CLARKSON UNIVERSITY

**THE DETECTION AND MEASUREMENT OF THE ELECTRICAL
MOBILITY SIZE DISTRIBUTIONS ASSOCIATED WITH RADON
DECAY PRODUCTS**

A Report

by

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ABSTRACT

THE DETECTION AND MEASUREMENT OF THE ELECTRICAL MOBILITY SIZE DISTRIBUTIONS ASSOCIATED WITH RADON DECAY PRODUCTS

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Clarkson University, 1996

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It has been known for a long time that exposure to radon and its decay products causes an increased risk of lung cancer. Therefore, the infiltration of radon into the indoor environment may represent a significant threat to public health. This potential threat has evoked extensive interest over the past several years in the fundamental chemical and physical properties of the radon decay products. Early studies showed that there are two distinct physical form of the radon progeny: either attached to the ambient aerosols, or still in the status of ions, molecules or eventually small clusters. The "unattached" activity would give a higher dose per unit of airborne activity than would the activity associated with the "attached" decay products that are rather poorly deposited.

In this research, a system for the determination of the unattached radon decay products electrical mobility size distribution by measuring their electrical mobilities has been developed. This measurement was based on the fact that about 88% of ^{218}Po atoms have unit charge at the end of their recoil after decay from ^{222}Rn , while the remainder are neutral. The essential part of the experimental setup is the radon-aerosol chamber with the Circular Electrical Mobility Spectrometer (C. E. M. S.) located inside. The C. E. M. S. Is used for sampling and classifying the charged radioactive clusters produced in

the chamber. An alpha sensitive plastic, CR-39 disk, was placed in the C. E. M. S., and was used both as an inlaid disk electrode and as the alpha particle detector. The system and its classification mechanism are described in detail in various chapters.

The C. E. M. S showed good performance in the fine inactive particles' classification. If it also works well for the radon decay products, it can offer a very convenient size distribution measurement for those radioactive ultrafine particles. However, current experiments did not obtain an acceptable resolution. Discussion and suggestions are made for this problem. If a good resolution can be obtained, the C. E. M. S. would be able to monitor changes in the size distributions as a function of environmental variations. The resultant information should permit a better estimation of the potential human health hazard from the inhalation of radon decay products present in indoor air.

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CHAPTER 1. INTRODUCTION

Radon-222 is one of the decay products belong to uranium-238 decay series. The principal basis for present concern about exposures to radon's decay products is the experience with lung cancer incidence among underground miners. In 1879, Härting and Hesse (1879) diagnosed a sharply higher lung cancer rate among miners. About 45 years later, Ludewing and Lorenzer (1924) suggested that the high lung cancer incidence among the Schneeberg miners may be attributed to the high radon concentration of 10^3 to 5×10^4 Bq m⁻³. It took another 30 years (Shapiro, 1954; Aurand et al., 1955; Bale and Shapiro, 1956) to recognize that the true cause for the high lung dose is not the radon, but the inhalation of the short-lived radon decay products. Until the late 1970s, the radon problem was thought to be largely associated with special industrial processes - primarily the mining and milling of uranium and with building materials (UNSCEAR, 1977). With the discovery of very high level of radon in ordinary living houses in Reading Prong area of eastern Pennsylvania, New Jersey, New York, as well as in Canada and Sweden in the late 70s and early 80s, attention has focused on radon in ordinary homes: its sources, its properties, its health implications, and how it might be controlled.

A principal characteristic of radon that gives it more radiological significance than earlier members of the decay chain is the fact that it is a noble gas. Once this chemically inactive element atom is formed, it is free to move through material's pore space, and a microscopic transport is possible. Radon can, therefore, reach air or water to which humans have access, provided that transport is sufficiently rapid to be completed before the radon decays. ²²²Rn has a relative long half-life, 3.8 days, and no chemical reaction

will occur until it decays further.

These characteristics allow radon formed either in building materials or in the ground within approximately a meter of building understructures can reach the indoor environment.

The second important characteristic of radon is that it decays to radionuclides that are chemically active and relatively short-lived. The decay chain from ^{226}Ra to ^{206}Pb is shown in Figure 1.1. From this figure, it is found that each of the first four radon decay products, polonium-218 (RaA), lead-214 (RaB), bismuth-214 (RaC), and polonium-214 (RaC'), has a half-life of less than 30 min. ^{210}Pb is the effective end decay product of the radon series. With a half-life of 22.3 years, its low specific activity effectively terminates the radon decay chain for most practical human exposure considerations. Therefore, if deposited in the lung after being inhaled, the radon decay products are likely to decay to ^{210}Pb before removal by lung clearance mechanisms.

The radiation released by the decay of the short-lived progeny imparts the dose to the cells at risk in the bronchiol epithelium to which the increased risk of lung cancer is attributed. The alpha radiation from the polonium isotopes contributes the radiologically significant dose, primarily because alpha particles deposit their energy within such a small thickness from the surface of tissue.

From the standpoint of respiratory deposition, the respiratory system can be divided into three regions: the extrathoracic region, the tracheobronchial region, and alveolar region. Inhaled particles deposit in the various regions of the respiratory system by the complex action of three most important mechanisms: impaction, sedimentation, and diffusion, which are governed by inhaled particle sizes. Once deposited, particles are retained in the lung for varying times depending on their chemical properties, their

Figure 1.1 The schematic diagram of uranium-238 decay series

location within the lung, and the type of clearance mechanism involved. The radon progeny damage to people's health is depended on the radiation dose imparting to the tissues and their distribution within the regions of respiratory tract. Therefore, the health effects of the radon decay products are strongly dependent upon their aerodynamic behavior in the indoor atmosphere. However, the behavior is complicated by their chemical activity: the decay products can attach to airborne particles, to indoor surfaces, and to human respiratory, where they can deposit either directly or after attachment to airborne particles. Figure 1.2 illustrates the various mechanisms for changing the state or presence of the decay product, other than radioactive decay itself.

Figure 1.2 Sketch of the behavior of radon decay product in indoor air

Early studies commonly assumed that there are two distinct physical forms of the

radon progeny: either attached to the ambient aerosols, or still in the status of ions, molecules or eventually small clusters. This assumption is based on the experimental observation that an remarkable distinction exists between the diffusivities of both forms: of the order of 10^{-6} to 10^{-4} $\text{cm}^2 \text{s}^{-1}$ for the attached fraction while values of the order of 0.01 to 0.1 for the unattached fraction (Raes 1985). Chamberlain and Dyson (1956) first identified that the highly diffusional size of radon progeny would more effectively deposit in the tracheo-bronchial region of the respiratory system. This "unattached" activity would give a higher dose per unit of airborne activity than would the activity associated with the "attached" decay products that are rather poorly deposited. For this reason, a more accurate understanding of the nature of these unattached radioactive clusters is required.

As ^{222}Rn decays, an α particle is emitted and the residual ^{218}Po nucleus recoils in the opposite direction. The initial kinetic energies of α particle and ^{218}Po nucleus are 119 keV and 5.49 MeV, respectively. Both of them lose their energies by the end of their path, and the energy loss rate for ^{218}Po nucleus is four times that of the α particle. Thus, substantial local molecular excitation and ionization occur.

As the α particle leaves the ^{222}Rn atom, it will autoionize the residual ^{218}Po . As it recoils, its velocity will be larger than the velocity of the bound, outer-shell electrons, and thus will become a highly charged ion. As it slows, it picks electrons back up so that it is almost neutral by the time it achieves thermal velocity and thus 88% are singly charged positive ions and 12% are neutral (Wellisch, 1913; Porstendörfer and Mercer, 1979; Chu and Hopke, 1985). The ^{218}Po ions may lose their charges by electron transfer, electron scavenging, and small ion recombination (Frey *et al.* 1981, Chu and Hopke 1988). Different fractions of $^{218}\text{Po}^+$ neutralization will lead to different apparent molecular diffusion coefficients, since the diffusion coefficient of the ion is less than half that of the

neutral species (Porstendörfer 1968, Porstendörfer and Mercer 1979). Thus, ionic charge of these decay products is one of the parameters controlling plateout.

Many previous studies have examined radon decay product's behavior by the detection of its diffusion coefficient. Raabe (1968) observed that the diffusion coefficient of ^{218}Po decreased with increasing humidity. Raghunath and Kotrappa (1979) studied the effect of relative humidity and ventilation rate on diffusion coefficients of radon and thoron and found that the diffusion coefficient decreased as the residence time of the radionuclide in the diffusion chamber increased. Busigin et al (1980) suggested that it is likely that charged Po-218 in air reacts with oxygen to form polonium dioxide ions, and estimated that its ionization potential is about 10 eV. Goldstein and Hopke (1985) further determined that the ionization potential should be in the range 10.35-10.53 eV. This determination was based on the following principle: The diffusion coefficient changes with particle physical diameter and is affected by particle charge, so that, the growth and neutralization of radon decay products can be detected by their diffusion coefficient measurement. The chemical and physical behavior of radon decay products in suspended gas can be understood by the analysis of this parameter, therefore. Several instrument have been developed and corresponding methods are evolved in diffusion coefficient detection, such as diffusion tube method and wire screen method. However, it is difficult for this kind measurement to offer more detail information, since obtained diffusion coefficient is an average value. This invokes an searching for another techniques which can provide more accurate measurements.

