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ATOMIC ENERGY OF CANADA LIMITED

L'ÉNERGIE ATOMIQUE DU CANADA LIMITÉE

THORON-IN-BREATH MONITORING AT CRNL

Détection à Chalk River du thoron rejeté par la respiration

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Chalk River, Ontario

April 1985 avril

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THORON-IN-BREATH MONITORING AT CRNL

by

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Résumé

On décrit dans ce rapport le détecteur TIBM (Thoron-In-Breath Monitor) ayant été développé à Chalk River. Ce détecteur permet d'estimer la quantité de thorium (Th-232 et/ou Th-228) se trouvant dans l'organisme humain. La détection des émanations de thorium dans la respiration est fondée sur le fait que le thoron (Rn-220) est un produit de désintégration du thorium. Le thorium déposé dans l'organisme produit donc, *in vivo* du thoron dont une partie est expirée. Lors de la détection, la personne examinée expire de l'air dans une chambre collectrice où le thoron se transforme en Po-216 et Pb-212 dont la charge est positive. Les produits de filiation sont recueillis sur un disque en mylar recouvert de ZnS et placé sur une électrode. L'étalonnage de l'instrument indique que 48 ± 8% des atomes de Pb-212 engendrés dans la chambre sont recueillis sur l'électrode à une tension de 4000 volts. Le rendement de la collecte s'est avéré indépendant du débit de l'air dans la chambre lequel se situe entre 5 et 10 l/min^{-1}, ce qui correspond à la gamme de respiration de la plupart des personnes au repos. Le rendement de la collecte est réduit par la tension si celle-ci est inférieure à 3500 volts. Par contre, il reste constant si la tension est supérieure à 3500 volts. L'activité minimale que l'instrument TIBM peut détecter dans la chambre où les personnes expirent l'air de leurs poumons est estimée à 0,012 Bq par minute ou 16 Bq par jour.

Les expériences effectuées avec le détecteur TIBM ont montré que la détection des émanations de thorium est facile à faire et que les résultats en matière de personnes contaminées ou non, peuvent être aisément interprétés. Les études se poursuivent en ce qui concerne la relation existant entre le thoron expiré par les poumons et le thorium déposé dans l'organisme et donc entre le thoron expiré et la dose reçue.

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ABSTRACT

This report contains a description of the thoron-in-breath monitor (TIBM) developed at CRNL. This monitor can be used to estimate the amount of thorium (Th-232 and/or Th-228) in humans. Thoron-in-breath monitoring is based on the fact that thoron (Rn-220) is a decay product of thorium, and hence deposited thorium produces thoron in vivo, a fraction of which will be exhaled. During the monitoring the subject exhales into a collection chamber where the thoron decays into Po-216 and Pb-212, both positively charged. The charged daughters are collected on a ZnS coated mylar disk placed on an electrode. The calibration of the instrument indicates that 48 ± 8% of the Pb-212 atoms generated in the chamber are collected on the electrode at a voltage of 4000 V. The collection efficiency was found to be independent of the flow through the chamber in the range from 5 to 10 L min^{-1}, which corresponds to the range of breathing for most subjects at rest. The collection efficiency is also a function of voltage for voltages below 3500 V, above which it is constant. The minimum detectable activity of TIBM, in terms of thoron activity exhaled, was estimated to be 0.012 Bq exhaled per minute or 16 Bq exhaled per day.

Experiences with the TIBM indicate that the monitoring is easy to perform and the results in terms of contaminated vs uncontaminated subjects can be easily interpreted. Work on relationships between thoron exhaled and deposited thorium and hence between thoron exhaled and dose, is continuing.

