

Theoretical Calculation on Alpha Track Density by Using an Electrostatic Ion Spectrometer

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ABSTRACT

To develop a technique of theoretical alpha track density calculation for comparison with measured track density, an electrostatic ion spectrometer was specially designed and fabricated. The mobility spectrum of first radon daughter(Po-218) in the range of $0.07 \sim 5.0 \text{ cm}^2 \text{ V} \cdot \text{s}$ from the radon chamber was measured using the electrostatic ion spectrometer. Measurement was taken in a radon chamber operated using dry particle free air passed through silica gel, activated charcoal and molecular sieve filters. The mobility of a new-born Po-218 ion measured by the electrostatic ion spectrometer was determined to be $1.92 \text{ cm}^2 \text{ V} \cdot \text{s}$. A comparison of the theoretical and measured alpha track densities was completed and uncertainties concerning the shape of the spectrum were analyzed. It was found that the discrepancies in track densities are primarily due to the neglect of wall loss of ions in the theoretical track density calculation.

Key words; radon daughter, Po-218, radon chamber, track density, mobility spectrum, electrostatic ion spectrometer.

INTRODUCTION

It has been known for a long time that exposure to radon and its daughters contributes to an increased risk of lung cancer. Recently there are findings of individual houses with extremely high radon concentration level and there is increasing

concern that indoor radon and its daughters threat public health. There are still, however, many uncertainties as to the fundamental chemical and physical properties of the radon daughters.

Radon-222 belongs to the U-238 series which decays to stable Pb-206 through 8 alpha-particle and 6 beta-particle emissions. Radon itself is

an inert gas. However, its daughters are chemically active. Radon-222, having a half-life of 3.825 days, decays into a series of radioactive daughters, polonium-218(RaA), lead-214(RaB), bismuth-214(RaC) and polonium-214 (RaC'). The series emit 2 alpha and 2 beta particles in the decays to the long-lived lead-210(RaD).

Since polonium-218 (RaA) is formed from radon decay, its chemistry is very important in understanding the behavior of the radon daughters in indoor air. Polonium-218 has a long half-life enough to permit electrostatic collection for the study. The polonium-218 nucleus has a recoil energy of 110 keV and is found to be a singly charged positive ion 88% of the time [1,2,3]. The neutral species occurs the remaining 12% of the time.

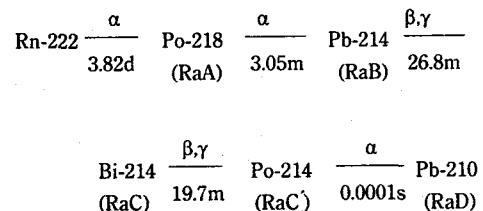
An electrostatic field can be employed to collect the positively charged ions on a detector where subsequent decays are recorded. The radon concentration is deduced from the collected activity. Monitors for environmental radon measurements using this method have been described by several investigators [4,5,6]. The collection efficiency of these monitors to a greater or lesser degree depends on the electric mobility of the polonium ions, which is a function of their size and charge [7]. A knowledge of the size of the polonium atom immediately after formation is determined using these electrostatic techniques. The removal of radon daughters by electrostatic deposition has been proposed [8,9] as a means to reduce the airborne radioactivity level in the work and living environments.

The purpose of this study was to develop a

technique of theoretical track density calculation and compare the theoretical track density with the experimental track density taken by using an electrostatic technique. For this purpose, an electrostatic spectrometer capable of measuring the sizes of the new-born polonium-218 ions in well-defined carrier gases and for controlled humidity conditions was specially designed and fabricated.

THEORETICAL TRACK DENSITY CALCULATION

The sequence of disintegrations of each atom of radon, as it transforms through the stages represented by radon daughters, can be represented schematically as



where the type of radiation and half-life are given for each nuclide. The extremely minor branches are omitted.

