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Study of the Atmospheric Chemistry of Radon Progeny in Laboratory and Real Indoor Atmospheres

Progress Report for the Period

May 1, 1993 to January 31, 1994

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ABSTRACT

This report covers most of the first year of the current grant period of Clarkson University's continuing study of the chemical and physical behavior of the ^{218}Po atom immediately following its formation by the alpha decay of radon. Because small changes in size for activity in the sub-10 nm size range result in large changes in the delivered dose per unit exposure, this behavior must be understood if the exposure to radon progeny and its dose to the cells in the respiratory tract are to be fully assessed. In order to pursue this general goal, two areas of radon progeny behavior are being pursued; laboratory studies under controlled conditions to better understand the fundamental physical and chemical processes that affect the progeny's atmospheric behavior and studies in actual indoor environments to develop a better assessment of the exposure of the occupants of that space to the size and concentration of the indoor radioactive aerosol. Thus, two sets of specific goals have been established for this project.

The specific tasks of the controlled laboratory studies are 1.) Determine the formation rates of $\cdot\text{OH}$ radicals formed by the radiolysis of air following radon decay, 2.) Examine the formation of particles by the radiolytic oxidation of substances like SO_2 , ethylene, and H_2S to lower vapor pressure compounds and determine the role of gas phase additives such as H_2O and NH_3 in determining the particle size, 3.) Measure the rate of ion-induced nucleation using a thermal diffusion cloud chamber, and 4.) Measure the neutralization rate of $^{218}\text{PoO}_x^+$ in O_2 at low radon concentrations.

The specific tasks of the exposure studies in occupied indoor spaces are 1.) Initiate measurements of the activity size distributions in actual homes with occupants present so that the variability of the indoor activity size distributions can be assessed with respect to indoor aerosol sources and general lifestyle variations of the occupants, 2.) Initiate a prospective study of the utility of measurement of deposited ^{210}Pb embedded in glass surfaces as a measure of the long-term, integrated exposure of the population to radon, and 3.) Develop the methodology to determine the hygroscopicity of the indoor aerosol so that the changes in deposition efficiency of the radioactive indoor aerosol with hygroscopic growth in the respiratory tract can be assessed.

This report describes the progress toward achieving these objectives.

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INTRODUCTION

The health effects of the radon decay products are strongly dependent upon their behavior in the indoor atmosphere. Particularly for ^{218}Po , the partitioning between the "unattached" state and the "attached" forms, combined with the preexisting aerosol particles, has a significant impact on the calculation of the dose to the lung resulting from a given decay product concentration. Thus, the chemical and physical behavior of the ^{218}Po atom immediately following its formation by the alpha decay of radon must be considered if its health implications are to be fully assessed. This report of our continuing program to investigate the chemical and physical behavior of radon progeny covers the period from May 1, 1993 to January 31, 1994 since with the renewal of the grant, the starting date for the grant period was adjusted to May 1.

LABORATORY STUDIES

The principal areas of the laboratory studies during the current period have been 1.) Determination of the formation rates of $\cdot\text{OH}$ radicals formed by the radiolysis of air following radon decay, and 2.) Measurement of the rate of ion-induced nucleation using a thermal diffusion cloud chamber, 3.) Development of systems to measure the ion-cluster behavior of the $^{218}\text{Po}^+$ in the presence of a variety of typical indoor volatile organic compounds.

OH Radical Production Rate

We have measured the HO_x radical concentrations produced from radon decay with the chemical amplification system developed by Cantrell *et al.* (1984). In the chemical amplifier system, $\cdot\text{OH}$ radicals produced by ^{222}Rn decay in air participate in a series chain reactions with high concentrations of CO (10%) and NO (ppm), resulting in high concentrations of NO_2 . The product NO_2 is measured by luminol chemiluminescence. These reactant gases as well as radon-laden N_2 is mixed into a stream of purified air. The air then passes through a flow-through detector system based on the gas-surface reaction of NO_2 with a 0.01 M Na_2SO_3 , 0.05% methanol, 0.001M aqueous luminol solution of pH 12 (Maeda *et al.*, 1980). Contaminant NO_2 in the reactant NO gas is removed by passing the gas stream over FeSO_4 . A manuscript describing these studies has been accepted for publication in the *Journal of Atmospheric Chemistry*.