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1. INTRODUCTION

Research and development on the thorium fuel cycle has been underway for some time at the Chalk River Nuclear Laboratories (CRNL). At present, CRNL purchases and processes about one tonne of thorium containing compounds (mainly thorium oxide and thorium nitrate) (1). Handling and processing of thoria (ThO_2) powders or other thorium compounds can result in a potential human intake of thorium containing compounds. Thorium compounds can find their way into the human body by either ingestion, inhalation or through wounds. Due to the low solubility, the ingested thoria powders (ThO_2) are almost completely eliminated (2). Inhalation of insoluble thoria powders represents the more hazardous route of the radiological exposure. The inhaled thoria particles will deposit on the walls of the lung airways, and will be removed from the lungs depending on their location. The larger particles (about 5 μm and above), deposited mainly in the tracheo-bronchial region, will be removed within a week by the mucociliary clearance process (3, 4). The smaller thoria particles (up to 5 μm), deposited in the pulmonary region, will be cleared from the lungs by the dissolution in the blood of the lungs and by the lymph transport. The clearance from the pulmonary region is slow with the half-life of the order of years (thorium oxide is therefore classified as a Y compound). From the lungs, thorium is transported to other organs of the body such as skeleton, liver, spleen, kidney, etc. (5).

International Commission on Radiological Protection (2) recommends the values for the annual limit on intake (ALI) for Th-232 and Th-228 of 100 Bq and 600 Bq, respectively. At CRNL the control limit for internal contamination is set at 1/20 ALI (6) which corresponds to the investigation level (IL) defined and recommended by ICRP (7). At CRNL the investigation level is also taken as a control level for removal of workers from further contamination. This level corresponds to an intake of 5 Bq for Th-232 and 30 Bq for Th-228 and about 15% of these amounts will remain in the lung for long times after an intake. The derived investigation level (DIL) for lung monitoring is therefore taken to be 1 Bq and 5 Bq for Th-232 and Th-228, respectively.

At present three different approaches are being used to measure the internally deposited thorium: whole body counting (8), lung counting (9) and excreta analysis (10). The minimum detectable activity ($MDA_{2\sigma}$) of the whole body counter is of the order of 50 Bq for Th-228 and from 50 to 140 Bq for Th-232 (depending on the time when thorium was extracted from the ore). The minimum detectable activity of the lung counter is about 5 Bq for Th-228 and from 5 to 13 Bq for Th-232. Comparing the values for DIL's with those of MDA's one can see that the detectabilities of neither the whole body counter nor the lung counter are adequate for monitoring of thorium. A further drawback of the lung monitor is that the peak due to the K-X-rays emitted by Ra-224 (the activity actually measured) is broad and difficult to identify. In addition, the presence of any other radioactive contaminants (such as Xe-133) in the person will interfere with the identification of the peaks. Therefore, it was decided to develop a method which would have a better sensitivity and a better reliability of identifying thorium than the existing methods. In this report, the development, calibration and performance of the instrument which identifies internally deposited thorium by detecting thoron gas (Rn-220) in breath of a subject, is presented. At the end of this report, the work done on the interpretation of the results in terms of the relationship between the amount of thoron exhaled and dose received by the subject due to internally deposited thorium (11, 12) is briefly discussed.

2. TECHNICAL APPROACH

2.1 Background

Thoron-in-breath monitoring (TIBM) is based on the fact that thorium, internally deposited in a subject, will decay producing thoron which will be exhaled. Five different approaches have been reported for monitoring thoron in breath.

(i) collection of exhaled air in a plastic bag, followed by counting of alpha activity by means of an ionization chamber (13),

(ii) collection of exhaled thoron by cooled charcoal and counting alpha activity due to thoron decay (14),

(iii) collection of thoron daughters on a filter and measuring the alpha activity of the thoron daughters (15),

(iv) passing of thoron through a ZnS detector and counting the alpha activity of thoron decay (16), and

(v) electrostatic collection of thoron daughters followed by counting of alpha activity due to the decay of thoron daughters (17).

The instrument described in this report is based on electrostatic collection of positively charged thoron daughters and is similar in design to the one described by Keane and Brewster (17).