Equations which describe the growth of activity of Rn and RaA in a source which is initially pure radon, i.e. no radon daughters presents, can be derived from the following principles. In this research only Rn-222 and Po-218 have been used for the calculation of track density of the Po-218, since they have long enough half-lives to permit studies to be done. For Rn-222 and Po-218, let us assign subscripts A and B. Then, let us consider

a sample in a 55 gallon container with volume V and ventilation rate λ_{ven} (Fig.1).

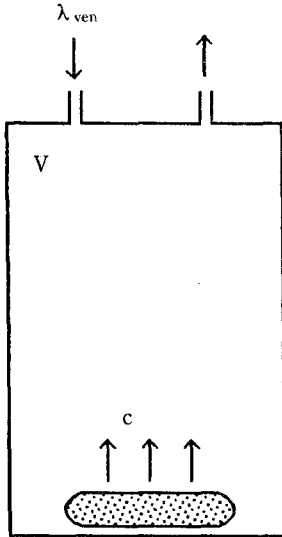


Fig. 1. Container with radon-exhaling material.

If the total exhalation rate (number of atoms per unit time) of the sample is c , the concentration of Rn-222 and Po-218 (number of atoms per unit volume), N_A and N_B , will grow with the time according to the equations

$$\frac{dN_A}{dt} = \frac{c}{V} - \lambda^* N_A, \quad \dots\dots(1)$$

$$\frac{dN_B}{dt} = \lambda_A N_A - \lambda' N_B, \quad \dots\dots(2)$$

where: λ_A is the decay constant of the radon (per sec),
 λ_B is the decay constant of the RaA (per sec),
 λ_{ven} is the ventilation rate (per sec),

λ^* is the sum of λ_A and λ_{ven} (per sec),
 and

λ' is the sum of λ_B and λ_{ven} (per sec).

In these equations, it is assumed that Rn-222 atoms and Po-218 atoms are removed from the container only by radioactive decay and ventilation. If c is assumed to be constant independently of N_A and N_B , simultaneous equations(1) and (2) have the solutions:

$$N_A = \frac{c}{V\lambda^*} + \frac{c}{V} \left(\frac{1}{\lambda_A} - \frac{1}{\lambda^*} \right) e^{-\lambda^* t} \dots\dots(3)$$

$$N_B = \frac{c}{V\lambda_B} e^{-\lambda' t} + \frac{c\lambda_A}{V\lambda^*\lambda'} (1 - e^{-\lambda' t}) + \frac{c(\lambda^* - \lambda_A)}{V\lambda^*(\lambda' - \lambda^*)} (e^{-\lambda' t} - e^{-\lambda^* t}). \dots\dots(4)$$

If the effect of wall loss to diffusion is neglected, the total number per unit volume of radon daughter atoms produced during time t is given by:

$$U = \int_0^t N_B(t) \lambda_B dt, \quad \dots\dots(5)$$

$$U = \frac{c}{V} \left[-\frac{1}{\lambda'} (1 - e^{-\lambda' t}) + \frac{\lambda_A \lambda_B}{\lambda^* \lambda'} t + \frac{\lambda_A \lambda_B}{\lambda^* \lambda'} e^{-\lambda' t} (1 - e^{-\lambda' t}) \right] \dots\dots(6)$$

$$+ \frac{\lambda_B (\lambda^* - \lambda_A)}{\lambda^* (\lambda' - \lambda^*)} \left\{ \frac{1}{\lambda'} (e^{-\lambda' t} - 1) - \frac{1}{\lambda^*} (e^{-\lambda^* t} - 1) \right\}.$$

Another problem of great practical importance for the track density of Po-218 is diffusion to the inner wall of the tube of the Po-218 atoms flowing through the tube. Mathematical solutions have

been obtained for a few simple cases such as laminar flow through a tube having a circular or a rectangular cross section. The solution for a circular tube will give the penetration fraction of entering atoms that exit through a tube of diameter d_c as a function of the dimensionless deposition parameter, μ , as given by [10]:

$$\mu = \frac{4DL}{\pi d_c^2 U} = \frac{DL}{Q} \quad \dots\dots(7)$$

where: D is the particle diffusion coefficient (cm^2/s),

L is the length of the tube (cm),

U is the average velocity through the tube ($U=Q/A$)(cm/s)

Q is the volume flow rate through the tube (cm^3/s), and

A is the cross-sectional area of the tube (cm^2).

