that at short range, the interaction energies assume realistic values unlike the the Hamaker form which diverges as contact is approached. Another way of presenting this energy data is given in Fig. V.13 where the relative difference between the discrete energies and the Lifshitz-Hamaker energy is given. Note that at long range the relative difference is less than 15% and diminishes with cluster size as expected. We intend to use a more accurate approximation for the Lifshitz interaction in the future as an alternative to determine the extent to which the result shown here is an artefact of the Hamaker approximation.

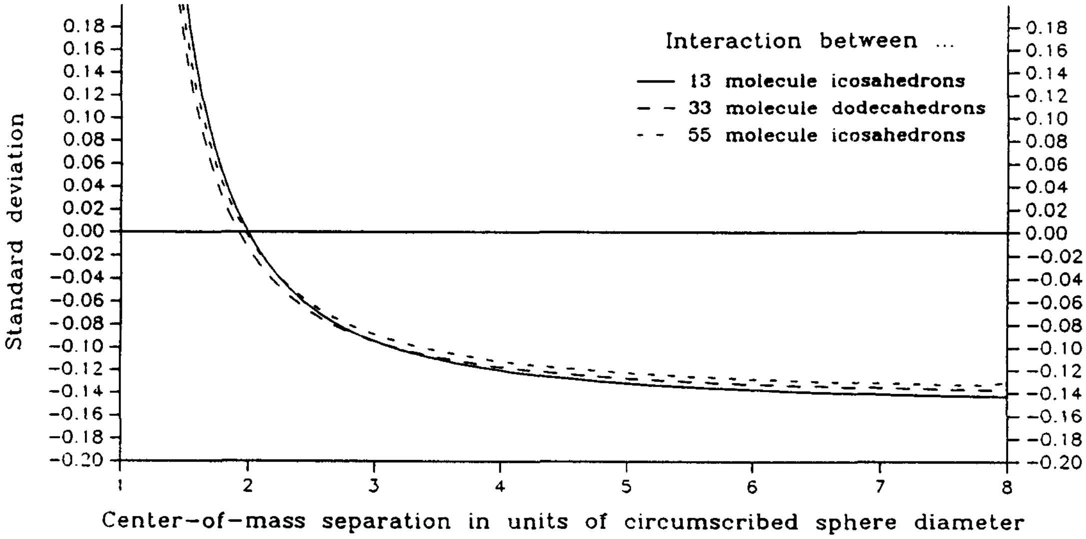

Fig. V.13 : Comparison between Hamaker and exact potentials

For the purpose of describing cluster evolution relevant to indoor radon daughter properties, the objective is to develop readily-calculable collision rates which reflect the dependences of those rates on composition as well as geometry. High-accuracy computations for cluster collision trajectories would be impractical for the number and variety of clusters that ultimately must be treated in modeling calculations. To utilize the potentials described above in developing the required pragmatic approach, collision rates of the clusters were computed by us in two locked-orientation configurations of the clusters and the results compared with the rates for spheres. A cluster collision was treated as if it were between spheres with the interaction energy as a function of separation being that which was computed in the locked-configuration orientations. The results of our computations are displayed in the accompanying Table VI.1. There, Interaction type corresponds to the cluster size, orientations, or types of interactions. The 13, 33, and 55 on the left are the cluster size (i.e. number of molecules); S3 and S5 are for interactions of the clusters oriented with mirror symmetry along the 3- and 5-fold axes of symmetry; AVE is for the rates computed with the potential being the angular weighted average of the preceeding two energies; and