2.2 Design

General features of the thoron-in-breath monitoring can be seen in Figure 1. The subject is asked to breathe through a Hans Rudolph type inspiratory/expiratory valve (Bionetics Ltd., Montreal, Quebec). To ensure mouth breathing the subject's nostrils are closed with a noseclip, and exhaled breath is then fed into a 37.7 L collection chamber. Thoron decays with a half-life of 55 seconds into Po-216 which in turn decays with a half-life of 158 msec into Pb-212. Assuming a respiratory minute volume of 7 L min^{-1} and complete mixing, the mean residence time of thoron in the chamber is over 5 half-lives allowing for 98% of thoron to decay in the chamber. The tubing between the mouth of the subject and the collection chamber represents a dead space where thoron decays. The fraction of exhaled thoron that will enter the collection chamber (γ) can be estimated by (17):

$$\gamma = e^{-\lambda_{12} \frac{V_d}{\phi}}$$

where λ_{12} is the decay constant for thoron (0.76 min^{-1}), ϕ is the flow rate, and V_d is the volume of the tubing (0.3 L). Assuming the flow rate of 7 L min^{-1}, γ can be estimated to be about 97%.

Thoron daughters Po-216 and Pb-212 produced in air (see Appendix A) from thoron decay are positively charged which results from the emission of secondary electrons during the recoil of the nucleus when an alpha particle is ejected (18). Positively charged Po-216 and Pb-212 (half-life of 10.6 hours) are collected on a ZnS coated mylar disc mounted in front of a brass electrode. The mount of the mylar disc is displayed in Figure 2.

TIBM procedure consists of three steps: equilibration, collection, and measurement of alpha activity. Equilibration (10 minutes) allows the concentration of Tn in the collection chamber to reach the equilibrium, it furthermore allows the measurement of the respiratory flow (in L min^{-1}) of the subject with a wet test meter, and finally allows the subject to settle down and assume normal breathing. Due to the relatively high backpressure generated by the wet test meter, the wet test meter is connected only during the equilibration period. The collection of daughter products (i.e. voltage on) lasts 30 minutes after which the mylar disc is placed in an alpha counter for an overnight count.

2.3 Counting of Alpha Activity

Three different counters are available in the Dosimetric Research Branch to measure alpha activity of collected thoron daughters.

(i) Beckman Widebeta II - an alpha/beta counter with an efficiency of 21%,

(ii) ZnS screen/photomultiplier tube counter (referred to hereafter as an alpha counter) with an efficiency of 80%, and

(iii) a ruggedized partially depleted silicon surface barrier detector located in an evacuated chamber. The efficiency of this detector was estimated by known quantities of calibrated Th-228 on a stainless steel disc. The efficiency for this detector was found to be 30.5%. The efficiencies of Widebeta II and alpha counter were evaluated on the basis of this estimate (values given above).

2.4 Calibration

2.4.1 Alpha Spectra of Th-228 and Tn Daughters

Figure 3 shows an alpha spectrum of Th-228 (in equilibrium with its daughters) taken with a ruggedized surface barrier detector. The source was prepared by evaporating a small amount of Th-228 solution on a stainless steel disc. The spectrum shows six peaks corresponding to six alpha particles of different energies. Figure 4 shows an alpha spectra of thoron daughters collected electrostatically on a mylar disc counted at different times after collection.

Thoron daughters emit alpha particles of three different energies, 6.05, 6.78 and 8.78 MeV. Due to the short half-life of Po-216 (158 msec) this isotope will decay before it can be counted. Therefore, only two peaks, one at 6.05 MeV due to the decay of Bi-212, and the other at 8.78 MeV due to the decay of Po-212, are observed.

2.4.2 Evaluation of TIBM Efficiency

Thoron-in-breath monitor (TIBM) was calibrated by passing air (at 5 L min^{-1}) through a Th-228 solution of known activity. Thoron gas is swept out of the solution into the collection chamber where thoron decays (see Figure 1).

The collection efficiency (ε_{col}) of the thoron-in-breath monitor is defined as the ratio of the rate of the Pb-212 atoms collected on the electrode to the net rate of the Pb-212 atoms generated in the chamber.

$$\varepsilon_{col} = \frac{(d\ N_3/dt)_e}{(d\ N_3/dt)_n}$$

where N_3 represents the number of Pb-212 atoms. (For the equations describing the equilibration, collection and the ingrowth and decay of alpha activity consult Appendix A.) The efficiency (ε_{col}) was determined by varying the value for ε_{col} until a good agreement was achieved between the calculated ($A_\alpha(t)$ vs t, see Appendix A) and measured activities. The alpha activity due to the decay of Bi-212 and Po-212 (denoted by $A_\alpha(t)$) was calculated by integrating the set of differential equations given in Appendix A. Integration was carried out by means of the FORSIM integration package (19). Figure 5 shows a comparison between the calculated activity and experimental measurements. The average value of seven calibrations was found to be (48 ± 8)% (at the collection voltage of 4000 V) which is in good agreement with the value of (54 ± 4)% reported by Keane and Brewster (17) for 4000 V. The errors are expressed as sample standard deviations.