The penetration fraction P as a function of μ with an accuracy of 1% for all values of μ can be calculated using the following expression[10]:

$$P = 1 - 5.50 \mu^{2/3} + 3.77 \mu \quad \text{for } \mu < 0.007, \quad \dots\dots(8)$$

$$P = 0.819 \exp(-11.5\mu) + 0.0975 \exp(-70.1\mu) + 0.0325 \exp(-179\mu) \quad \text{for } \mu > 0.007, \quad \dots\dots(9)$$

the solution for determining the penetration of an atom flowing through a rectangular tube follows.

The deposition parameter is defined by:

$$\mu = \frac{DLW}{Qh} \quad \dots\dots(10)$$

where: L is the length of the channel in the direction of flow (cm),

W is the width of the channel (cm), and h is the separation distance between the plates (cm).

Simplified equations for penetration, accurate to 1% for all values of μ , are obtained from[10]:

$$P = 1 - 2.9 \mu^{2/3} + 0.4 \mu \quad \text{for } \mu < 0.003, \quad \dots\dots(11)$$

$$P = 0.910 \exp(-7.54\mu) + 0.0531 \exp(-85.7\mu) + 0.0153 \exp(-249\mu) \quad \text{for } \mu > 0.003 \quad \dots\dots(12)$$

Equations (7) to (12) were used here to calculate the penetration fraction of the sample tube (Fig.2) installed in the spectrometer. A more refined calculation has been made by dividing the sample tube into three rectangular channels and one circular tube as shown in Fig. 2.

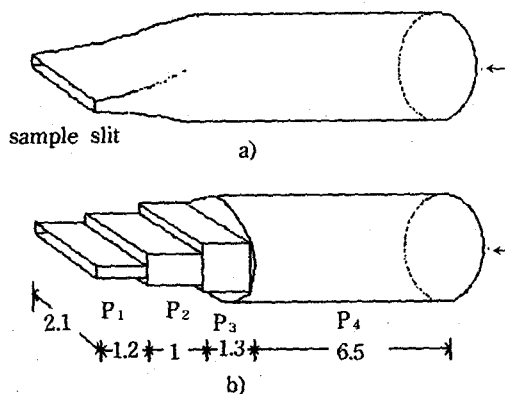


Fig. 2. a) Original inlet samlet tube, b) mathematical model of the inlet sample tube.

When positive Po-218 ions are discharged between the charged plates through the sample tube from the container, all of the positive Po-218 ions are attracted to the negatively charged plate and are accumulated in a predictable manner as

illustrated in Fig. 3. When all positive ions are assumed to attach to the negative plate, the total number of Po-218 ions deposited on the plate can be calculated using equation (6).

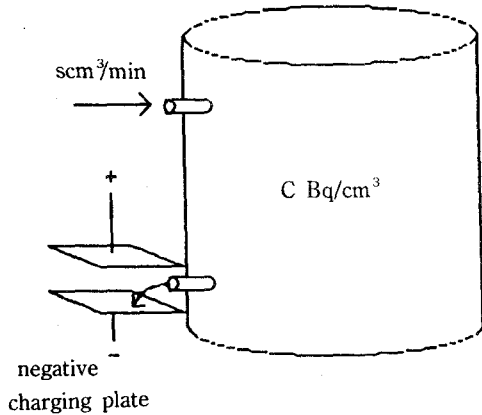


fig. 3. Schematic diagram for the accumulation of the positive ions on the negative charging plate (The concentration of radon in the container is $C \text{ Bq/cm}^3$, in an air stream which penetrates through the sample tube at a volumetric flow rate of $s \text{ cm}^3/\text{min}$ during the sampling time t).