Table VI.1 : Collision rates and derived parameters from alternative methods

Interaction type	13S3	13S5	13AVE	13HAM	33S5	33S3	33AVE	33HAM	55S3	55S5	55AVE	55HAM
r12 (Å)	14.71	17.02	17.02	17.02	20.25	23.99	23.99	23.99	23.77	28.21	28.21	28.21
Marlow (extended)												
Flux (Å^3/sec)	180.1	182.3	181.1	321.9	216.2	218.7	217.4	282.4	234.2	235.9	234	329.3
Capt. radius (Å)	26.98	27.15	27.05	36.07	37.31	37.53	37.41	42.64	44.12	44.28	44.1	52.32
C.R.E.F.	3.365	2.544	2.527	4.491	3.395	2.447	2.432	3.159	3.444	2.464	2.444	3.44
Fuchs												
Flux (Å^3/sec)	130.8	135.9	133	148.9	159.2	165.1	163.2	179.1	172.9	177.8	175	193.5
Capt. radius (Å)	22.99	23.44	23.19	24.53	32.02	32.61	32.42	33.96	37.91	38.44	38.13	40.1
C.R.E.F.	2.444	1.896	1.856	2.077	2.5	1.847	1.826	2.003	2.543	1.857	1.827	2.021
Sceats												
Flux (Å^3/sec)	150.3	154.1	151.8	165.7	181.7	186.1	184.6	198.6	197.1	200.5	198.5	214.9
Capt. radius (Å)	24.65	24.95	24.77	25.88	34.21	34.61	34.47	35.76	40.47	40.83	40.61	42.27
C.R.E.F.	2.808	2.15	2.119	2.312	2.853	2.081	2.065	2.222	2.898	2.094	2.073	2.245

HAM is the Lifshitz-Hamaker potential. R12 is the separation of the spheres at contact. The collision formulas used are designated Marlow (extended), which is the formula presented above in this report of activities, Fuchs, which is the standard free molecular regime aerosol collision rate formula, and Sceats which is a recently-published formula that represents an alternative approximation to Fuchs'. The results are presented as "Flux(Å^3/sec)" which is the collision rate density, "C.R.E.F." which is the enhancement of the rate due to the forces (in effect, Flux/πR12^2) or the collisiion rate enghancement factor, and "Capt. radius" which is the effective collision cross sectional radius. As has been found earlier, the enhancement from Fuchs' rate is lower than from the other rate and as shown for the Hamaker form, much less sensitive to composition, a deficiency which has been pointed out earlier as difficult to reconcile with data. Examination of these results and their dependences implies that as calculated they are not entirely representative of the collisions being described. An analysis of a better approach suggests that utilization of the angular weighted average separations at contact, R12, would better reflect the actual physics of the long-range forces and capture trajectories. Table VI.2 below displays the rates with appropriate separations as calculated according to this averaging procedure. Now the rotationally-averaged C.R.E.F.'s are within the range imposed by the limiting cases for the cluster orientations (S3 and S5 in Table VI.1). The relative differences between the collisions rates derived from the macroscopic and microscopic rates are 49%, 15.1% and 14.4% resepectively for the 13,33, and 55 molecule clusters' interactons. This time, a convergence is obseved as the clusters increase in size. Though a greater change in the relative difference between the 33-molecule dodecahedron and 55-molecule icosahedron might have been expected, its absence is just a manifestation that the dodecahedral structure is a better approximation to a sphere than is the icosahedral structure, a property of these polyhedrons noted elsewhere in this study.

Table VI.2 : Collision parameters based on an average contact separation
(from the Marlow-extented approach)

Interaction model	13AVE	13HAM	33AVE	33HAM	55AVE	55HAM
Averaged r_{12} (Å)	15.58		22.59		25.44	
Collis. rate (Å^3/sec)	181.1	**269.6**	217.4	**250.3**	234.0	**267.8**
Capture radius (Å)	27.05	**33.01**	37.41	**40.15**	44.10	**47.18**
C.R.E.F.	**3.016**	4.491	**2.743**	3.159	**3.006**	3.440

We are currently engaged in an experimental study of the trace gas dependences of the electrical mobility distributions of ^{218}Po cluster ions formed in radon decays. An aspiration mobility separator has been developed to examine cluster ions in the 0.05 – 5.0 cm^2/volt-sec range. The radioactive ions are deposited along a strip of alpha-track film which is exposed for 4 hours per run to a radon concentration of 600 pCi/l and then etched. Decay tracks are then counted to give the distributions. Initial runs were made in the first half of 1989 at the University of Illinois using Dr. P. K. Hopke's radon chamber and they will be continued in the fall of 1989 in Dr. Hopke's new facility at Clarkson University.