2.4.3 Effect of Voltage on Collection Efficiency

The higher the voltage (and electrical field) the faster the charged thoron daughters (Po-216 and Pb-212) will reach the electrode. Figure 6 shows the relationship between the collection efficiency and voltage on the electrode. Above 4000 V the collection efficiency remains constant indicating that all available charged thoron daughters are collected. The calibration and all routine measurements were carried out at 4000 V.

2.4.4 Effect of Flow Rate on Collection Efficiency

Normal subjects breathe anywhere from 5 L min^{-1} to 10 L min^{-1} when resting. With higher flow rates the residence time of thoron in the collection chamber will be shorter and less thoron daughters will be collected on the electrode (i.e. the apparent collection efficiency will decrease). The collection efficiency varies with the flow in the following manner (17):

$$\varepsilon_{col} = \varepsilon^{o}_{col} \quad e^{-\lambda_{12} \frac{V_d}{\phi}} \quad (1 - e^{-\lambda_{12} \frac{V_c}{\phi}})$$

where ε^{o}_{col} is the measured collection efficiency, and λ_{12}, V_d, V_c and ϕ are the thoron decay constant, dead volume, chamber volume and flow rate, respectively. Figure 7 shows the comparison between the calculated collection efficiency ε_{col} (ε^{o}_{col} was taken as 48% at the flow of 5 L min^{-1}) and the measured collection efficiency. At the low rates the collection efficiency will decrease due to the decay of thoron in the dead space (for volumes below 2 L min^{-1}). One can conclude from Figure 7 that the efficiency does not depend strongly on the flow rate in the range from 5 to 10 L min^{-1}.

3. COUNTING PROCEDURE

Besides the thoron daughters, radon daughters are also collected on the electrode. The collection of radon daughters on the electrode was also simulated by a computer program similar to the one developed for thoron daughters (Appendix A). In about 2 1/2 hours after the end of the collection period the alpha activity due to the decay of radon daughters decays to about 2% of the activity at the end of the collection period. Due to the interference from the radon daughters, especially at the beginning of the counting period, the early part of the decay was fitted to the decay of radon daughters and only the counts from 140 minutes on were attributed to the decay of thoron daughters (after the counts due to the radon daughters were subtracted). In routine measurements the decay of thoron daughters is followed overnight for a total counting time of 840 minutes (i.e. from 140 minutes to 980 minutes after the end of the collection period). In this time interval 55% of the decays of thoron daughters will occur.

4. MINIMUM DETECTABLE ACTIVITY

The minimum detectable activity was calculated using the following assumptions: the duration of the collection period is 30 minutes, collection efficiency is 48%*, detector efficiency is 0.8 and counting was carried out over a period from 140 minutes to 980 minutes after the collection period when 55% of the decays occur. The average background of the alpha counter was measured to be 0.01 cpm (with a ZnS coated mylar disc in the sample holder).

If one assumes the flow of 1 Tn atom per minute leaving the subject's mouth over a collection period of 30 minutes, during the counting period of 840 minutes one would expect a total of 6.3 counts due to the decay of thoron daughters collected on the mylar disc. The uncertainty in the background count ($2\sqrt{N}$ where N is the number of background counts in 840 minute period) is 5.8 counts which corresponds to the rate of thoron exhalation at mouth of 0.92 Tn atoms per minute (equivalent to 1325 Tn atoms exhaled per day). In terms of thoron activity exhaled the minimum detectable activity corresponds to 0.012 Bq of thoron exhaled per minute or 16 Bq of thoron exhaled per day.

When the control subjects (subjects who never worked with thorium, i.e. "uncontaminated") were monitored for thoron in breath, in average 30 counts above background were detected in the counting period of 840 minutes.

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* One can easily show that the efficiency of Pb-212 atoms collected is equivalent to the ratio of Tn atoms detected to Tn atoms entering the chamber.