A laser-induced fluorescence system was established using an excimer laser system. We determined the sensitivity of the system with $\cdot\text{OH}$ produced by as high a radon concentration as we could put into the chamber. The response of the system was found to be insufficient to clearly

observe a signal above the noise and thus, we have ceased further work on this aspect of the measurement. It is clear from these studies that there is not a sufficient production of OH radicals following the decay of ^{222}Rn to have a significant effect on indoor atmospheric chemistry. Thus, further work in this area is not planned.

Ion-Induced Nucleation

With respect to the ion-induced nucleation, there are two proposed areas for study. The first one is to examine the potential for ion-induced nucleation of volatile organic compounds by radon decay in indoor environments. The second one is to study the combined oxidation and nucleation mechanism for the $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ system with radiolytic formation of H_2SO_4 .

For the first area, we have already finished the experimental work and have submitted two papers to *Journal of Chemical Physics* (He and Hopke, 1993a&b). The first of these papers has been accepted for publication. The objectives of this experimental study are twofold. The first objective is to verify if there is an effect (or effects) of ^{222}Rn on the particle formation for most VOC vapors found in indoor air by the mechanism of ion-induced nucleation. The second is to obtain a more detailed understanding of the role of the molecular properties such as orientation and distortion polarization on ion-induced nucleation.

Our approach is based on the experimental determination of the critical supersaturation for both ion-induced nucleation and homogeneous nucleation (without radon added) in a thermal diffusion cloud chamber and relating the trends of observed critical supersaturation values with the molecular polarization, both orientation and distortion, for the different classes of compounds under investigation. There are two indicators of the catalytic effect of the ion on the nucleation process. First, alpha tracks are visible in the nucleation zone (about 80% chamber height) when observed with the forward scattered light if sufficient vapor molecules nucleate on the ions at supersaturated conditions. The second indicator is the difference in the critical supersaturations between homogeneous and ion-induced nucleation at the same nucleation rate. A lower critical supersaturation is expected for ion-induced nucleation.

For a specific substance, if there is a critical supersaturation difference between the homogeneous nucleation and ion-induced nucleation and there are ion-tracks appearing in the chamber, this substance can undergo ion-induced nucleation. If there is no obvious difference between the critical supersaturation, ion-induced nucleation is not an important particle formation mechanism for this substance. By examining the relative critical supersaturation difference between

the homogeneous and ion-induced nucleation in terms of some common molecular properties (polarization) over in a broad range, the ion-induced nucleation behavior for different VOCs can be grouped according to these properties. Subsequently, these results may be useful in the development and testing of an improved ion-induced nucleation theory that would be able to predict the nucleation behavior in terms of the properties of the vapor molecules.

The VOCs used in our study and their properties are listed in the Table 1. These compounds include short chain alcohols, carboxylic acids, aldehydes and ketones, aromatic hydrocarbons compounds and paraffins. The choice of these compounds is based on following consideration. First, they have relatively high concentrations and range from the order of micrograms per cubic meter to more than 100 times higher depending upon indoor VOC sources and ventilation rates (Daisey and Hopke, 1993). Second, a broad range of distortion (from 3.29 to $15.9 \times 10^{-24} \text{ cm}^{-3}$) and orientation polarization (from 0 to 2.88 Debye) is covered. Third, the choice of the compounds was limited to the substances whose boiling points must be much higher than their triple points because the condensing vapor must not be allowed to freeze on the condensation surface and the temperature of the liquid pool must be below its boiling point.