The lower negatively charged plate carries an alpha track film (LR 115 II) with the sensitive surface oriented toward the interior of the device. The alpha track film is used to detect positive Po-218 ions. The film is covered with Al foils to discriminate all radon daughters except Po-218. A $10 \mu\text{m}$ thick Al foil absorber can be chosen for the detection of Po-218 atoms due to the peak in the detection efficiency at this thickness while discriminating most of the Po-214 alpha particles as shown in Fig. 4[11]. Factors which should be considered for the calculation of the track density are the alpha track detector efficiency, the

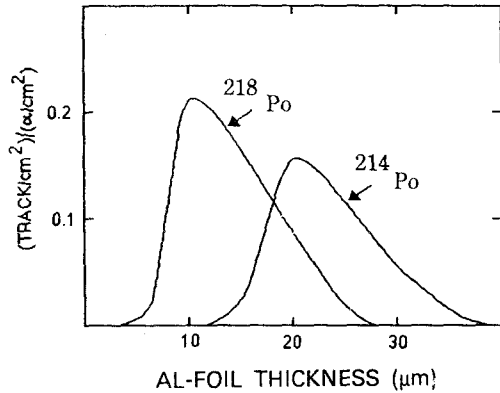


Fig. 4. Calculated efficiency for an alpha source plated out on a Al-foil covered LR 115 II detector [11].

fraction of the positive Po-218 ions, the sample tube penetration fraction, and the actual area of the film per total area of the charging plate. Therefore, the actual number of positive Po-218 ions deposited on the film per mm^2 , namely, the theoretical track density, is

$$\begin{aligned}
 & U \times (\text{the fraction of positive Po-218 ion}) \\
 & \times (\text{alpha track detector efficiency}) \\
 & \times (\text{tube penetration fraction}) \\
 & \times (\text{the actual area of the film per the total} \\
 & \quad \text{area of plate}) \\
 & \times (\text{per the actual area of the film}). \dots(13)
 \end{aligned}$$

Consider a positive ion penetrating between the charged plates. Then assume that a uniform electric field exists between these plates shown in Fig. 5. Let the applied voltage be V , H the distance between the plates, and v the average flow velocity. The ions of mobility K become attached to the negatively charged plate at a distance x from the sample tube slit where x is given by:

$$x = bHv/KV. \dots\dots(14)$$

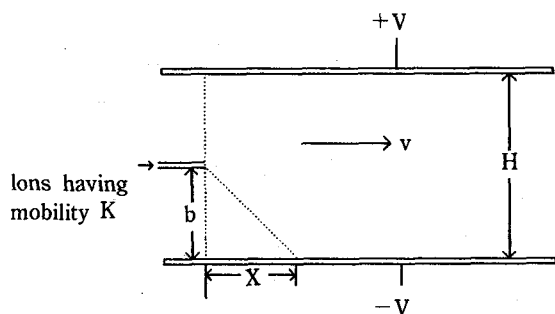


Fig. 5. Schematic diagram showing ion trajectory in the electrostatic ion spectrometer.

EXPERIMENTAL SYSTEM AND MEASUREMENTS

The experimental system used in this study was designed to provide a means of calibrating and characterizing the electrostatic spectrometer developed to measure an experimental track density. This system consisted of a radon chamber, filter system, manometer, flowmeters, vacuum pumps, and a plastic chamber.

The spectrometer was composed of two parallel brass plates and two side pieces of plexiglass forming a rectangular chamber as shown in Fig. 6. Both ends of the chamber were designed to hold a uniform array of hollow cylinders and a plastic filter which creates a uniform flow of the gas. This suppresses turbulence that might otherwise disturb the flow as it enters the chamber. A uniform electric field was obtained by connection to a DC high voltage supply.