One can define the upper limit for the uncontaminated subjects as 2σ above this average value (30 counts + 2 $\sqrt{N}$ = 41 counts). The value of 41 counts then represents the lower limit of detection for thorium exposure which is equivalent to 0.08 Bq of Tn exhaled per minute.

5. RELATIONSHIP BETWEEN THORON EXHALED AND THORIUM DEPOSITED IN HUMAN SUBJECTS

In order to make use of the measured thoron in breath, the relationship between exhaled thoron and deposited thorium must be known. A model used to describe this relationship is described briefly here. A more detailed account of the calculations can be found elsewhere (11, 12). In the model the following assumptions were made:

(i) the deposition and retention of thorium in the body are that recommended by ICRP (2),

(ii) the inhaled particles have a log-normal distribution with an activity median aerodynamic diameter of 1 μm,

(iii) the amount of thoron emanated from the particle results entirely due to the recoil of the alpha particle (20) and, furthermore, that the emanation of thoron follows the formalism described by Quet, et al. (21),

(iv) the exhalation of thoron from the lungs will follow mechanism developed by Peterman and Longtin (11), and

(v) the fractions of thoron reaching the lungs (from the other organs) will be similar to the estimates found for thorotrast patients (13, 16, 22) and animals (23-25) exposed to thorium.

The output of the model are the relationships between the activity of thoron exhaled in a day per activity of ancestor inhaled vs days post exposure and the dose rate received in a day per activity of thoron exhaled vs days post exposure (12). Thus, if one knows the time of the exposure, by measuring the amount of thoron exhaled, one can estimate the effective dose equivalent a person is receiving as a result of thorium exposure. It should be pointed out that some of the parameters, which are not known, needed for the calculations were extrapolated and/or roughly estimated and, therefore, the above relationships are somewhat uncertain.

6. DISCUSSION AND CONCLUSION

So far some forty subjects have been monitored for thoron in breath. Except for occasional interferences from large concentrations of radon the results of repeated counting on the same subjects are reproducible. The results of the counting are simple to interpret (in terms of contaminated vs uncontaminated) and do not suffer from the ambiguity of gamma peak assignments as is often encountered in the lung and whole body measurements. The interpretation of the amount of Tn exhaled in terms of amount of thorium injected (13) or thorium measured in thorax (26) is more difficult. A larger scatter was observed when thoron exhaled was related to the amount of thorium injected (13) or measured in thorax (26). This probably indicates different depositions of thorium and/or different exhalation rates (13).

In summary, although TIBM is simple to perform and the results in terms of contaminated vs uncontaminated are simple to interpret, the relationship between the amount of thoron exhaled and thorium in various organs (and hence dose) are more complicated, and considerable uncertainty exists. Work on improving our understanding of the relationship between thoron in breath and deposited thorium is continuing.