Except for benzene and the aliphatic compounds, alpha tracks and relatively lower critical supersaturations were observed for all of the substances listed in Table 1. These results indicate that these substances are capable of undergoing ion-induced nucleation. The general agreement between our results and the results from previous study (Rabeony and Mirabel, 1987) for o-xylene and the alcohols is good although different ionization methods were used and the focal point in this study is critical supersaturation differences whereas the focal point in prior studies was the sign effects of the ion on nucleation. For benzene, the high vapor pressure and narrow difference between the boiling and freezing points makes the critical supersaturation measurement very difficult. However, no clearly observable alpha tracks occurred inside the chamber, indicating that the ions may had little nucleating effect on benzene. For the C_6 to C_9 straight chain paraffins, there were no obvious differences between the critical supersaturations for homogeneous and ion-induced nucleation at the same temperature. No alpha-track were observed after radon was added to the system for these compounds.

Our experimental results also show that the nucleation rates increase substantially with increasing radon at low activity concentrations, but leveled off at high concentrations because of vapor depletion preventing the growth of droplets to a measurable size. For short chain alcohols, this result is shown in Figure 1.

Table 1. List of Compounds Examined for Ion-Induced Nucleation and their Associated Molecular Properties

VOC	Molecular weight	Boiling point (C)	Freezing point (C)	Vapor Pressure (Torr)	Surface tension (dyn cm ⁻³)	Dipole (Debye)	Polarization (10 ⁻²⁴ cm ³)
Methanol	32.042	64.40	-97.70	55.42	23.28	1.700	3.29
Ethanol	46.069	78.30	-114.10	23.54	25.06	1.690	5.41
1-Propanol	60.097	97.20	-126.30	7.04	25.88	1.680	6.74
n-Butanol	74.123	117.70	-89.3	2.05	25.38	1.666	8.88
Formic Acid	46.026	100.60	8.30	19.79	38.77	1.41	3.40
Acetic Acid	60.053	117.90	16.60	6.07	29.10	1.74	5.10
Propionic Acid	74.080	140.83	-20.70	1.18	27.58	1.75	6.90
Acetone	58.080	56.20	-95.00	116.42	25.86	2.88	6.40
M. E. Ketone	72.107	79.59	-86.70	45.51	26.30	2.78	8.13
Propionaldehyde	58.080	47.93	-80.00	164.99	27.22	2.52	6.50
n-Butyraldehyde	72.107	74.78	-96.40	52.37	26.22	2.72	8.20
Benzene	78.155	80.10	5.53	45.52	30.09	0.00	10.74
Toluene	92.141	110.63	-94.91	12.43	29.71	0.36	12.30
o-Xylene	106.17	144.41	-25.18	2.47	31.41	0.62	14.90
n-Hexane	86.178	68.74	-95.30	75.70	19.06	0.00	11.90
n-Heptane	100.206	98.43	-90.60	20.65	21.19	0.00	15.00
n-Octane	114.23	125.70	-56.80	5.23	22.50	0.00	15.90
n-Nonane	128.26	150.80	-53.50	1.71	23.50	0.00

Figure 1. . Nucleation rate as a function of radon concentration within the chamber.

Figure 2. Relative critical supersaturation differences changes against the total polarization (10^{-24}cm^3).

From the experimental data, the relationship between the ion-induced nucleation and the polarization of the vapor molecules is examined. Figure 2 is the plot of $\Delta S_{CR}/S_{CR}$ against the total polarization of the vapor molecules. In this figure, $\Delta S_{CR}/S_{CR}$ is the values of the critical supersaturation differences between the homogeneous nucleation and ion-induced nucleation divided by the critical supersaturations for homogeneous nucleation. It provide a measure of the relative importance of the ion on the nucleation process. From the experiment results shown in Figure 2, several observations can be made. First, the increasing trend of $\Delta S_{CR}/S_{CR}$ with respect to both orientation and distortion polarization of the vapor molecules suggests that the effect of the ion on the nucleation process increases with the polarization of the vapor molecules. Second, compared to the distortion polarization, orientation polarization plays a more important role in the ion-induced nucleation process. Third, there is considerable scatter in this figure. The reasons for the scatter may result from that other physical properties also contribute to the variation of the critical supersaturation, such as saturation vapor pressure and surface tension. Also, even with the orientation polarization itself, there are different impacts on the nucleation process. From a study of the homogeneous nucleation of highly polar liquids at lower temperature, El-Shall and coworkers (Wright and El-Shall, 1993) suggested that dipole orientation may result in an increase in surface tension and spherical droplet elongation. These change will then increase the free energy barrier and the critical supersaturation for nucleation. From this discussion, it can be seen that the impact of the polarization of the vapor molecules on the nucleation process is a complicated issue. In order to establish a detailed understand of ion-induced nucleation, further theoretical analysis will be necessary.