The upper brass plate carries an alpha track film with the sensitive surface oriented toward the interior of the tube. The film was covered with $10\mu\text{m}$ thick Al foils to discriminate against all

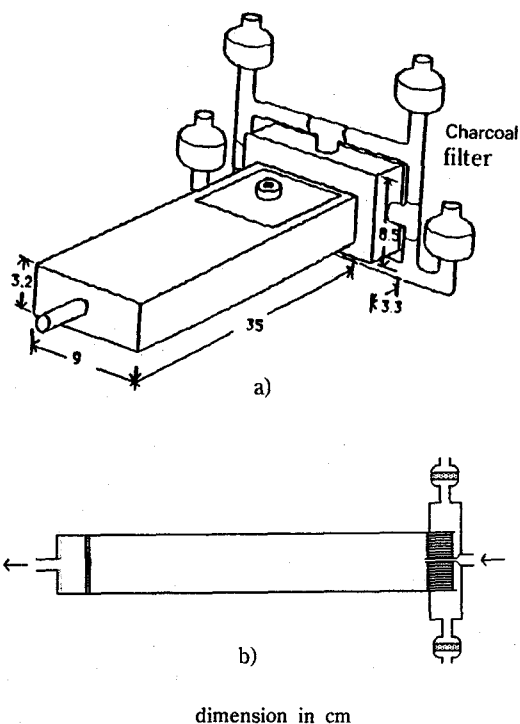


Fig. 6. a) The oblique view, b) the front view of the electrostatic ion spectrometer.

radon daughters except Po-218 [11]. The upper and lower brass plates were charged to potential $+240\text{ V}$ and -240 V , respectively, so that all of the positive Po-218 ions penetrating through the sample tube were attracted toward the upper, negatively charged plate.

Since the field between the plates was uniform, each ion followed a specific trajectory before it is deposited on the plate. In practice the apparatus was operated for a long time (240 min) enough to collect measurable activity on the plate. Subsequent analysis of the track density on the upper plate as a function of distance from the projection of the slit yielded a band of tracks.

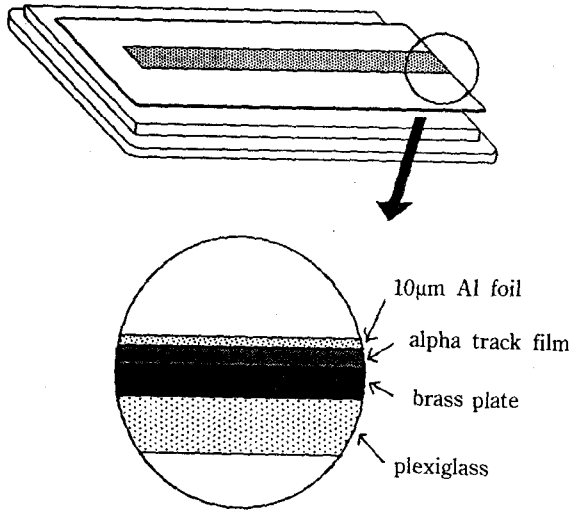


Fig. 7. The removable section of the electrostatic ion spectrometer.

The layout of the experimental system is shown in Fig. 8. The radon chamber was fabricated from a 55 gallon drum and was designed with the air flow inlet and outlet tubes located at the top of the drum. Uranium ore was used as a radon source and was placed inside on the bottom of the chamber. The radon chamber was operated using dry particle free air passed through silica gel, activated charcoal and molecular sieve filters to remove moisture and organic vapors. The air flow into the chamber was maintained at 800 cm³/min, equivalent to the spectrometer sample flow rate.