The property that 88% of the polonium clusters carry unit positive charge at the end of the recoil path permits a better understanding of their behavior by the investigation of the electrical mobility of polonium clusters. Polonium is a chemically

active decay product of radon and can react with substances in the suspended gas, or attach onto the existing aerosol particles. Both of these processes will alter the size distribution of this radon decay product clusters, as well as the specific charge of the clusters. As a result, their electrical mobility will be changed. The purpose of this research is to develop and test a system, by which, the electrical mobility spectrum of polonium clusters can be obtained. The electrical mobility data can be converted to the size distribution. By carefully controlling the situation, the behavior of the radon decay products can be well understood. To reach this goal, the Circular Electrical Mobility Spectrometer (C. E. M. S.) is used for separating particle according to their electrical mobility. CR-39, an alpha sensitive plastic, was used to collect the particles and, later, removed and processed for detection.

In Chapter 2, several charging mechanisms are introduced. Then, the motion of charged particles in external electric field is described. Once the particle begin to move, it will experience the resistance to forward movement immediately. If the particle moves with the Reynolds number smaller than one, the relationship between the resistance and movement velocity can be given by Stokes's law. In a quite short time, the resistance will equal to the electrostatic force, the particle reaches its terminal velocity.

Chapter 3 is a review of several kinds of electrical mobility analyzers that are used for the classification and analysis of charged aerosol particles. Their principles for electrical mobility detection are introduced. The advantage and limitation of these instruments are also mentioned.

In Chapter 4, an experiment system which is employed for the electrical mobility measurement of radon decay products are described. The key component of the system is Circular Electrical Mobility Spectrometer, which classifies the decay products according to

their electrical mobility. The classified particles radially deposit on the CR-39 plate. After etching, the spectrum of their deposition can to be observed.

Experimental work performed in a laboratory radon chamber is related in Chapter 5. Experiment results and some problems encountered during this period are also discussed.

Chapter 6 concludes the thesis. Problem resolutions and further experiments are suggested.

CHAPTER 2. ESSENTIAL CONCEPTS AND THEORIES IN ELECTRICAL TECHNIQUES

2.1 Introduction

The importance of electrostatic charge to aerosol mechanics rests on two basic facts. First, a majority of aerosol particles carry some electric charge, and some may be highly charged. Second, the force exerted on small electrically charged particle in an electric field is many times that of gravity. Under well-controlled conditions, this electrical force can be used to design many important types of aerosol control equipment, and aerosol sampling and measuring instruments.

Aerosol charge is of importance to the study of many natural phenomena as well as to the many aspects of aerosol technology and research. The high electrical mobility of charged aerosols in an electric field had been used since about 1900 (White, 1963) for industrial gas cleaning. There are several mechanisms by which aerosol particles can acquire charge: static electrification, charging by small ions (diffusion charging and field charging), thermionic charging, and self-charging.

Static electrification causes particles to become charged as they are separated from bulk material or other surfaces. Thus, particles are usually charged by this mechanism during formation, resuspension, or high-velocity transport. In general, the particle charge acquired by static electrification is difficult to predict and in most cases is undesirable.

Charging by small ions requires the production of either bipolar or unipolar ions and the particles become charged due to collisions between the ions and the particles.

This mechanism is often used to produce aerosols with certain charge distribution. Thermionic charging of particles is primarily caused by thermionic emission of either electrons or ions when particles are heated to a sufficiently high temperature (Yeh and Cheng 1980). For a radioactive aerosol, particles may gain charges due to a self-charging mechanism by the loss of either alpha or beta particles and the ejection of valence electrons, or the release of charged fragments from atoms of particles during alpha, beta, or gamma radiation (Yeh *et al.* 1976; Yeh *et al.* 1978).

When particles are charged by small ions, or they are in equilibrium charge state, the charge distribution is known. In this situation, the particle size distribution can be obtained by the detection of particle behavior in electric field. Just after α -decay of radon, 88% of the decay products form ultrafine unit-charged clusters. The study of their electrical behavior provides a powerful method to understand their dynamic characteristics.

2.2 Charging by Unipolar Ions

When aerosol particles are exposed to unipolar ions, the particles become charged due to collision of the particles and the ions. The complete charging theory, valid for all particle sizes and all charging conditions, is not yet fully known. Various limiting forms of the theory, valid under certain idealized conditions, have been derived by various investigators. These theoretical equations are useful in predicting the charge on particles provided, of course, the particles are charged under such conditions as to meet the requirements set forth in deriving the theoretical equations.

2.2.1 Diffusion Charging

The charging of aerosol particles in the absence of an applied electric field may be considered as a diffusion process in which the gaseous ions diffuse toward the particle surface and impart their electric charges to the particle. Many theoretical and experimental strides on diffusion charging have been reported (Bricard 1948; Fuchs 1947; Liu and Pui 1977; Marlow and Brock 1975; Pui *et al.* 1988; White 1951). To estimate the order the magnitude of the number of charges, n , acquired by a particle of diameter d_p , White's (1951) equation can be used:

$$n = \frac{d_p k T}{2 e^2} \ln \left(1 + \frac{d_p \bar{c} \pi e^2 N_i t}{2 k T} \right) \quad (2.1)$$

where

k = Boltzmann's constant (1.38×10^{-16} dyn cm/K)

T = absolute temperature (K)

e = elementary unit of charge (4.8×10^{-10} statC)

$\bar{c}$ = mean thermal speed of the ions ($\approx 2.4 \times 10^4$ cm/s)

N_i = ion concentration (ions/cm³)

t = charging time (s)

2.2.2 Field Charging

With an applied electric field of sufficient intensity, the ordered motion of the ions caused by the electric field may be the predominant charging mechanism by which the ions are driven onto the particle. Assuming that diffusion charging can be neglected, the following field charging equation has been derived (White 1951):

$$n = \left(\frac{3 \epsilon}{\epsilon + 2} \right) \left(E \frac{d_p^2}{4 e} \right) \left(\frac{\pi e Z_i N_i t}{1 + \pi e Z_i N_i t} \right) = n_s \left(\frac{\pi e Z_i N_i t}{1 + \pi e Z_i N_i t} \right) \quad (2.2)$$

where

E = applied electric field (statV / cm)

ϵ = dielectric constant of particle

Z_i = mobility of ions ($\approx 450 \text{ cm}^2 / \text{statV s}$)

n_s = saturation charge (reached when sufficient charging time is applied).

When particles are charged under moderate electrical field intensities, both the field and diffusion mechanisms of particle charging should be considered. Cochet (1961) derived the following equation, which agreed with his experimental data:

$$n = \left(1 + K \frac{n_i}{2} \right)^2 + \frac{2}{1 + K n_i} \left(\frac{\epsilon - 1}{\epsilon + 2} \right) \left(E \frac{d_p^2}{4 e} \right) \left(\frac{\pi e Z_i N_i t}{1 + \pi e Z_i N_i t} \right) \quad (2.3)$$

Sato (1987) suggested that the sum of the charges calculated using the classical diffusional and field theories independently (equations 2.1 and 2.2) agreed best with his experimental data.

2.2.3 Charging Mechanism of ^{218}Po Clusters

The charge distribution of ^{218}Po clusters at the end of their recoil path is the result of self-charging, electron stripping by air molecules, and neutralization by electron exchange from the support gas molecules. As the decay product of ^{222}Rn , ^{218}Po will automatically have two positive charges and obtain initial kinetic energy. The moving ^{218}Po ions are knocked by the ambient gas molecules and outer shell electrons are

stripped. The negative ions, therefore, form ions cloud around the ^{218}Po ion track. As the ion slows, it will extract electrons from the gas molecules and yield singly charged ions 88% of the time. This high percentage of charged Po molecules has led a number of investigations using the collection of the ^{218}Po in an electrostatic field as a measure of the ^{222}Rn content of the air.

2.3 Charge Neutralization

When aerosol particles are exposed to bipolar ion atmospheres, after a sufficiently long time, the particles will come to a state of charge equilibrium with the ionic atmosphere due to frequent collisions between the particles and ions (Willeke and Baron 1992, p.414). This equilibrium charge state is called the Boltzmann equilibrium or residual charge distribution, and can be written as

$$f_n = \frac{\exp(-n^2 / 2 \sigma^2)}{\sum \exp(-n^2 / 2 \sigma^2)} \quad (2.4)$$

where

$$\sigma^2 = \frac{d_p k T}{2 e^2} \quad (2.5)$$

and f_n is the fraction of particles of size d_p having n elementary units of charge. For particles larger than $0.05 \mu\text{m}$ diameter, Eq. 2.4 can be approximated by the normal Gaussian distribution:

$$f_n = \frac{\exp(-n^2 / 2 \sigma^2)}{\sqrt{2 \pi \sigma^2}} \quad (2.6)$$

However, a study by Reischl *et al.* (1983) indicated that for particle diameters less than $0.05 \mu\text{m}$. Boltzmann's law underestimates the charged fraction, and Fuchs' (1963) theory should be used.

The equilibration time can be greatly shortened by increasing bipolar ion concentration. Either a radioactive source or a bipolar ion generator can be used to approach it. ^{85}Kr is the most commonly used radioactive source. As a rule of thumb, to adequately discharge aerosols to near-Boltzmann equilibrium, the $N_i t$ product should be of the order of 6×10^6 ions/cm³s. The ion concentration, N_i , is dependent on the strength of the ion source used, whereas the charging time t can be adjusted by changing the flow rate through the discharger (Willeke and Baron 1992, p.416).