The second area of ion-induced nucleation study is to study H_2SO_4 - H_2O system with radilytic formation of H_2SO_4 . These studies will be discussed below in the section describing the research yet to be performed.

Neutralization Rates

We have been collaborating with Drs. Y.S. Cheng and coworkers at ITRI on the neutralization of the $^{212}Pb^+$ ion formed in the decay of ^{220}Rn . In these studies, we have repeated the type oif experiment performed by Goldstein and Hopke (1985), but with ^{220}Rn rather than ^{222}Rn . the results of this study have been submitted for publication in *Health Physics* (Cheng *et al.*, 1994). We expect that the revised version recently submitted will be accepted for publication. From the results

of these studies, it appears that we may have overestimated the ionization potential of the $^{218}\text{PoO}_x$ in our earlier study. Thus, we need to continue these studies to better define our methods to estimate this basic property of this airborne polonium species.

We want to complete our earlier studies of the neutralization in ultrapure O_2 (Shi and Hopke, 1991). In order to complete these experiments, we have been communicating with Professor A.J. Howard of the Department of Physics, Trinity College, Hartford, CT. Professor Howard has developed a system for measuring the neutralization rate for $^{218}\text{Po}^+$ ions under well controlled conditions (Howard and Strange, 1991). He has obtained results in good agreement with our earlier measurements (Shi and Hopke, 1991). It will be easier to make the critical measurements of the O_2 neutralization rates using their system. We are planning to have Dr Howard and his students visit us in order to begin these experiments. However, he had some minor repairs that needed to be made to his system and we expect that early in 1994, he will be able to visit Potsdam to initiate these experiments.

Characterization of $^{218}\text{Po}^+$ Ion Clusters

We have begun two avenues of study to examine the nature of the ^{218}Po species formed immediately after their formation from ^{222}Rn decay. These studies involve the use of two different methods to measure the "size" of the $^{218}\text{Po}^+$, a circular electrical mobility spectrometer and a hypersonic impactor.

Circular Electrical Mobility Spectrometer

In our original proposal we have suggested continuing the experiments begun in conjunction with Professor William Marlow and his student that resulted in the work presented in Yoon *et al.* (1992). However, there was a serious problem with the resolution in that system. Although we have borrowed the unit from Prof. Marlow, we have subsequently discovered an alternative system that would appear to have a better potential for providing the resolution needed to examine the electrical mobility of the initial cluster ions. This system is a new form of mobility analyzer originally developed at CEA in Fontenay-aux-Roses by Pourpux and Boulard (). The circular electrical mobility analyzer (C.E.M.S.) should be able to provide much better resolution in the mobility spectrum because of much better control of the flow characteristics within the spectrometer. The circular electrical mobility spectrometer design is shown in Figure 3. The particles enter through a circular slit in the top plate (#1) and mix with the sheath air. The field carries the charged particles

to the collection surface (#6). The original system uses a Si wafer such as those used to make integrated circuits as the collection surface. A surface scanner can thus be used to measure the particles collected on the surface where the mobility spectrum is observed by examining the radial pattern of particle deposition.

Figure 3. . Schematic diagram of the CEA circular electrical mobility spectrometer.

In our study, we are modifying this design by substituting a disk of CR39 etched track detector for the Si wafer. We can collect the activity onto the plastic that has been coated with a thin conducting surface and the subsequent decay will cause a track to be deposited. After etching the disk, we can examine the track density as a function of radial distance in order to obtain the mobility spectrum. We plan to examine ions in the range of 0.03 to $0.3 \text{ V cm}^2 \text{ s}^{-1}$. We can use a voltage of $\leq 100 \text{ V}$ and a flow rate of $\leq 20 \text{ Lpm}$ to obtain this measurement range.