In the present study, external aerosols were not used. The calibration and characterization studies

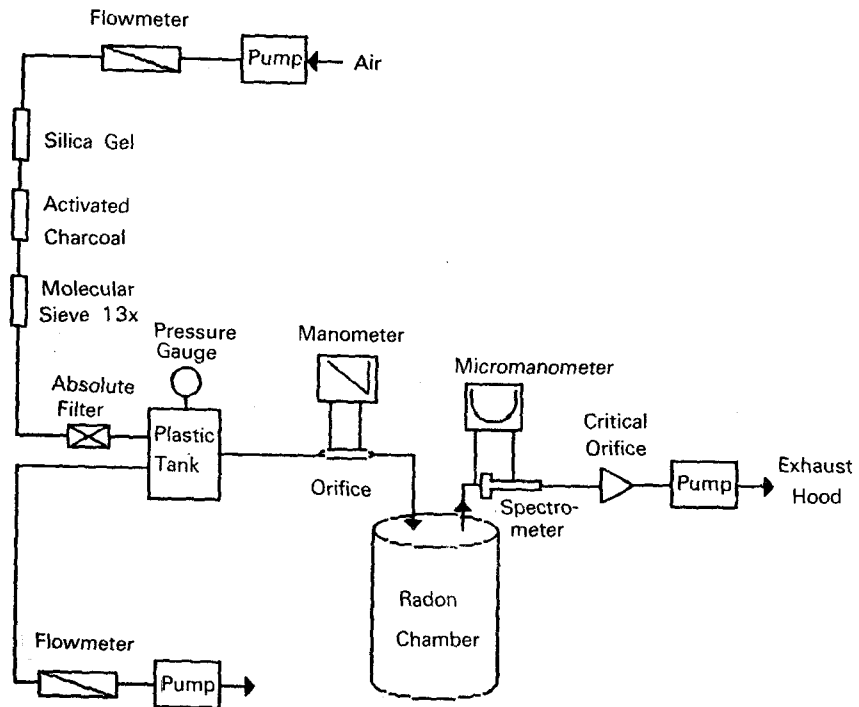


Fig. 8. The layout of the radon chamber system.

of the electrostatic spectrometer were performed using radon gas exhaled from uranium ore within the chamber. The radon concentration of the chamber used was $2 \times 10^{-4} \text{Bq/cm}^3$ measured by the charcoal canister[12].

Po-218 ions and clusters deposited on the upper charged plate after an experiment were detected as follows: A $6 \times 13.5 \text{ cm}^2$ section of the collector plate was removable and fit flush with the collector plate (Fig. 7). After exposure this section was removed and separated from a contact sheet of LR-115 alpha track film[13].

Alpha particles on the collector plate were detected by a $112 \mu\text{m}$ thick LR 115 alpha track film. To make the track visible, a film developing and analysis procedure developed by Jessick [14] was employed. The dried, etched film was placed on a microscope slide and the image of the tracks was recorded by a video camera using a computer-controlled image analysis system and video monitor [11]. To determine the track density distribution, the video screen was moved progressively from the line in 1 mm increments along the longitudinal centerline. This rectangular video screen was calibrated to a length of 0.97 mm and width of 0.72 mm. Due to the fact that the horizontal flow distribution is flat at the centerline, only tracks in a 1 cm wide strip lying along the longitudinal center line of the film were counted. The track densities of 5 areas on each strip along the line were averaged and the background subtracted. The background was determined on the film where no ions were deposited and was about 0.4 tracks/mm^2 .

RESULTS AND DISCUSSION

Calibration of the specially-designed electrostatic spectrometer was performed using the radon chamber system discussed in the previous section. The resulting track density vs. travel distance was translated into track density vs. ion mobility spectrum shown in Fig. 9 using equation(14). The most probable mobility appears to be $1.92 \text{ cm}^2/\text{V} \cdot \text{s}$. The ion mobility spectrum obtained with the radon chamber (250 min residence time) in filtered room air (Fig. 8) indicates the formation of ultrafine aerosol particles in the $1\text{-}1.7 \text{ cm}^2/\text{V} \cdot \text{s}$ mobility range. This peak broadening in the $1\text{-}1.7 \text{ cm}^2/\text{V} \cdot \text{s}$ mobility range is due possibly to the presence of unknown trace gases and organics.