2.4 Behavior of Charged Particles

2.4.1 Interaction Between Two Charged Particles

The force between two point charges is governed by Coulomb's Law:

$$F = \frac{n_1 n_2 e^2}{r^2} \quad (2.7)$$

Where

n_i ($i=1,2$) = the amount of unit charges carried by particle i

r = distance between the two particles (cm).

In addition to the Coulombic force, a charged particle and a uncharged particle can attract each other by image forces.

2.4.2 Behavior of a Charged Particle in External Electric Field

2.4.2.1 The Motion of an Aerosol Particle in Suspended Gas

When a particle moves through the medium in which it is suspended, the motion results in the resistance from the suspended gas which is defined as the gas resistance force or drag force F_D , and can be expressed as

$$F_D = C_D \frac{\pi}{8} \rho_g d_p^2 V^2 \quad (2.8)$$

where

C_D = coefficient of drag

ρ_g = density of suspended gas

d_p = particle diameter

V = particle motion velocity

Because of the low velocities and small particle sizes involved, most particle motion occurs at low Reynolds numbers. Thus, Stokes' law has wide application to the study of aerosols. In this situation, the drag force can be given as

$$F_D = \frac{3 \pi \eta V d}{C_c} \quad Re < 1 \quad (2.9)$$

Where η is the viscosity of the gas, C_c is the Cunningham slip correction factor derived by Cunningham (1910) as,

$$C_c = 1 + 2.52 \lambda / d_p \quad (2.10)$$

where λ is the mean free path of the gas. Use of the Cunningham correction factor

extends the region of application of Stokes's law to particles of 0.1- μm diameter.

Experimental measurements of slip, made by measuring the settling velocity of particles of known size and density, have provided data necessary to develop an empirical equation that extends the range of Stokes' law to below 0.01 μm .

$$C_c = 1 + \frac{\lambda}{d_p} [2.514 + 0.8 \exp (-0.55 d_p / \lambda)] \quad (2.11)$$

This expression is referred to as the slip correction factor (Hinds 1982). However, equation (2.11) overestimates the slip effect on the molecular clusters in the 0.5-1.75 nm size range. In order to use equation (2.11) to calculate the slip correction factor of particles with diameter larger than 0.5 nm, a semi-empirical factor can be used to fit equation (2.11). This work has been reported by Ramamurthi and Hopke (1989). The substitution of

$$d_p^* = d_p (1 + 3 e^{-2.20 \times 10^7 d_p}), \text{ where } d_p \text{ is in cm,} \quad (2.12)$$

for d_p in equation (2.11) for allows the use of equation (2.12) to estimate the slip correction, C_c .

2.4.2.2 Behavior of a Charged Particle in Electric Field

When an aerosol particle carrying n elementary units of electrical charge is placed in an electric field of intensity E , it experiences an electrostatic force of

$$F_{elec} = n e E \quad (2.13)$$

which causes the particle move through the medium and quickly reaches its terminal

velocity, a condition where the drag force of the air on the particle will be exactly equal and opposite to the electrostatic force. At this condition,

$$F_D = F_{elec} \quad (2.14)$$

$$v = \frac{n_p e E}{3 \pi \eta d_p C_c} \quad Re < 1 \quad (2.15)$$

where v is the terminal velocity. Equation (2.15) can be written in terms of particle mechanical mobility, B , as

$$v = neEB \quad (2.16)$$

Equation (2.15) shows that the terminal velocity is proportional to the intensity of the applied electric field. The proportionality constant is, by definition, the electrical mobility, Z , i.e.

$$Z = \frac{v}{E} = \frac{neC_c}{3\pi\eta d_p} = neB \quad (2.17)$$

2.5 Conclusions

Mechanisms for particles charging have been introduced in this chapter. Sooner or later, highly charged particles will lose most of their charges and come to a charge equilibrium state. The motion in a uniform electric field of charged particles in suspended gas can be described by applying force balance between the electrostatic force and the gas resistance force. It is easy to understand the importance of the charge to aerosol particles' behavior, especially in a strong external electric field. Many kinds of

instruments have been produced for aerosol charge distribution measurement. If the aerosol distribution is previously known, those instruments can be used to determine aerosol particle size distribution. This process is going to be introduced in detail in the next chapter.

CHAPTER 3. CURRENT ELECTRIC MOBILITY ANALYZERS

3.1 Introduction

Instruments used for measuring electrical mobility include the Millikan Cell (Millikan, 1935; Sano *et al.*, 1953; Ivanov *et al.*, 1974; Arnold, 1979), parallel-plate electrical mobility spectrometer (Megaw and Wells, 1969; Maltoni *et al.*, 1973; Takahashi and Kudo, 1973; Yeh *et al.*, 1976; Porstendörfer *et al.*, 1984), and concentric electrical mobility analyzer (e.g., electrical aerosol analyzer and differential-mobility analyzer) (Whitby and Clark, 1966; Knutson and Whitby, 1975b). The Millikan cell method is based on a single-particle measurement; as such, it is tedious and time-consuming for charge or size distribution measurements.

Electrical Mobility Analyzers also can be cataloged into integral-mobility analyzer and differential-mobility analyzer (DMA). In the integral-mobility analyzer, particles with a sufficiently high electrical mobility are collected, while those with lower mobilities penetrate through the mobility analyzer and are sensed by an aerosol sensor (e.g., electrometer or particle counter). The mean electrical mobility, Z_p , of the threshold can be calculated. This kind of instrument can classify particles into two fractions, and can be used to obtain particles with higher or lower electric mobilities than the threshold.

In differential-mobility analyzer, only particles with specified electrical mobility will pass through an aerosol outlet, and be counted. Particles with higher electrical mobilities will be deposited onto the electrode before they reach the outlet. Particles with lower electrical mobilities pass by the outlet, and are collected by a filter or exit with the excess gas flow. Therefore, differential-mobility analyzer can be used to determine

the electrical mobility distribution of the aerosol and, thus, its size distribution by varying some parameter (generally, voltage V) and measuring the corresponding particle penetration as the function of this parameter. Also, it is the instrument to be used to produce approximately monodispersed aerosols. This kind of electric mobility analyzer have been widely used in aerosol science.

The circular electrical mobility analyzer is a kind of parallel plate differential-mobility analyzer. It is gaining interest because of its portable size, relatively low cost, and satisfactory resolution for ultrafine particles. Comparison of circular electrical mobility analyzer, TSI standard 3071 DMA (Li and Pui, 1976), a short version of the TSI DMA, and a Hauke 3/150 DMA has been made by Fissan *et al.* (1996). Their evaluation were performed under the condition of $10 \text{ l}\cdot\text{min}^{-1}$ sheath air and $1 \text{ l}\cdot\text{min}^{-1}$ aerosol flow rates and at a flow ratio of 10:1. The experimental results showed that circular electrical mobility analyzer gave the lowest particle loss characteristics and fair resolution for the sub-20 nm particle size range among the four DMAs.

3.2 Parallel-plate Electrical Mobility Spectrometer

Figure 3.1 depicts a schematic diagram of a parallel-plate electrical mobility spectrometer, showing a particle trajectory with respect to flow streamlines within the spectrometer. The parallel plates A and B are separated from each other by a distance of $2h$, and are respectively charged to potentials $+V/2$ and $-V/2$ with respect to ground. The aerosol is drawn into the device as a thin ribbon through a narrow-slot nozzle extending across the spectrometer, midway between the plates. Clean, filtered, and laminar air flow carries the aerosol through the device from the nozzle towards the outlet. As the aerosol and clean air flow through the channel, charged particles move across the clean air under

the electrostatic force. Those particles with electrical mobility higher than the critical value Z_{pc} are deposited onto the plate, others will penetrate the spectrometer. The Z_{pc} can be determined by:

$$Z_{pc} = \frac{2 v h^2}{UL} \quad (3.1)$$

where

v = mean flow velocity

h = half the inter-plate distance

U = is the potential difference between the plates

L = the length from the nozzle to the outlet

Figure 3.1 Schematic diagram of the parallel-plate electrical mobility spectrometer. (Taken from *Aerosol Measurement-Principles, Techniques, and Applications*, page 416, with the permission from Van Nostrand Reinhold).

By varying the voltage U and measuring the corresponding particle penetration, the electrical mobility distribution can be obtained. In this way, the parallel plate electrical mobility spectrometer is used as integrated electrical mobility analyzer.

The particle collecting plate can also be covered by some kind of special particle sensitive material, which can be removed and analyzed after each experiment, so that the mobility distribution of particles can be determined. In this situation, the parallel plate electrical mobility spectrometer is used as differential mobility analyzer. The electrical mobility of the particles that deposit at a distance x from the entrance is given by the formula similar to equation 3.1.

$$Z_p = \frac{2 \nu h^2}{U x} \quad (3.2)$$

This kind instrument was first developed by Erickson (1922).

A variation to the basic design of Fig. 3.1 has been reported by Johnston (1983). In his design, the outlet flow of the electrical mobility spectrometer was split into two equal-flow outlets; thus, both the positively and negatively charged particles can be measured directly by an optical particle counter. This arrangement improves the speed of measurement and data analysis. It has been used of measure the electrostatic charge of laboratory aerosols and for workplace aerosols (Johnston, *et al.*, 1985, 1987).