We have been given a system constructed in France by the same shop as made them for the CEA. In this system the top was made of aluminium. In addition, we have constructed 2 additional stainless steel systems using the CEA plans. One of these systems was constructed for the group at the Inhalation Toxicology Research Institute who are conducting parallel measurements on thoron progeny ions. We have acquire 6 140 mm CR39 disks from TASL at the University of Bristol in England. They had some difficulties in making these test disks and we may have some problems obtaining more in the future. We are exploring alternative sources of this material. We have acquired a CCD video camera for our optical microscope as well as image analysis software that can

provide counts of the tracks per unit area as a function of radial distance. Thus, all of the facilities are now in place and initial tests are being performed. We expect to begin detailed experiments once a continuing source of CR39 disks has been obtained.

Hypersonic Impaction of Radon Progeny.

The ultimate aim of this project is to determine the size of radon progeny as a function of the prevalent gas composition in order to have a better understanding of the behavioral pattern of radon progeny in indoor atmosphere, and to better assess the exposure of the radon progeny and its dose to the cells in the respiratory tract. The C.E.M.S. provides information on the ion mobility of the ^{218}Po . Although we can relate the mobility to a diffusion coefficient and then to size, we cannot directly obtain a molecular weight. If we can use inertial separation of the Po species, then we can obtain an aerodynamic diameter that will include the particle density. Thus, from the diffusion diameter and the aerodynamic diameter, we can estimate the density and the molecular weight. This ability will permit us to finally determine the nature of the Po species in the presence and absence of O_2 , estimate the number of water vapor molecules that cluster around the $^{218}\text{Po}^+$ as a function of relative humidity, and a number of other problems that have previously been intractable.

To achieve this objective, a hypersonic impactor based on the work of Prof. Juan de la Mora of Yale University (de la Mora *et al.*, 1990 a&b; de la Mora and Schmidt-Ott, 1993) has been designed using plans provided to us by Prof. de la Mora and built as shown in Figure 4. This impactor utilizes the distance between the jet nozzle and the impaction plate to control the particle inertia. It is made of a stainless steel high vacuum cross with windows on its four arms and consists of two main parts, namely: nozzle and collection assemblies. In the nozzle assembly, a micrometer screw gage is attached to the nozzle via the stagnation chamber. This permits the distance between the nozzle and the impaction plate to be carefully varied.

The collection plate or the impaction plate is a particle-implanted passivated surface (PIPS) detector which is connected to a computer-controlled multichannel analyzer via a pre-amplifier and an amplifier. A α -particle detector is employed in order to utilize the radioactivity of the radon progeny to quantify in real time the amount of these progeny collected on the impaction plate. By integrating the peaks for both ^{218}Po and ^{214}Po , we get one alpha count per deposited nucleus since the counting efficiency for each nuclide is essentially 50%. Thus, we are able to detect each deposited nucleus with a very high signal-to-noise ratio.

The fabrication of the impactor has been completed, and it is currently being tested. The procedure is to generate the progeny inside a radon chamber under well controlled laboratory conditions and then sample with the hypersonic impactor for various nozzle-plate distance. A plot of collection efficiency as a function of nozzle-plate distance will permit the determination of the aerodynamic diameter. Initial test-runs suggests that the impactor and the electronics setup for data acquisition are working.

The radon chamber is cylindrical in shape, and it has an internal volume of 32 liters with 6 sampling ports and 8 BNC bulkhead fittings for electrical and electronic connections. Radon-laden nitrogen from the radon source flows into the chamber at 2 liter per min and leaves via a downstream gas pump which exhausts into the hood. Initially, we are trying to determine the aerodynamic diameter of ^{218}Po in high purity nitrogen (99.998%) to ensure that the freshly-formed ^{218}Po does not form molecular clusters with the gas impurities. Several experiments have been conducted in this respect. However, from the experimental results, it seems that ensuring uniform progeny concentration over a sufficiently long period of time, which is a *sine qua non* in the use of a one-stage impactor for size spectroscopy, inside the chamber coupled with the need to have statically significant number of ^{218}Po particles deposited on the impaction plate is the main difficulty at present. Thus, efforts are being made towards solving these two problems by increasing the concentration and minimizing the wall losses.