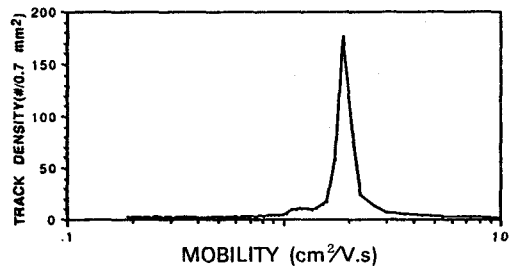


Fig. 9. Track density vs. ion mobility spectrum of Po-218 using the radon chamber system (5.5% RH).

The ion spectrum resolution is reported as the half-width of band at one-half its maximum value and expressed as a percentage of the measured ion mobility. The resolution of the our experimental system was 16.2%. The spectrometer was designed to have a resolution of 15%. Better ion spectrum resolutions were obtained using a critical orifice instead of a rotameter between the spec-

trometer and vacuum pump.

The theoretical track densities calculated using equation (13) was used to compare with the measured track densities. The fraction of positive Po-218 ions was taken to be 0.88 [1,2,3] and the alpha track detector was assumed to have 20% efficiency under a 10 μm thick Al foil, as was shown in Fig. 4. The calculated penetration fraction of the sample tube used in the experiment and the actual area of the film per the total area of plate were 0.088 and 0.296, respectively. Considering that the positive Po-218 ions are deposited on the film during the sampling time of 240 min, the theoretical track density was 11.4 ions/ mm^2 . However, the measured track density from Fig. 5 was 10.3 ions/ mm^2 . The difference between these two track densities is due primarily to neglect of consideration of wall loss of ions in the theoretical track density calculation. The diffusion loss of ions in the spectrometer was negligible due to the very short travel time of the trapped ions in the diffusion region.

CONCLUSIONS

This study has focussed on the development of new calibration method of the electrostatic ion spectrometer. A well-controlled experimental chamber was used in this study. The electrostatic spectrometer could be used to study the nature of the charged Po-218 ions formed by radiolysis and its dependence on the trace gas and water vapor concentrations. The mobility of a new born Po-218 ion measured by a specially-designed ion spectrometer was estimated to be $1.92 \text{ cm}^2/\text{V} \cdot \text{s}$. The uncertainties concerning the shape of the

spectrum were also analyzed. It was realized that the use of a fabricated critical orifice between the spectrometer and vacuum pump could obtain better ion spectrum peak.

A comparison of the theoretical and measured alpha track densities was completed to investigate the performance of the specially-designed ion spectrometer. It was concluded that the difference between these two track densities is primarily due to the neglect of wall loss of ions in the theoretical track density calculation.

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정전기분광분석방법에 의한 알파입자비적밀도의 이론적계산연구

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요 약

알파비적밀도의 이론계산방법을 개발하기 위하여 정전기방법을 이용한 분광계를 특별히 설계 제작하였다. 이 정전기분광계를 사용하여 라돈의 첫째딸핵종인 Po-218의 이온에 대해 $0.07 \sim 5.0 \text{cm}^2/\text{V}\cdot\text{sec}$ 범위의 이동도스펙트럼을 측정하였다. 이측정은 실리카겔, 활성탄, 분자구조의 시브필터를 통과한 탈입자가스를 사용하는 라돈챔버내에서 수행되었다. 제작된 정전기분광계를 사용하여 챔버내에서 새로 생성된 Po-218 이온의 이동도는 $1.92 \text{cm}^2/\text{V}\cdot\text{s}$ 인것으로 측정되었다. 알파비적밀도의 이론적계산방법에 의해 얻어진 결과를 측정에 의해 얻어진 결과와 비교하였으며 측정된 스펙트럼에 관계된 불확실도를 분석하였다. 이론계산방법에 의해 얻어진 알파비적밀도와 측정된 값과의 차이에 대한 이유는 이론계산방법에서 이온의 라돈 챔버에서의 벽제거효과를 고려하지 않았기 때문인 것으로 밝혀졌다.

Key words: 라돈딸핵종, Po-218, 라돈챔버, 비적밀도, 정전기이온분광계, 이동도스펙트럼.