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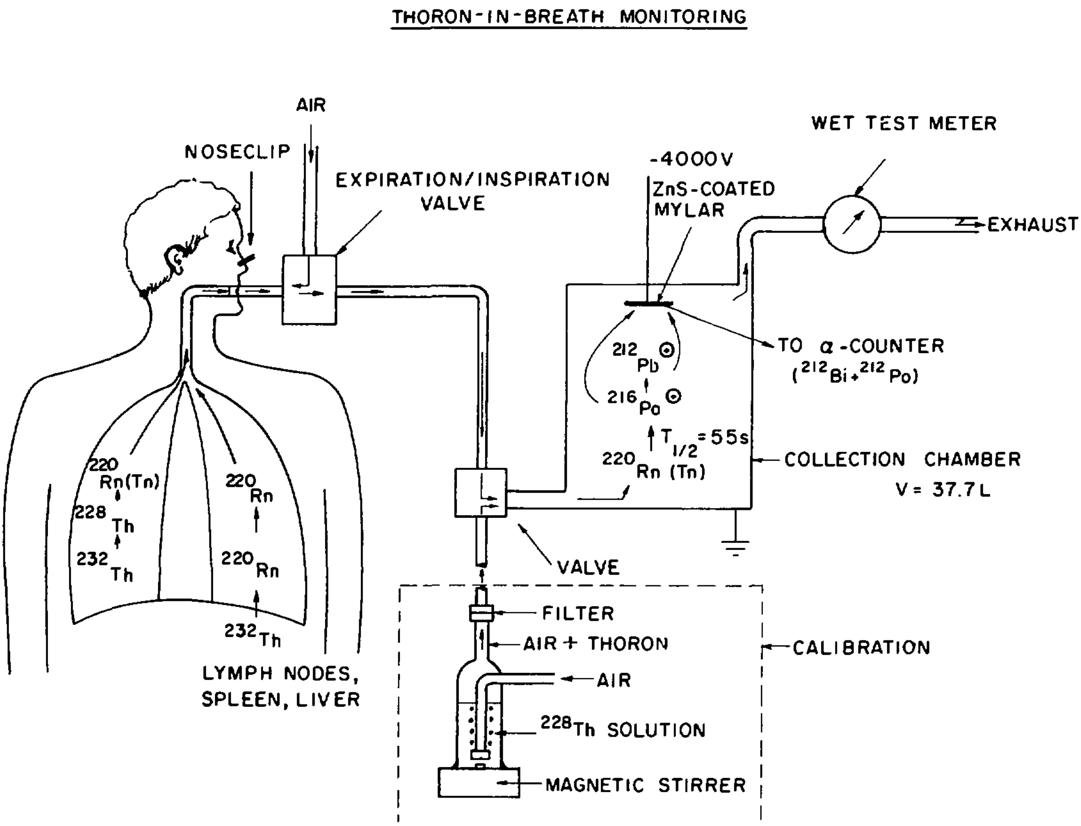