After solving the above-mentioned problems, we intend to conduct a series of laboratory experiments to determine the size of molecular clusters form by ^{218}Po in the presence of other gases, such as O_2 , H_2O , SO_2 , NO_x , and VOCs that are typically found in both outdoor and indoor environments. Above all, we would like to see how ^{218}Po size changes with relative humidity and the effect of the presence and absence of O_2 on the mass of the Po species.

Thus, the laboratory aspects of the project are underway and making good progress.

Figure 4. Cross-sectional view of the hypersonic impactor
REAL INDOOR ATMOSPHERE STUDIES

The specific tasks of the exposure studies in occupied indoor spaces are 1.) Initiate measurements of the activity size distributions in actual homes with occupants present so that the variability of the indoor activity size distributions can be assessed with respect to indoor aerosol sources and general lifestyle variations of the occupants, 2.) Initiate a prospective study of the utility of measurement of deposited ^{210}Pb embedded in glass surfaces as a measure of the long-term, integrated exposure of the population to radon, and 3.) Develop the methodology to determine the hygroscopicity of the indoor aerosol so that the changes in deposition efficiency of the radioactive indoor aerosol with hygroscopic growth in the respiratory tract can be assessed.

Exposure Assessments in Occupied Houses

The measurement of activity-weighted size distributions in normally occupied homes are made using the automated, semi-continuous, graded screen array system described by Ramamurthi and Hopke (1991). During the present period, we have not made any additional measurements in homes, but have concentrated on analyzing and compiling the results of our prior studies. Several of these

studies have now been published or accepted for publication (Hopke *et al.*, 1993a; Wasiolek *et al.*, 1993; Hopke *et al.*, 1994).

The International Commission on Radiological Protection (ICRP, 1994) has adopted a lung deposition model, the dose conversion coefficients that have been varying over the past several years will remain constant. We need to go back to earlier studies and recalculate the dose based on the final model and associated dosimetry which we are doing in collaboration with Dr. A.C. James of Pacific Northwest Laboratories. With the move toward the second phase of the BEIR VI study of radon risk, these results will be needed to help improve the dosimetric extrapolation from mines to homes as suggested by the earlier NRC study (NRC, 1991).

We are continuing to study the various approaches to the deconvolutions of the activity-weighted size distributions in order to understand the nature of the differences that have been seen (Hopke and Paatero, 1994). At this time, it is not clear why these differences exist and which results are correct. Further studies in other homes are planned for the near-term future and we have a considerable amount of existing data from past studies that requires analysis and preparation of appropriate manuscripts.

Prospective Study of ^{210}Po in Glass

Prior studies by Dr. R. Lively of the Minnesota Geological Survey (Lively and Ney, 1987) and Christer Samuelsson of Lund University, Sweden (Samuelsson, 1988) on the relationship between measured ^{210}Po embedded in glass and ambient radon concentrations. When ^{214}Pb or ^{214}Bi is deposited on a surface in a room, they will adsorb to the surface. When the resulting ^{214}Po emits its α particle, the ^{210}Pb nucleus recoils with sufficient energy to embed it about 40 nm into the surface. Thus, it is thus below the surface and is not removed by cleaning, but it is close enough to the surface that the 5.3 MeV α from ^{210}Po can be observed.

All of the previous work has been done using existing glass surface in the house and a year of α measurements. Lively and Steck (1991) has demonstrated the kind of wide variability that can occur even in the annual average radon concentration. Also people are not always certain when the particular piece of glass was placed into its current environment. Thus, some uncertainty exists in the value of the calibration constant. Furthermore, there has not been a study of the variations that might exist within a room or between rooms in the same house. These are the questions our study hopes to address.