3.3 Concentric Electrical Mobility Analyzer

Figure 3.2 shows two types of electrical mobility analyzers, similar to the parallel plate electrical mobility spectrometer but with a cylindrical configuration. The aerosol is introduced into the mobility analyzer by particle-free, clean air. An adjustable voltage is

applied to the inner electrode with the outer cylinder grounded. As shown in Fig. 3.2a, particles with a sufficiently high electrical mobility are collected, while those with lower mobilities penetrate the mobility analyzer and are sensed by an aerosol sensor (e.g., electrometer or particle counter). The median precipitation voltage, V , for particles of a uniform mobility Z_p , is given by

$$Z_p = \frac{(Q_{tm} - 0.5 Q_{am}) \ln(r_2/r_1)}{2\pi VL} \quad (3.3)$$

where

Q_{tm} = total flow rate

Q_{am} = aerosol flowrate

r_1 = the inner electrode radii of the analyzer

r_2 = the outer electrode radii of the analyzer

L = is the length of the inner electrode

V = the voltage on the collector rod

Again, when the electrode voltage is varied, the threshold value of Z_p to get to the detector changes. Therefore, the cumulative curve of the total number of particles that have electrical mobilities lower than Z_p as a function of the voltage between electrodes is obtained. If the particles charge distribution is known previously, the corresponding cumulative curve of total particles number against particle size can be obtained. Thus, this kind of design is used as integral-mobility analyzer.

A variation of this instrument yield a design of a differential-mobility analyzer (DMA), as shown in Fig. 3.2b. A DMA was first used by Rohmann (1934), and later again proposed by Chapman (1937, 1938) for determining the mobility of fine particles.

Figure 3.2 Schematic diagram of (a) an Electrical Aerosol Analyzer and (b) a Differential-Mobility Analyzer. (Reprint from *Aerosol Measurement-Principles, Techniques, and Applications*, page 417, with the permission from Van Nostrand Reinhold).

There are two basic designs of differential mobility analyzers. The cylindrical arrangement with two flow inlets and two flow outlets dates back to Hewitt (1957). A more practical version was described by Whitby and Clark (1966). Knutson and co-workers (Knutson and Whitby, 1975a; Knutson and Whitby, 1975b; Knutson, 1976) developed and discussed in detail a design based on that of Hewitt.

The schematic diagram is shown in Figure 3-2b. As shown in the figure, an extraction slit has been cut in the center rod. Therefore, particles with higher electrical mobility will be collected on the upper portion of the rod and those with lower mobility will be carried along with the major outlet flow. Only those particles with a narrow range of electrical mobility will pass through the narrow slit and will be carried out by the minor sampling flow to the aerosol sensor. The mean electrical mobility of the particles, Z_p , extracted through the slit, is given by (Knutson and Whitby 1975a)

$$Z_p = \frac{[Q_t - (Q_s + Q_a)/2] \ln(r_{2m}/r_{1m})}{2\pi V L} \quad (3.4)$$

where

Q_s = sample flow rate

Q_a = aerosol flow rate

Q_t = the total flow in the analyzer.

The DMA has higher resolution than the integral-mobility analyzer due to its design. However, it may require an aerosol sensor with higher sensitivity for detection, because only a narrow mobility fraction is extracted. Generally, in order to measure particle size distributions, particles need to be charged and the charge distribution should be known. This is because, for a given electrical mobility, there can be different particle

sizes with different numbers of electrostatic charges associated with them.

3.4 Radial Differential Mobility Analyzer

The cylindrical DMA has become the standard design and has been widely used for the generation of monodisperse aerosols and for the size classification of polydispersed aerosols. However, the DMA of Knutson and Whitby was designed for the classification and analysis of particles greater than 10 nm in size. Kousaka *et al.* (1986) and Reineking and Porstendorfer (1986) found that the cylindrical DMA is severely limited for measuring ultrafine aerosol particles, i.e., those below 20 nm in diameter. The aerosol is introduced into the column through a narrow annular region. The transmission efficiency drops dramatically below 20 nm due to diffusion losses in that annulus and in flow passages at the bottom of the analyzer column through which the classified aerosol exits the classifier (Kousaka *et al.*, 1986). The long residence time in the classification region allows Brownian diffusion to substantially broaden the instrument transfer function for ultrafine particles.

Both issues have been addressed with modifications to the cylindrical DMA design. More radial design changes made recently yielded more dramatic improvements. Among them, a design based on the flow between parallel circular disks obtained most successful because its short aerosol inlet and analyzer column limited the Brownian diffusion.

A representative of this design—Radial Differential Mobility Analyzer (RDMA) is shown in Figure 3.3, which developed by Zhang, *et al.* (1995). Particle-free sheath air flow enters the RDMA tangentially into a circular channel, and then passes through a ring made of porous material, that evenly distributes the sheath flow and provides a

uniform flow field. The aerosol flow is also introduced tangentially into the inlet channel of the RDMA. The airflows exit through the outlets at the center of the two disks. The classified aerosol is extracted through a port in the electrode opposite the aerosol inlet; the excess air flow is exhausted through the port on the same side as the aerosol entrance.

Figure 3.3 Cross-section of Radial Differential Mobility Analyzer (Taken from Zhang *et al.*, 1995, with the permission from Aerosol Science and Technology).

The primary region of the RDMA for particle classification is formed by the stainless steel housing disk electrode and an inlaid stainless steel disk electrode that is electrically insulated from the rest of the RDMA and is connected to a negative high voltage power supply. The housing of the RDMA is grounded. The resulting electric field in the analyzing region attracts positively charged particles from the aerosol layer and causes them to migrate across the sheath air flow toward the counter electrode as the radial flow carries them towards the center line.

Particles with relatively high mobility deposit on the counter electrode upstream of the sample outlet. Those with relatively low mobility are discharged with the excess flow.

Particles within a narrow range of intermediate mobilities can reach the sampling hole and are extracted with the sample flow.

Similar to the cylindrical DMA talked above, the aerosol entrance slit has finite width and the precise value of R_{in} is uncertain. This is one of the reasons that cause the broadness of the electrical mobility of selected outlet aerosol. As the results, the lowest mobility articles to be transmitted is

$$Z_{pmin} = \frac{Q_{sh} - Q_s}{\pi(R_2^2 - R_1^2)} \frac{b}{V} \quad (3.5)$$

The highest mobility particles is

$$Z_{pmax} = \frac{Q_{sh} + Q_a}{\pi(R_2^2 - R_1^2)} \frac{b}{V} \quad (3.6)$$

So that, the mean value of the electrical mobility of the aerosol out of the exit is

$$Z_p = \frac{2Q_{sh} + Q_a - Q_s}{2\pi(R_2^2 - R_1^2)} \frac{b}{V} \quad (3.7)$$

where

Q_{sh} = sheath flow rate

Q_s = sample flow rate

Q_a = aerosol inlet flow rate

R_2 = the mean radius from the aerosol entrance slit to the axis.

R_1 = the radius of outlet pore

b = the spacing between the electrodes of the RDMA

V = the voltage between the electrodes

In different fields of aerosol research, the particle size may be dramatically different from one another. Unfortunately, there is no way to apply one single design of electrical mobility analyzer with satisfactory results over the whole size spectrum of actual interest, so that selection of a proper design of the analyzer have to be done before each experiment. Two aspects need to be considered for the selection. First, the size range of the aerosol to be detected. Second, the aerosol flow rates to be handled. For small particles, the design is mainly governed by considerations to minimize the effects of diffusion with respect to the losses. For large particles, hence small electrical mobilities, the limited applicable field strength dictates the use of long devices requiring special care to ensure laminar flows in the classification chamber.

CHAPTER 4. DESIGN OF AN AEROSOL CLASSIFICATION SYSTEM BASED ON THEIR ELECTRICAL MOBILITIES

4.1 Introduction

The objective of this research was to develop a particle classification system based on particle electrical mobility. With this system, the electrical mobility diameter of radon decay products can be determined. After the achievement of this purpose, the second step we were going to do was trying to find the relationship between electrical mobility diameter and aerodynamic diameter of radon decay products, so that either of them can be obtained from the others. As mentioned in Chapter 1, the "unattached" decay products play an important role in radon research because they could more effectively deposit in the tracheo-bronchial region of the respiratory system and would give a higher dose per unit of airborne activity than that could be given by "attached" fraction. On the other hand, the "unattached" radon decay products are chemical active. The chemical reactions with ambient compounds may change their aerodynamic behavior significantly, and make their fate more complicated after they are formed. A good understanding of this variation will increase the accurate estimation radon risk to human being and provide a better scientific basis for finding a way to lower the risk. By simulating different situations in experiments, and after carefully analyzing and comparing their different behavior in different situations, we can learn the details about what chemical reactions occurred to those products.

Circular Electrical Mobility Spectrometer (C. E. M. S.), which was first developed by Pourprix, *et al.* (1990), is the essential instrument in this system. Its classification

principle is the same as the Radial Differential Mobility Analyzer, which was discussed in chapter 3. The C. E. M. S. should be a suitable instrument for the measurement of the electrical mobility of radon decay products because:

1. As we mentioned in Chapter 1, about 88% of ^{218}Po atoms have unit charge at the end of their recoil path after decay from ^{222}Rn , while the remainder are neutral. Thus, the electric technique is a good means for the investigation. The simple charge distribution also implies that a simple function can be used to obtain size distribution.

2. The unattached decay products have diameter around 1 nm. As we discussed in last chapter, C. E. M. S. is suitable for the measurement on ultrafine particles as small as 1 nm.