Two additional activities supportive of measurement efforts in the Indoor Radon Program are also in progress. In one, the angular position of attachment of high-diffusivity particles to a cylinder situated transverse to an air stream is being calculated. Direct simulation via finite element computations of the gas flow serve as the basis for these computations. The motivation for this study is that it serve as a model for the attachment of radon daughters to ultrafine mesh screen that are widely used in high-diffusivity radon daughter measurements. At present for Reynolds numbers up to 2, no deviation from attachment based upon strictly laminar flow has been observed.

The second supportive activity is motivated by observations at the 1988 University of Illinois Radon Intercomparisons Measurement Workshop. The aerosol size distribution in the 3-10 nm size range from a radon chamber as measured by a differential mobility analyzer using the charge accumulated strictly by background (i.e. room air) charging agreed with the activity-size distribution in the same aerosol. We are currently doing modeling calculations under a variety of assumptions to determine if this was a fortuitous result or one that can be anticipated. If it is a result of general validity, then it might suggest a means of determining the activity-size distribution based upon total activity and DMA determined size distribution.which would be much less time-consuming than the methods currently used.

Plans for continuation of present objectives and possible new objectives in consideration of past results

As discussed above, major components of Aerosol Microphysics of Indoor Radon were originally intended to provide theoretical and computational information for the heterogeneous-chemical modeling of radon daughter properties. Since the modeling activity did not develop elsewhere as planned, we propose in our renewal application to expand the scope of this project to include this activity. A second addition we propose is the inclusion of gasdynamics in both our activities of experimental support (Task 1) and fundamental research for modeling. Apparently neither heterogeneous modeling nor the effects of room airflow are being systematically developed elsewhere and without this capability, comprehensive understanding of what affects the way radon exposure is related to dose to the critical cells of the respiratory system will not be available.

Graduate students educated.

Aerosol Microphysics of Indoor Radon has wholly supported the education and research of the following three degree students:

Ms. Xiao-Wei Zhu completed her Master of Science in Health Physics with a thesis <u>Ultrafine Aerosol Diffusion Charging: An improved Interaction Potential for Metallic Particles.</u>

Mr. Alexis Amadon is scheduled to present and defend his thesis for the Master of Science in Nuclear Engineering on September 8, 1989. The title is <u>Interaction Potentials of Molecular Clusters with Applications to Collision Rate Calculations.</u>

Mr. Suk-Chul Yoon is conducting research for his Doctor of Philosophy Degree in Health Physics with completion anticipated for the spring semester of 1990. The dissertation will be on the trace-gas dependence of polonium cluster ion electrical mobilities.

In addition, three other students have been temporarily supported in research for this project:

Mr. David L. McFarlane performed the calculations resulting in the published paper " A theoretical fit of charging data: comments on "Unipolar charging of ultra-fine particles' "

Mr. Subburamu Venkatesh is currently performing calculations modeling the charge state of the ultrafine aerosol under various assumptions, computing the resultant size distribution that would be measured by a differential mobility analyzer, and comparing the result with the size distribution of the high-diffusivity fraction of radon daughters.

Mr. Anand Gangadaharan is currently calculating the attachment of high diffusivity particles to small cylinders in a flowing gas stream.

Finally, Ms. Valli Arunachalam, who has a Master's Degree in Physics from the University of Madras, is joining our group and will begin research work on a topic of importance in indoor radon studies.

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The jellium potential in diffusion charging of ultrafine metallic particles. X.-W. Zhu and W. H. Marlow. to be submitted to Aerosol Science and Technology in 1989.

Exact formula for collision rates of free-molecular regime particles interacting via finite, attractive contact potential. A. Amadon and W. H. Marlow. (in preparation for 1989 submission).

Calculation of molecular cluster collision frequencies including van der Waals' interactions. A. Amadon and W. H. Marlow. (in preparation for 1989 submission).

Aerosol charge vs radon daughter diffusion in the measurement of the high-diffusivity fraction of radon daughters. S. Venkatesh and W. H. Marlow. (in preparation for 1989 submission).

Other work in progress which will result in 2 or 3 additional publications to be submitted in 1990 will include the studies discussed above of high-diffusiviry radon daughter attachment to cylinders and the dependence of the electrical mobility distribution of ^{218}Po cluster ions on atmospheric trace gases.