Our study plan was to place 150 pairs of 8" x 10" pictures into houses. So far we have placed 144 pair of pictures into homes from California to New York. Another 20 pictures in groups of ten have been placed into two schools (one in Maine and the other in South Dakota). In 50 houses, the pictures are placed in the most lived in room of the house (living or family room). In 50 houses, the pair of pictures go in the master bedroom and in the remaining 50 houses, the pair are split between the two rooms. Within each group of 50 placed in the same room, we have split them into two groups. In the one case, both pictures are placed on an inside wall and in the other, the pair are split between inside wall and outside wall. For the 50 split between two rooms, half have both pictures on inside walls and half have both on outside walls. The pictures are being placed for 18 months. With each picture, there is at least one etched track detector. For 50 individual pictures chosen at random, there are 2 detectors as a QA check. We also have storage blanks and spiked samples.

In February 1993, we began the process of retrieving the exposed pictures. Upon return to the laboratory, each picture is individually wrapped in aluminum foil to prevent any inadvertent further exposure in the laboratory. After an appropriate time to permit the ^{210}Po to come into secular equilibrium with the ^{210}Pb , we will count the activity in the glass with pulse ionization chambers for a sufficiently length of time to provide better than 10% counting statistics. In the meantime, the alpha track detectors are sent to the vendor to be counted. This study will give us a much tighter handle on the utility of this method for long term exposure assessment and therefore much improved epidemiology. However, several more years will be needed to complete this phase of our studies.

Hygroscopic Growth of Particles

The final task of this portion of the project was the development of the tandem differential mobility analyzer (TDMA) system to measure the hygroscopic growth of the indoor aerosol when it encounters > 99% relative humidity within the respiratory tract. The system has been developed that does permit the measurements to be made (Li *et al.*, 1992). The system has been used to perform a series of measurements on several types of indoor particle sources. Growth studies have been conducted on particles present in cigarette smoke (mainstream and sidestream), incense smoke, and produced by candles, natural gas and propane flames. The particles from cigarette smoke are introduced to first DMA right after they are generated from the burning tip. The aerosols from natural gas and propane gas flame are generated in a 55 gallon steel barrel and then introduced into the first DMA. The average residence time of aerosols in the barrel is about 4 minutes. The

residence time of all aerosols in the humidifier is about 20 seconds, approximately equal to the residence time of aerosols in human's respiratory tract. These results were presented by Li and Hopke (1993). Additional studies of consumer products including hair spray, aerosol cleaning products, and pesticides have been performed and the results of these studies was presented in Li's Ph.D. thesis (1993), at the Indoor Air '93 conference in Helsinki in July 1993 (Hopke *et al.*, 1993b) and a manuscript is being prepared for submission to *Aerosol Sci. Technol.*

FUTURE RESEARCH PLANS

The thermal diffusion cloud chamber is operating satisfactorily and measurements are in progress. We will now begin a series of studies on ion-induced binary nucleation. We will again start with a variety of hydrocarbon compounds in He as the support gas. Ultimately, we plan to look at the combined oxidation and nucleation mechanism to study the binary nucleation of water and sulfuric acid with *in-situ* production of the H_2SO_4 through radon radiolysis. In such a system, the gas phase will consist of He, O_2 , and SO_2 . The initial task in this part of the study has been to extend the computer model of the chamber to permit calculation of heat and mass transfer for 2 condensable materials (H_2O and H_2SO_4) and up to 3 gas-phase components. We will need to set the gaseous species concentrations such that there a negative density gradient still exists from the bottom to the top of the chamber.