A formula for the mean electrical mobility calculation of the deposited particles, which was deduced by Pourprix, *et al.* (1990) will be introduced in this chapter. It is the starting point of the calibration experiments. The application of this formula is based on the following assumptions: (1) the flow in the C. E. M. S. is laminar, axisymmetric, and incompressible; (2) particle inertia and Brownian motion are neglected; and (3) the electric field is uniform. This chapter is a discussion of the design developed for the C. E. M. S. to meet the above operational specifications.

4.2 Description of the C. E. M. S. Design

The schematic cross-section of the Circular Electrical Mobility Spectrometer (C. E. M. S.) is shown in Figure 4.1. The circular, flat stainless steel top and base form a small chamber, with a circular flat concentric Teflon plate sandwiched in between. All of them are parallel. A stainless steel ring is mounted on the Teflon plate to support a carbon coated or Mylar film covered CR-39 disk. The CR-39 disk can be taken out for

Figure 4.1 The Schematic Cross-section of the Circular Electrical Mobility Spectrometer

the further analysis after sampling and has to be changed after each experiment. The CR-39 plate and the stainless steel ring are electrically conductive, but electrically separated from the rest of the C. E. M. S..

Particle-free air enters the C. E. M. S. through the sheath flow inlet that is located at the center of the stainless steel base, then it spreads radially between the Teflon plate and the inner side of the stainless steel base plate. After passing through a glass fiber filter (Pallflex Inc.) hoop which is used to evenly distribute the sheath flow, and surrounding the Teflon plate, it forms a uniform, laminar, radial flow towards the center line in the gap between CR-39 disk and the stainless steel top where particle collection occurs.

The aerosol flow is introduced into the C. E. M. S. from aerosol inlet slit. The slit is a sharp-edged ring with a 0.4 mm-wide gap that provides for a smooth, turbulence-free merging of aerosol flow. After charged particle's collection, the excess aerosol flow and the sheath airflows exit through the outlet at the center of the stainless steel top. The primary region of the circular electrical mobility spectrometer for particle classification is formed by the stainless steel top disk electrode and the CR-39 disk which was used both as inlaid disk electrode and as particle detector. During the experiments, the housing of the C. E. M. S. was grounded. The CR-39 was connected to a negative voltage power supply. Therefore, a uniform electrostatic field in the analyzing region was formed except in the small region that was very close to the excess flow outlet. Positively charged particles in this region were attracted from the aerosol layer and transported across the sheath air flow toward the inlaid electrode as the radial flow carried them toward the center line. The radial velocity of the flow increases as it approaches the center line. To maintain the laminar radial flow throughout the whole collection region,

we only need to keep the Raynolds number lower than 2,300 in the region near the exit. Therefore, generally, the flow rate should not exceed $32.5 \text{ L}\cdot\text{min}^{-1}$. The collection region for the interesting particles should not be too near to the airflow outlet in case unexpected influences come from either non-uniform electric field, flow turbulence, or both.

As soon as a charged particle comes into the classification region, it will experience electrostatic force and start to move to the counter electrode. In the experiments, the $^{218}\text{Po}^+$ cluster will move towards the CR-39 negative disk electrode. Since the particle is suspended by the gas, it will get accelerated, and reach its terminal velocity. This process takes negligible time.

Carried by the radial sheath air flow, a particle with a electrical mobility higher than the critical value will deposit on the CR-39 plate. The critical electrical mobility is defined here as the value possessed by a particle which is collected by the electrostatic force exactly at the edge of the excess flow outlet of Circular Electrical Mobility Spectrometer. Particles with relative higher mobility deposit at positions nearer the aerosol inlet slit. The deposition position will move towards center line as particles electrical mobility become smaller and smaller. Since the C. E. M. S. is axisymmetric, the deposition of particles with the same electric mobility will form a concentric circle on CR-39 disk with radius of R_2 . The neutral particles or the particles with the electrical mobility smaller than critical value are removed through the excess flow outlet. The analysis of the deposition spectra can give the information of particles electrical mobility distribution.

Figure 4.2 shows how the deposition position of charged particles depends on their electrical mobility.

A formula was given by Pourprix, *et al.* (1990) to calculate the electrical mobility,

$$R_2 = \left(R_1^2 - \frac{Q}{\pi ZE} \right)^{1/2} \quad (4.1)$$

Therefore, for fixed values of Q and E , a single value of R_2 corresponds to each electrical mobility Z .

Figure 4.3 Picture of sphere particles ($0.72 \mu\text{m}$ diameter, carrying one elementary charge) precipitated on a 6" wafer by the C. E. M. S. (From Pourprix, *et al.*, 1990).

The examination and calibration of Circular Electrical Mobility Spectrometer has been done for a particle size of $0.72\ \mu\text{m}$ (Pourprix *et al.*, 1990), and the results fitted the calculated value very well. The deposition ring of radius R_2 was clear and sharp (figure 4.3).

CR-39 is a kind of polycarbonate foil that has been widely used as radon detectors in recent years. It has a relatively stable sensitivity to α rays and low background. During the experiment, ^{218}Po clusters deposited on the CR-39 disk. The decay of these clusters to ^{214}Po release α particles. The kinetic energy of the emitted α particles enabled them to penetrate into CR-39 disk, and leave tracks in it. After the sampling ended, the CR-39 disk was etched by concentrated sodium hydroxide solution until the tracks were large enough to be observed by an optical microscope.

4.3 Conclusions

The essential elements of the design of radon decay product electric mobility diameter determination system have been presented in this chapter. Two points must be emphasized here. First, charged particles are collected by a uniform electric field. To reach this point, the coating or the covering on the CR-39 disk must be very even, and the interesting particles should not be deposited too close to the excess flow exit. Second, the application of Eq. (4.1) requires the aerosol flow rate is small compared with the carrier gas flow rate so that it can be negligible. Another advantage of this small aerosol flow is the avoidance from disturbing the laminar carrier gas flow by its confluence. For ultrafine particles, their inertia motion can be neglected, but the Brownian motion can not be omitted. However, the Brownian broadening can be limited by carefully selecting instrumental parameters such as flow rate and electric voltage. Both the aerosol inlet

width and the Brownian broadening leads to only being able to determine average electric mobility of particles.

In Chapter 5, the prototype sampling, etching, and counting system will be introduced. The principle experimental results will also be presented.

CHAPTER 5. EXPERIMENTAL CALIBRATION AND CHARACTERIZATION

5.1 Introduction

Previous studies have shown that the C. E. M. S. gives good performance for the inactive ultrafine particles, and equation (4.1) provides a good fit to the experiment results (Fissan *et al.*, 1996). The aim of this study was the development of an experimental protocol that employs the C. E. M. S. for the determination of the electrical mobility diameter of radon decay product $^{218}\text{Po}^+$. As mentioned in Chapter 1, 88% of ^{218}Po atoms have a unit positive charge at the end of their recoil, and form clusters with diameters around 1 nm. Two kinds of studies have been conducted:

- 1) Under carefully controlled situation, form ultrafine $^{218}\text{Po}^+$ clusters with well defined diffusion coefficients. Calibrate the C. E. M. S., determine instrumental parameters, such as volumetric flow rate, applied electrical voltage, etc. with these clusters.
- 2) Examine equation 4.1 with the experimental results to see if it is still applicable to such ultrafine particles.

This chapter provides a description of the experimental system utilized to generate these radioactivity clusters including details of the radon-aerosol chamber designed for this purpose and its operation. The sampling, etching, and counting systems are also described.

5.2 Experimental System

The experimental system consists of two distinct units. The first is the radon-aerosol generation system and sampling equipment. This unit includes the radon sources,

commercial tank gases such as N_2 and O_2 , compressed air purification apparatus, the flow control equipment, and the radon-aerosol chamber, in which the C. E. M. S. was located. The second unit included the CR-39 etching, counting and analysis system. Each of these units is described in detail in this section.

5.2.1. Radon-Aerosol Generation and Sampling System

A schematic diagram of the radon-aerosol chamber, including the C. E. M. S., operation controls, and other subsidiary equipments is shown in Figure 5.1. The chamber is made of plexiglas, with a volume of about 200 liters. Two inlet ports are fixed at the left side of the chamber. One of them was for the introduction of radon-laden gas or gas mixture into the chamber, another was used to introduce the dry, clean, aerosol free compressed air into the C. E. M. S. directly.

The radon gas was from a flow-through Pylon Rn-1025 source with continuous output about $1.39 \times 10^5 \text{ MBq} \cdot \text{min}^{-1}$. The source contained a dry radium (^{226}Ra) that provided calibrated quantities of radon gas with an accuracy of $\pm 4\%$. A compressed nitrogen gas or oxygen gas cylinder was used to supply pure nitrogen or oxygen. A three-way joint was used after the gas regulator and adapter to divide the nitrogen or oxygen gas into two branches. One branch went through the flow-through type radon source at a mass flow controlled rate of $100 \text{ cm}^3 \text{ min}^{-1}$, and then into the mixer-manifold. The other branch entered the mixer-manifold directly with a valve located upstream to adjust the pure gas amount in the mixer-manifold. The radon-laden gas and the pure gas then well mixed in the mixer-manifold, and passed through the outlet port of the mixer-manifold and into the plastic chamber. Other trace gases, such as NO , SO_2 , and so on, could also be introduced into radon-aerosol chamber for generating different nanometer sized

Figure 5.1 Schematic diagram of the laboratory radon-aerosol chamber

particles. The effect on ^{218}Po cluster of these trace gases can then be studied. A output port located at the right side of the chamber provided a route for the excess radon-laden gas or gas mixture to an exhaust hood. A metering valve was used to control the leakage rate that allowed the pressure in the chamber to be a little bit higher than the ambient air pressure to prevent the room atmosphere from penetrating into the chamber. A three-way joint was located just before the metering valve to create a route for gas reaching the vacuum valve. Prior to each experiment, the route for cylinder gas to radon source was shut off while the metering valve 1 was opened. Thus, only the pure nitrogen or oxygen passed through the manifold and entered the radon-aerosol chamber. At the same time, the vacuum valve was turned on to remove the resident air in the chamber. The chamber would be full of the cylinder gas after a while. This pre-treatment was for the development of a well controlled situation and to be certain that the aerosol generated in the chamber was free from the influence of unknown component in the air.