Present state of knowledge, significance in the field of environmental matters, and needed future investigations.

To compute the radon daughter activity-size distribution in sufficient detail for dosimetry, more experimental and computational information is required than is currently available. Compounding the problem is the possibility that compositions, in addition to diffusivities, of the ultrafine particles may play significant roles in determining the ultimate biological effects. For example, if the molecule containing the ^{218}Po atom is chemisorbed onto an insoluble particle, then its deposition on a critical cell may not result in its being dissolved by the body fluids and residing on the critical cell surface but rather 3-5 nm further away. Conversely, if the host particle is an electrolyte, then growth and deposition higher in the respiratory system may occur. One may speculate that only a weakly soluble particles too small to serve as a heterogeneous nucleation site upon entry into the respiratory system could effectly transport activity deep into the respiratory system. Thus, the task of modeling indoor radon daughter evolution in the detail required for useful dose estimates may well grow beyond any means of accurate computation. In these circumstances, experimental data must be supplemented by modeling computations based upon a variety of pictures of the processes which effectively determine the required daughter properties. Since in this picture, the modeling is used as an investigative tool to extend the usefulness and validity of expeimental results and not simply as a means of curve fitting, modeling approaches themselves must be correct beyond any question in order not to introduce spurious error. At present, there is a mixed record on those approaches. The classical theory of nucleation seems to provide an adequate description of single-component particle formation which probably doesn't occur in the atmosphere. Multicomponent nucleation methods are under development, but adequate thermodynamic information on the precritical clusters is largely speculative and will require considerable effort to develop. In many cases of relevance, the difficulties of critical nucleation may possibly be bypassed for clustering of low vapor pressure molecules formed by radiolysis. Following the the formation of the smallest molecular clusters, subsequent clustering probably occurs irreversibly (due to available modes to dissipate the collisional energy). For both clustering and nucleation, the full chemistry of the decay path must be clarified for the subsequent modeling to be possible. With the addition of electrical charge, the possibility of charge-induced nucleation must be included. The question of interaction of the gas phase species with the room surfaces must also be taken into account, a study which has barely been undertaken and certainly not in the detail required to determine what feedbacks occur to affect radon daughter evolution. Finally is the question of even how to do the kinetic modeling correctly. We normally assume concentrations of species that are volumetric averages. However, in the decay path, transient, extremely high concentrations of various species occur and the effects of this high local loading is not clear for modeling.

As a result of a number of efforts around the world during the last twelve years, there is a growing realization that the submicrometer aerosol size spectrum is not always meaningfully expressed as consisting of "nucleation" and "accumulation" modes alone. The lacking element is new particles that appear in the aerosol size spectrum in the range of diameters from below 1 nm to about 10 nm. These particles are the manifestation of atmospheric chemical transformation processes that can be photochemical, thermal, or radiolytic in origin. Thus, the high-diffusivity fraction of radon daughters is only one element of the subject of new particle formation so that once the chemically stable state of the decay atom is reached, its interaction and growth processes are common across a broad range of subjects. In particular, the development of the ability to describe these processes for radon daughters will also find application throughout the field of air chemistry. One area of particular currency is in the field of global warming. An understanding of the formation and behavior of ultrafine particles is required for their contributions both to the growth of preexisting aerosol and for their roles in forming larger new particles (e.g. the nucleation mode). In both cases, contributions will be made to cloud condensation nuclei formation as well as to the presence of passive (i.e. noncondensing) nuclei. In addition to their effects on cloud microphysics, chemically specific particle formation also is important in the planetary albedo and in stratospheric aerosol questions.

The present division of Federal support for overall research program

In May, 1989, the Principal Investigator was awarded a new, 3-year research project by the Office of Naval Research entitled "Dynamical Processes of Gas-Phase Clusters." The content of this project is entirely complementary to Aerosol Microphysics of Indoor Radon. Its subject of study is the energy accommodation processes occurring in colliding clusters and how long-range interaction energies may affect cluster collisions and interactions with particular emphasis on cluster configuration. This project will occupy approximately 1/6 of our efforts in contrast with the 1/4 effort by Aerosol Microphysics of Indoor Radon.