A major focus of this next year will be studies of the nature of the polonium species immediately following their formation by ^{222}Rn decay. The combination of the circular electrical mobility spectrometer along with the hypersonic impactor should permit us to estimate the molecular weights of the species as a function of the atmospheric composition so that we will have a much better understanding of the chemistry of polonium in the indoor atmosphere. There is an initially important study of ^{218}Po in an O_2 atmosphere. Prior studies (Frey *et al.*, 1981; Goldstein and Hopke, 1985; Chu and Hopke, 1988; Cheng *et al.*, 1994) have clearly demonstrated that there is significantly different neutralization behavior when the $^{218}\text{Po}^+$ is formed in N_2 compared with O_2 . However, recently unpublished studies of Castleman (private communication, 1993) have found no reaction between Pb^+ or Bi^+ with O_2 even with center of mass energies of 75 eV. Thus, identification of the nature of the ionic species formed following the decay of the radon precursor is important to understanding its subsequent chemistry. This chemistry greatly affects the diffusion coefficient of the polonium species and hence its deposition efficiency in the extrathoracic region of the respiratory

tract. It will also provide a much better understanding of the effects of relative humidity or indoor VOCs on the polonium ion cluster size.

In our field studies, we have tested the first pulse ionization chamber. We have acquired the counting electronics to allow us to build 6 additional units so that we can efficiently count the glass when the ^{210}Po has adequately grown into secular equilibrium with the ^{210}Pb . The initial counts on our collected glass will begin in February 1994 and will take about a year to complete. There are some additional pieces of glass to be retrieved during the early part of 1994 and these pieces will be the last to be counted in the spring of 1995.

We will continue to find houses in which we can make exposure measurements. It appears that we will have additional houses to study during the next year in which we will measure activity size distributions in a series of normally occupied homes. The house we are planning to study first is one that is strongly affected by the presence of a high concentration of radon in water. The background level in the house is around 50 to 70 Bq m^{-3} . However, shortly after a shower is run, up to 2000 Bq m^{-3} has been observed in the bathroom. The transport of radon and progeny to the rest of the house and its impact on exposure and dose will be assessed. We continue to seek a mixture of various housing conditions (oil versus gas heating, hot water versus forced air, gas versus electric stoves, absence or presence of wood stove or fireplaces, use of space heaters (electric or kerosene), presence or absence of electrostatic air cleaners and/or humidifier on forced air systems, and effects of central air conditioning). In addition to the activity size distributions, we also plan to measure particle number distributions with the DMA and we hope to be able to measure deposition flux so that the room surface-to-volume ratio can be inferred. These additional measurements are by nature grab measurements, but they can provide the data to estimate the room model parameters necessary to better understand the dynamics of the radon progeny in the house. Further studies in cooperation with LBL and/or CEA may be undertaken to test the model as it is developed.

We are continuing to use the TDMA system to measure the hygroscopic growth of particles from a number of specific sources. The system is not readily portable. The need for several water baths to produce the temperature-controlled water for the wet-film reactor and for the bubblers used to temperature condition and humidify the sheath air for the second DMA means we will have to study aerosols in the laboratory and a very limited range of houses. We want to make additional laboratory studies of sources like a kerosene heater, and electric motors similar to those found in vacuum cleaners. We also plan to examine additional aerosol-spray products including spray

deodorant, FAM spray-on oil, household deodorizer (such as Lysol or Glade air freshener), and additional pesticide sprays.

We will move the system to Professor Hopke's home for additional studies of a free standing wood stove (Vermont Casting Model "Resolute"), an open fireplace, an electric broiler cooking meat (Jenn-Aire broiler), a deep-fat fryer, and background aerosol at several times during the year. We will also examine the particles in the presence of a number of people in the room to examine people as an aerosol source.

SUMMARY

We made continuing progress based on our previous efforts. All of the experimental systems are now on-line, and we are ready to begin collection of substantial new data characterizing the behavior of the radon progeny both in the laboratory and in the real world.

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CURRENT PROJECT PERSONNEL

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Other Professionals

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Graduate Research Assistants

Huiling Ding, Research Assistant, July 1, 1990 - August 31, 1993

Wei Li, Research Assistant, July 1, 1990 - July 31, 1993

Feng He, Research Assistant, August 15, 1991 - present.

O.O. Olawoyin, Research Assistant, July 1, 1993 - present.

Lin Fei, Research Assistant, August 16, 1993 - present.

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