The dry, clean, aerosol-free air used as carrier gas in the C. E. M. S. was obtained by following method: let the compressed air passed through silica gel, activated charcoal and 13X molecular sieve columns for the removal of moisture and organic vapors. The air was then filtered through a $0.2\ \mu\text{m}$ pleated membrane filter (Gelman Inc.) for the removal of particulate contamination. Mass flow controllers (Tylan Equipment Co.) were used to accurately measure and maintain the required air flows in the system.

The sampling system involving the use of a combination of positive-pressure air input and negative-pressure air removal was utilized. The compressed air from outlet of the mass flow controller provided the positive-pressure input. An oil-less, carbon-vane centrifugal pump (Doerr Inc.) provide the means for removal of air from the excess flow outlet of the C. E. M. S., that was then vented through an exhaust hood. The outlet air

flow rate was about 1~2 lpm in excess of the fixed input air flow rate to make sure a certain amount of the radon decay clusters generated in the chamber were drawn into the C. E. M. S. from the aerosol inlet slit, and were analyzed inside.

5.2.2 Pre-treatment of CR-39 Disks

In the C. E. M. S., a CR-39 disk was used as the inlaid disk electrode to collect ultrafine radon decay product cluster. Thus, the CR-39 disk must be pre-treated to be electrically conductive before it was employed in each experiment. Two methods were used for this purpose: carbon coating or Mylar covering. Carbon coating can produce a uniform, thin, and very even carbon film, but it is easy to damage the CR-39 disk because of heating during the coating process. For this reason, the later method was selected in the latter experiments.

5.2.3 Etching and Counting System

When the polonium cluster deposits on the CR-39 disk and emits an α -particle, the α -particle will penetrate the disk and leave a track in it. However, this track is too small to be observed by optical microscope, unless it is enlarged by etching. The etching process was not complicated. CR-39 disk was immersed in 6.25 N sodium hydroxide solution for a whole night and the temperature was kept at 72 °C by water bath. After etching, the track was enlarged with a diameter as big as 20 micro-meter in average.

The counting system included a optical microscope (Nikon Co.), a video camera (Sony Co.), a camera adaptor (Sony Co.), a monitor, and a personal computer. The enlarged tracks were magnified by the optical microscope, the picture was captured by the video camera and then translated to specific signals by the adaptor. The signals were

then transferred to the personal computer and recovered to a picture appeared in the monitor. The counting process was carried out using the commercial software package called Mocha. The captured picture only covered a very small area of the CR-39 disk, so several pictures had to be taken along the radius to get the number distribution of the tracks, and then the electrical mobility distribution of the particles can be obtained by applying the equation 4.1.

5.3 The Calibration of the Circular Electrical Mobility Spectrometer

The calibration experiments were proceeded for two main purposes:

- 1) Examine the resolution of the C. E. M. S. under well controlled situations. Good resolution is essential to permit the further experiments regarding the chemical reactions of radon decay product. For example, if the high resolution requirement is reached, it could be determined that whether $^{218}\text{PoO}_x^+$ is formed in the presence of oxygen or not.
- 2) Examine the fitness of equation 4.1 to ultrafine clusters.

5.3.1 Generation of Clusters with Certain Electrical Mobility Size Distribution

^{218}Po clusters with a given electrical mobility size distribution were generated during the calibration and characterization experiment. They were utilized to determine the efficiency of the C. E. M. S. on the ultrafine radioactive clusters.

The $^{218}\text{Po}^+$ clusters were generated in the radon-aerosol chamber by introducing radon into pure nitrogen. As discussed in Chapter 1, 88% of the $^{218}\text{Po}^+$ clusters following radon decay are found to be singly charged while 12% are neutral (Wellish, 1913; Porstendorfer and Mercer, 1979). Calculation and experiments have shown that the electrical mobility of the highest mobility small ions (primary ions) was about $2.0 \text{ cm}^2\text{s}^{-1}$

v^{-1} (Bricard, J., *et al.* 1966; Bricard, J., *et al.* 1965). This value was used to determine the experimental parameters.

5.3.2 Experimental Calibration—Procedure

The experimental procedure utilized for the determination of the plateout characteristics of the C. E. M. S. involved the following steps. Before each experiment, the radon-aerosol chamber was filled with nitrogen. The vacuum valve and the metering valve 1 (figure 5.1) were turned on, other valves and the mass flow controllers were shut off. Both of the flow rates entering and leaving the chamber were about 50 lpm. The volume of the chamber was about 200 liters. If we assume that the pure nitrogen well mixes with the air in the chamber instantly after its entering, 13 minutes later, the mole fraction of nitrogen in the chamber would reach 0.99. Actually, this nitrogen filling procedure was operated for 25 minutes before each experiment.

This step was followed by the sampling procedure. The vacuum valve and metering valve 1 were shut off in this process. The mass flow controllers, the pump, and metering valves 2 and 3 were turned on. The carrier air and the radon-nitrogen gas then entered the C. E. M. S. and radon aerosol chamber, respectively, at given flow rates. During the sampling, the aerosol in the chamber was drawn into the C. E. M. S. through the aerosol inlet slit at a flow rate of 1 to 2 lpm. In order to keep the gas pressure in the chamber a little bit higher than the atmosphere, metering valve 1 could be opened, and the amount of nitrogen entering the chamber was adjusted. The sampling procedure took 5 hours in each experiment. The CR-39 disk was then taken out and immersed in a 72°C, 6.25 N NaOH solution for 10 hours. The tracks of α -particles became large enough, and easy to observe under the optical microscope.

5.3.3 Experimental Calibration – Results

Several experiments have been done for the instrument calibration. Five of them are discussed here. The results are shown as Figures 5.2-5.6. The Y-axis represents the relative number of deposited particles, while the X-axis is the distance from the CR-39 disk center. The deposited track number was counted inward from the CR-39 edge, which is 7.0 cm from the center. Data points in the figures represent the relative, average values of deposit number within concentric, 2 mm wide rings.

Experiment #1. $Q_{in}=18.0$ lpm, $Q_{out}=19.2$ lpm, $U=1.48$ V, $d=0.35$ cm, $R_1=6.5$ cm.

Figure 5.2 Experimental result in the situation of $Q_{in}=18.0$ lpm, $Q_{out}=19.2$ lpm, $U=1.48$ V, $d=0.35$ cm, $R_1=6.5$ cm.

According to equation 4.1, R_2 should be 5.5cm. The result is shown in Figure 5.2. It is found that most particles deposited just under the aerosol entrance slit ($R=6.4$ cm). The deposit number decreased rapidly towards the center ($R=0$), and no other sharp

peak appeared. This peak was not identical to the prediction of equation 4.1. In order to be sure that the peak was caused by the particles precipitated by the Circular Electrical Mobility Spectrometer, and observe that how it move with the change of experimental parameters, the following experiments were performed.

Experiment #2. $Q_{in}=30.5$ lpm, $Q_{out}=32.1$ lpm, $U=1.35$ V, $d=0.35$ cm, $R_1=6.5$ cm.

Figure 5.3 Experimental result in the situation of $Q_{in}=30.5$ lpm, $Q_{out}=32.1$ lpm, $U=1.35$ V, $d=0.35$ cm, $R_1=6.5$ cm.

The higher flow rate and higher voltage shorten the retention time of the clusters in the C. E. M. S., and therefore, should limit the cluster diffusion. A sharp peak other than the one just under the inlet slit was expected by these changes, and it should appear at R_2 equal to 4.5 cm according to equation 4.1. However, the spectrum from the experiment (Figure 5.3) shows no great improvement. Comparing with figure 5.3, a larger

fraction of the deposited particles move inward, but it is still difficult to observe another clearly distinguished peak.

#3. $Q_{in}=30.5$ lpm, $Q_{out}=32.3$ lpm, $U=1.59$ V, $d=0.41$ cm, $R_1=6.5$ cm.

Figure 5.4 Experimental result in the situation of $Q_{in}=30.5$ lpm, $Q_{out}=32.3$ lpm, $U=1.59$ V, $d=0.41$ cm, $R_1=6.5$ cm.

R_2 should be 4.5 cm in this case.