FIGURE 1 Schematic presentation of thoron-in-breath monitoring

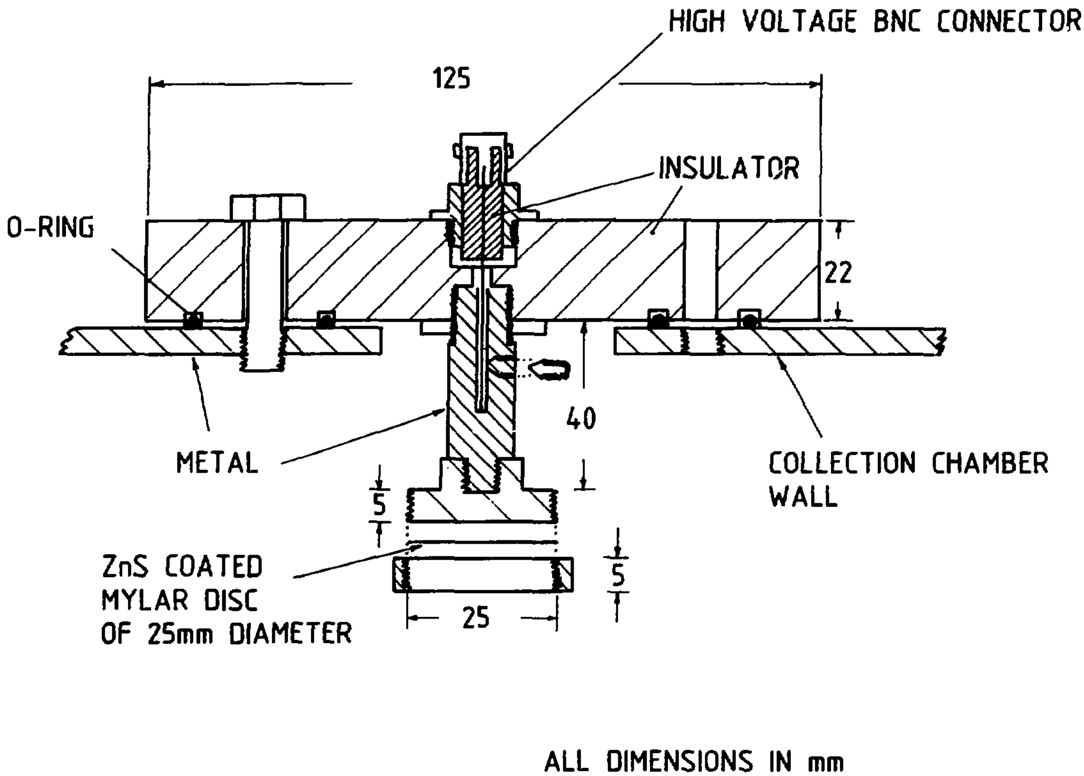


FIGURE 2 Schematic presentation of mylar disk mount