The distance between the two disk electrodes was enlarged to see if there was any change in the spectrum. The result is shown in Figure 5.4. No big improvement was obtained. Two smaller peaks appeared in this spectrum. Their average distance from the center line, R_2 , were 5.1 cm and 4.0 cm respectively. If it is assumed that these two peaks resulted from the deposition of $^{218}\text{Po}^+$ cluster, and equation 4.1 is correct, we can obtain the electric mobilities, and then, the electric mobility diameter of these clusters. The calculation involved equations 2.11, 2.12, and 2.17. The program, MATHCAD, and trial

and error method were used. The diameters of those clusters from calculation are 0.59 nm (for $R_2=5.1$ cm) and 0.84 nm (for $R_2=4.0$ cm). However, those peaks are too short and too broad (both of the half-height widths are greater than 0.5 cm) to be used for further analysis.

#4. $Q_{in}=42.1$ lpm, $Q_{out}=44.0$ lpm, $U=2.71$ V, $d=0.41$ cm, $R_1=6.5$ cm.

Figure 5.5 Experimental result in the situation of $Q_{in}=42.1$ lpm, $Q_{out}=44.0$ lpm, $U=2.71$ V, $d=0.41$ cm, $R_1=6.5$ cm.

R_2 should be 5.0 according to equation 4.1.

In this experiment, the flow rate and the voltage were further increased. The laminar flow can still be obtained in the region for locations beyond 7 mm radius from the center line. These increments were expected to narrow the peak, but they were not helpful as can be seen in Figure 5.5.

#5. $Q_{in}=18.0$ lpm, $Q_{out}=18.9$ lpm, $U=0$ V, $d=0.41$ cm, $R_1=6.5$ cm.

Figure 5.6 Experimental result in the situation of $Q_{in}=18.0$ lpm, $Q_{out}=18.9$ lpm, $U=0$ V, $d=0.41$ cm, $R_{in}=6.5$ cm.

The result of this blank test is shown in Figure 5.6. It is obvious that there are many tracks on the CR-39 disk in spite of the applied voltage being zero. Secondly, many particles moved and deposited inwards, but at the position just under the aerosol inlet slit, the number of deposited tracks was still dominant.

5.4 Conclusions

A description of experimental investigations performed in this study has been presented in this chapter. The design and operation of a radon-aerosol chamber developed for the purpose of generating the radon decay products with certain electrical

mobility size distribution utilized in the study was presented. Etching and counting systems were also introduced.

Experimental results for different sampling conditions were presented. They were characterized by a strong, sharp deposition peak just under aerosol inlet slit. This peak was not moved by changing experimental parameters. Smaller amounts of clusters moved inwards by the carrier air and deposited on the CR-39 disk by static electric field. Their deposition did varied by changing experimental parameters somehow. However, no sharp peaks were formed by this deposition, so that it is very difficult to proceed with the calibration. The discussion on these results will be presented in next chapter.

CHAPTER 6. DISCUSSIONS AND CONCLUSIONS

6.1 Introduction

In chapter 5, the calibration experimental results were described. The analysis and discussion on those results are presented in this chapter, followed by some suggestion for further investigations.

6.2 Discussion

Figure 6.1 Schematic diagram of the particle deposition on the aerosol inlet slit of the C. E. M. S.

When the particles enter the C. E. M. S. through the inlet slit, some of them will deposit on the edge of the slit by diffusion. For clusters with diameter below 1 nm, this plateout becomes very significant due to the high diffusion coefficient. The investigated cluster in the experiment, $^{218}\text{Po}^+$, has a high diffusion coefficient in nitrogen, 0.044 cm^2/sec (Frey, *et al.* 1981).

If the aerosol inlet slit is considered as a parallel plate diffusion battery, the penetration, P , as a function of μ , deposition parameter, can be expressed as (Hinds, 1982),

$$P = 0.910\exp(-7.54\mu) + 0.0531\exp(-85.7\mu) + 0.0153\exp(-249\mu) \quad \text{for } \mu > 0.003 \quad (6.1)$$

$$\mu = \frac{D L W}{Q h} \quad (6.2)$$

where,

h = plate spacing

W = plate width, which is much greater than h

D = particle diffusion coefficient

Q = volume flow rate

L = length of the channel in the direction of flow.

Figure 6.1 is a enlarged schematic diagram of the aerosol inlet slit. In this case, W is equal to 40.8 cm ($2\pi R_1$), h is about 0.2 mm, and L is about 0.2 mm too. If we suppose that Q is 1.5 l/min, therefore, μ is 0.07, P is about 0.6. This means that 40% of the clusters deposit at the edge. Every cluster emit a α -particle. If we further suppose that the α -particle emitted by the cluster at the edge has the same possibility to hit the CR-39 as which emitted from the cluster flying in the instrument, forty percent of the tracks is at the position just under the slit, sixty percent of them spread out between the inlet and the outlet. Moreover, the C. E. M. S. is surrounded by the radon during the experiment. When ^{222}Rn decays to ^{218}Po , α -particles are also emitted. Since the α -particle can move about 4.6 cm in the air, the decay of the ^{222}Rn atoms just above the

aerosol inlet slit can also leave tracks on the CR-39, and make the number increased further. Both of these factors might explain the large peak that appeared at about $R=6.4$ cm in each spectrum.

From Figure 1.1, we can find that not only the decay of $^{218}\text{Po}^+$ atom itself emits α -particle, but also its decay products, like ^{214}Po . In theory, people can distinguish these two kinds of decay by the α -particles' energies. However, it is extremely difficult in this experiment. Also, ^{222}Rn atoms can emit α -particles and leave tracks on CR-39 disk when they move through the C. E. M. S., but this only add a small increment of tracks due to it relatively long decay half-life. This may be a reason for the absence of a sharp peak between the aerosol inlet and the outlet. Another reason is due to the particle diffusion and thus deposition of neutral ^{218}Po will occur across the plate.

6.3 Suggestion

Figure 6.2 Schematic diagram of the detector-filter unit

The high cluster attachment rate to the edge of the inlet slit as they enter the Circular Electrical Mobility Spectrometer could be reduced by increasing the flow rate of the aerosol. However, this procedure is limited because equation 4.1 is based on the assumption that aerosol flow rate should be very small comparing with the carrier gas flow rate, and therefore, can be considered as zero. Alpha spectrometry could be used in place of CR-39 track analysis method. In this way, it is not the particles deposited by the C. E. M. S. are counted, but the particles flowing out of the C. E. M. S.. When radon decay products pass through the C. E. M. S., the clusters with higher enough electrical mobilities are removed by the C. E. M. S., while others are carried to the detector-filter unit, which is shown in Figure 6.2, through the excess flow outlet. Those radioactive particles are then collected by the membrane filter. A planar silicon barrier detector is positioned in the face of the filter, which is used to detect the alpha particles emitted by the radon decay products. The number of particles collected is determined by alpha spectroscopy. The signals of the detector are amplified by a preamplifier-amplifier combination and the spectra are collected with a multichannel analyzer. The multichannel analyzer measures the energy spectrum of a radioactive source by recording the pulse-height distribution produced by particles emitted from the source.

Since the alpha particle emitted by ^{218}Po atom has an energy different from other decays, studies on the ^{218}Po can be made without being disturbed by other radioactive particles. By changing experimental conditions, for example, by increasing the voltage applied on the C. E. M. S., an experimental result as shown in Figure 6.3 is expected. AB is extrapolated and hit the x axis at point C. This point is used to determine the electrical mobility of radon decay products.

Figure 6.3 Deposition curve

6.4 Conclusions

A system used for the electrical mobility determination of radon decay product was designed and established. Calibration experiments were performed. Some problems were encountered and needed to be solved. If it succeeds, the C. E. M. S. can offer a very easy and quick method to study radon decay products' behavior in different situation. The relationship between aerodynamic diameter and electrical mobility diameter can then be determined. The radon risk to human being can be more accurately and more comprehensively assessed.

REFERENCES

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- Albrecht, E. and Kaul, A., 1967. Assessment of Airborne Radioactivity in Nuclear Operations. International Atomic Energy Agency, Vienna.
- Arnold, S., 1979. Determination of particle mass and charge by one electron differentials. *J. Aerosol Sci.* 10:49-53.
- Aurand, K., Jacobi, W. and Schraub, A., 1955. Zur Biologischen Strahlenwirkung Des Radon Und Seiner Folgeprodukte. *Sonderband Zur Strahlentherapie*, 35: 237-243.
- Bale, W. F. and Shapiro, J. V., 1956. Radiation Dosage to Lungs from Radon and its Daughter Products. Proc. First Int Conf. On Peaceful Uses of Atomic Energy, United Nations, New York 13: 233-236.
- Bricard, J., 1948. Sur l'équilibre ionique de la basse atmosphere. *C. R. Acad. Sci. (Paris)* 226:1536-38.
- Bricard, J., Girod, P., and Pradel, J., 1965. Mobility spectrum of small radioactive ions in the air. *C. R. Acad. Sci.* 260:6587-6590.
- Bricard, J., Billard, F., Blanc, D., Cabane, M., and Fontan, J., 1966. Detailed structure of the mobility spectrum of the small radioactive ions in the air. *C. R. Acad. Sci.* 263:761-764.
- Busihin, A., van der Vooren, A. W., Babcock, J. C., Phillips, C. R., 1980. The nature of unattached RaA (^{218}Po) particles. *Health Phys.* 40:333-343.
- Chamberlain, A. C. and Dyson, E. D., 1956. The dose to the trachea and bronchi from the decay products of radon and thoron. *Br. J. Radiol.* 29:317-325.
- Chu, K. D. and Hopke, P. K., 1985. Continuous monitoring method of the neutralizing phenomena of Polonium-218. Pittsburgh, PA: Air Pollution Control Association; APCA Paper No. 85-85.5.
- Chu, K. D. and Hopke, P. K., 1988. Neutralization kinetics for polonium-218. *Environ. Sci. Technol.* 22:711-717.
- Cochet, R., 1961. Colloque Internationale-La Physique des Forces Electrostatiques et Leurs Applications, pp. 331-38. Paris: Centre National de la Recherche Scientifique.
- Cunningham, M. E., 1910. On the velocity of steady fall of spherical particles through fluid medium. *Proc. Roy. Soc.* 83A:376-365.
- Erickson, H. A., 1922. On the nature of the negative and positive ions in air, oxygen and nitrogen. *Phys. Rev.*, 2nd Series, 1, No. 2, p. 117.

Fissan, H., Hummes, D., Stratmann, F., Büscher, P., Neumann, S., Pui, D. Y. H., and Chen, D., 1996. Experimental comparison of four differential mobility analyzers for nanometer aerosol measurements. *Aerosol Sci. Technol.* 24:1-13.

Frey, G., Hopke, P. K., Stukel, J. J., 1981. Effects of trace gases and water vapor on the diffusion coefficient of polonium-218. *Science* 211:480-481.

Fuchs, N. A., 1947. The charges on the particles of aerocolloids. *Investiya Acad. Nauk. SSSR, Ser. Geogr. Geofiz.* 11:341

Fuchs, N. A., 1963. On the stationary charge distribution on aerosol particles in a bipolar ionic atmosphere. *Geofis. Pura Appl.* 56:185-93.

Goldstein, S. D. and Hopke, P. K., 1985. Environmental neutralization of polonium-218. *Environ. Sci. Technol.* 19:146-150.

Härtig, F. H. and Hesse, W., 1879. Der Lungenkrebs, die Bergkrankheit in den Schneeberger Gruben. *Vierteljahresschrift für gerichtliche Medizin Und öffentliches Sanitätswesen* 30: 296-309, 31:, 102-132, 313-337.

Hinds, W. C., 1982. *Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles*, pp. 45, New York: Wiley.

Ivanov, V. D., Kirichenko, V. N., Shan'gin, B. V., and Berezhnoi, V. M., 1974. Method and apparatus for the investigation of the electrical charging of beta-active 'hot' aerosol particles. *Colloid J. USSR* 36:427-30.

Johnston, A. M., 1983. A semi-automatic method for the assessment of electric charge carried by airborne dust. *J. Aerosol Sci.* 14:643-55.

Johnston, A. M., Vincent, J. H., and Jones, A. D., 1985. Measurements of electric charge for workplace aerosols. *Ann. Occup. Hyg.* 29:271-84.

Johnston, A. M., Vincent, J. H., and Jones, A. D., 1987. Electrical charge characteristics of dry aerosols dispersed by a number of laboratory mechanical dispersers. *Aerosol Sci. Technol.* 6:115-27.

Kirsch, A. A. and Zagnit'ko, A. V., 1981. Diffusion charging of submicrometer aerosol particles by unipolar ions. *J. Colloid Interface Sci.* 80:111-17.

Knutson, E. O. and Whitby, K. T., 1975a. Aerosol classification by electric mobility: Apparatus, theory, and applications. *J. Aerosol Sci.* 6:443-51.

Knutson, E. O. and Whitby, K. T., 1975b. Accurate measurement of aerosol electric mobility moments. *J. Aerosol Sci.* 6:453-60.

Knutson, E. O., 1976. Extended electric mobility method for measuring aerosol particle size and concentration. In *Fine Particles*, ed. B. Y. H. Liu, pp. 739-62. New York: Academic

Press.

Kousaka, Y., Okuyama, K., Adachi, M., and Mimura, T., 1986. *J. Chem. Eng. Jpn.* 19:401-407.

Liu, B. Y. H. and Pui, D. Y. H., 1977. On unipolar diffusion charging of aerosols in the continuum regime. *J. Colloid Interface Sci.* 58:142-49.

Ludewing, P. and Lorenzer, S., 1924. Untersuchungen in der Grubenluft in den Schneeberger Gruben auf den Gehalt an Radium-Emanation. *Z. Phys.* 22, 178.

Maltoni, G. G., Melandri, C. Prodi, V., Tarroni, G., De Zaiacomo, A., Bompane, G. F., and Formignani, M., 1973. An improved parallel plate mobility analyzer for aerosol particles. *J. Aerosol Sci.* 4:447-55.

Marlow, W. H. and J. R. Brock., 1975. Unipolar charging of small aerosol particles. *J. Colloid Interface Sci.* 50:32-8

Megaw, W. J. and Wells, A. C., 1969. A high resolution charge and mobility spectrometer for radioactive submicrometre aerosols. *J. Sci. Instrum. (J. Phys. E.) Series 2* 2:1013-16.

Millikan, R. A., 1935. Electrons (+ and -), protons, photons, neutrons, neutrons, and cosmic rays. *The Exact Evaluation of e*, chapter V, pp. 90-124. Chicago: The University of Chicago Press.

Porstendörfer, J., 1958. The diffusion coefficient and mean free paths of neutral and charged radon decay products in air. *Z. für Physik* 213:384-396.

Porstendörfer, J., Hussin, A., Scheibel, H. G., and Becker, K. H., 1984. Bipolar diffusion charging of aerosol particles-II. Influence of the concentration ratio of positive and negative ions on the charge distribution. *J. Aerosol Sci.* 15:47-56.

Porstendörfer, J. and Mercer, T. T., 1979. Influence of electric charge and humidity upon the diffusion coefficient of radon products. *Health Phys.* 15:191-199.

Pourprix, M., Daval, J., Berne, Ph., 1990. Device Permitting The Controlled Deposit of Fine Particles on Surfaces. American Association for Aerosol Research 1990 Annual Meeting. Philadelphia, PA.

Pui, D. Y. H., Fruin, S., and McMurry, P. H., 1988. Unipolar diffusion charging of ultrafine aerosols. *AS&T* 8:173-87.

Raabe, O., 1968. Measurement of the diffusion coefficient of radium A. *Nature* 217:1143-1145.

Raes, F., 1985. Description of the properties of unattached ^{218}Po and ^{212}Pb particles by means of the classical theory of cluster formation. *Health Phys.* 49:1177-1187.

Raghunath, B., Kotrappa, P., 1979. Diffusion coefficients of decay products of radon and thoron. *J. Aerosol Sci.* 10:133-138.

Ramamurthi, M. and Hopke, P. K., 1989. On improving the validity of wire screen "unattached" fraction radon daughter measurements. *Health Phys.* 56:189-194.

Reineking, A. and Pörschendorfer, J., 1986. *Aerosol Sci. Technol.* 5:482-486.

Reischl, G. P., Scheibel, H. G., and Pörschendorfer, J., 1983. The bipolar charging of aerosols: Experimental results in the size range below 20-nm particle diameter. *J. Colloid Interface Sci.* 91:272-75.

Sano, I., Fikitan, F., and Sakata, S., 1953. Colloid chemistry of tobacco smokes, distribution of size and charge of the particles. *J. Chem. Soc. Japan; Pure Chem. Sec.* 74:664-68.

Sato, T., 1987. Charging process of fine particles in unipolar ion flow. *Trans. I.E.E. Japan* 107:155-61.

Shapiro, J. V., 1954. An Evaluation of the Pulmonary Radiation Dosage from Radon and its Daughter Products. Univ. of Rochester Rep. UR-298.

Takahashi, K. and Kudo, A., 1973. Electrical charging of aerosol particles by bipolar ions in flow type charging vessels. *J. Aerosol Sci.* 4:209-16.

Wellish, E. M., 1913. The distribution of the active deposition of radium in an electric field. II. *Phi. Mag.* 6:623-635.

Whitby, K. T. and Clark, W. E., 1966. Electric aerosol particle counting and size distribution measuring system for the 0.015 to 1 μ size range. *Tellus* 18:573-86.

White, H. J., 1963. Industrial Electrostatic Precipitation. Addison-Wesley Publishing Co. Inc., Reading, Massachusetts.

White, H. J., 1951. Particle charging in electrostatic precipitation. *AIEE Trans.* 70:1186-91.

Willeke, K. and Baron, P., 1992. Aerosol Measurement: principles, techniques, and applications. New York: Van Nostrand Reinhold.

Wrenn, M.E., Spitz, H. and Cohen, N., 1975. Institute of Electrical and Electronics Engineers *IEEE Trans. Nucl. Sci.*, NS-22 645-648.

Yeh, H. C., and Cheng, Y. S., 1980. An experimental study of the effect of temperature upon aerosol charge state. *Environ. sci. Technol.* 14:726-29

Yeh, H. C., Newton, G. J., Raabe, O. G., and Boor, D. R., 1976. Self-charging of ^{198}Au -labeled monodisperse gold aerosols studied with a miniature electrical mobility spectrometer. *J. Aerosol Sci.* 7:245-53.

Yeh, H. C., Newton, G. J., and Teague, S. V., 1978. Charge distribution on plutonium-containing aerosols produced in mixed-oxide reactor fuel fabrication and the laboratory. *Health Phys.* 35:500-03.

Zhang, S. H., Akutsu, Y., Russell, L. M., Flagan, R. C., and Seinfeld, J. H., 1995. Radial Differential Mobility Analyzer. *Aerosol Sci. Technol.*

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