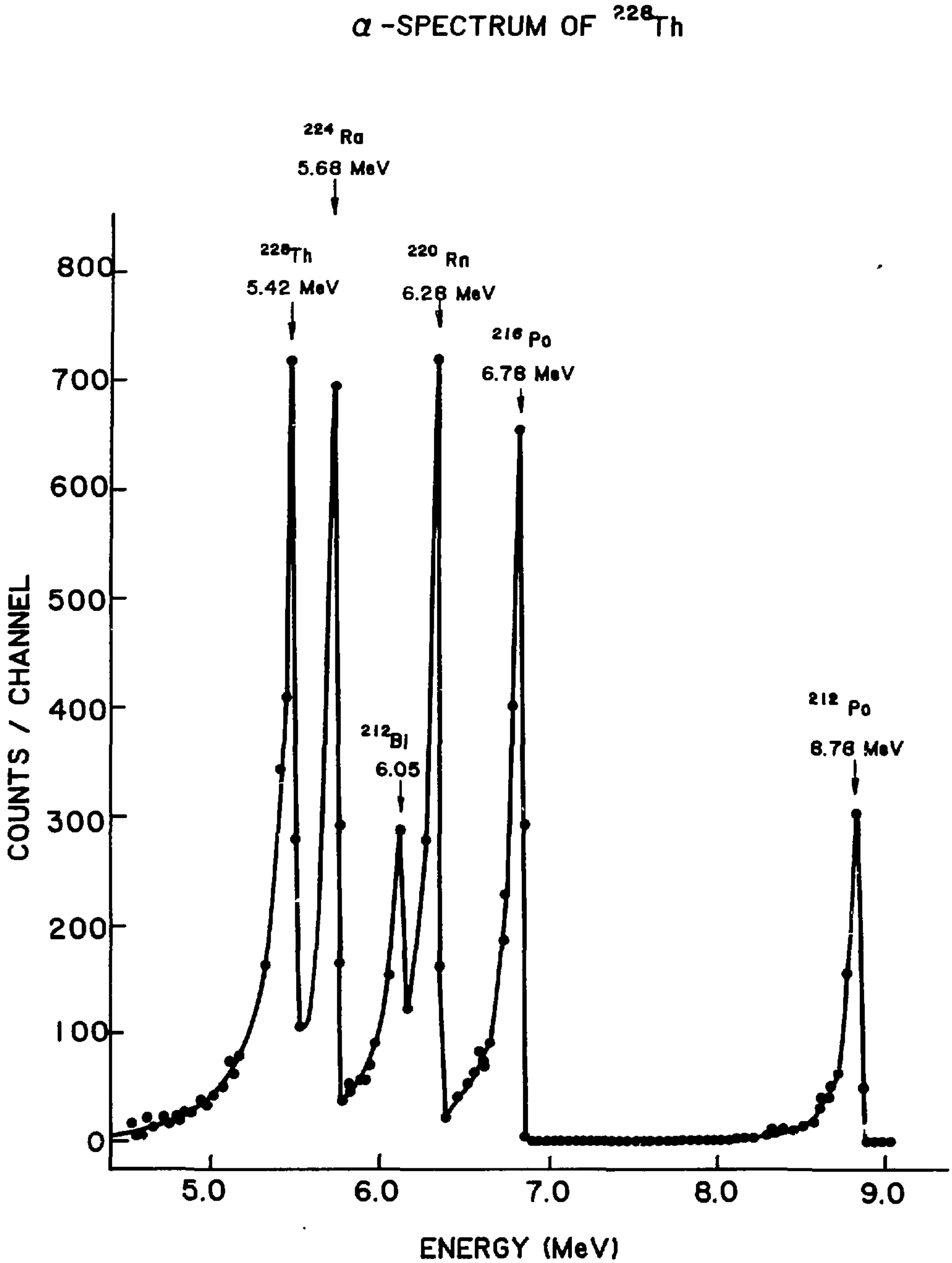


FIGURE 3 Alpha spectrum of Th-228

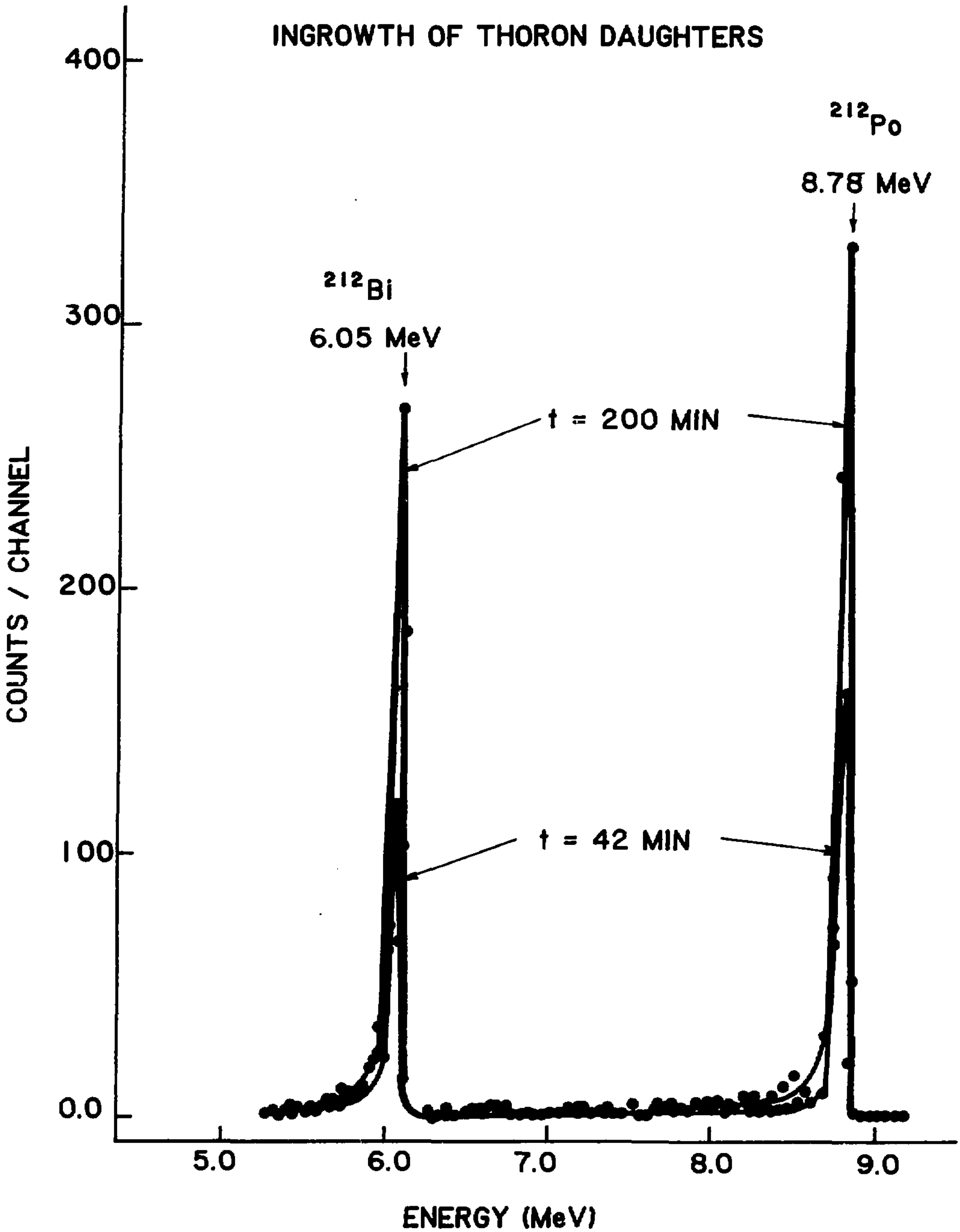


FIGURE 4 Alpha spectra of thoron daughters 42 and 200 minutes after collection

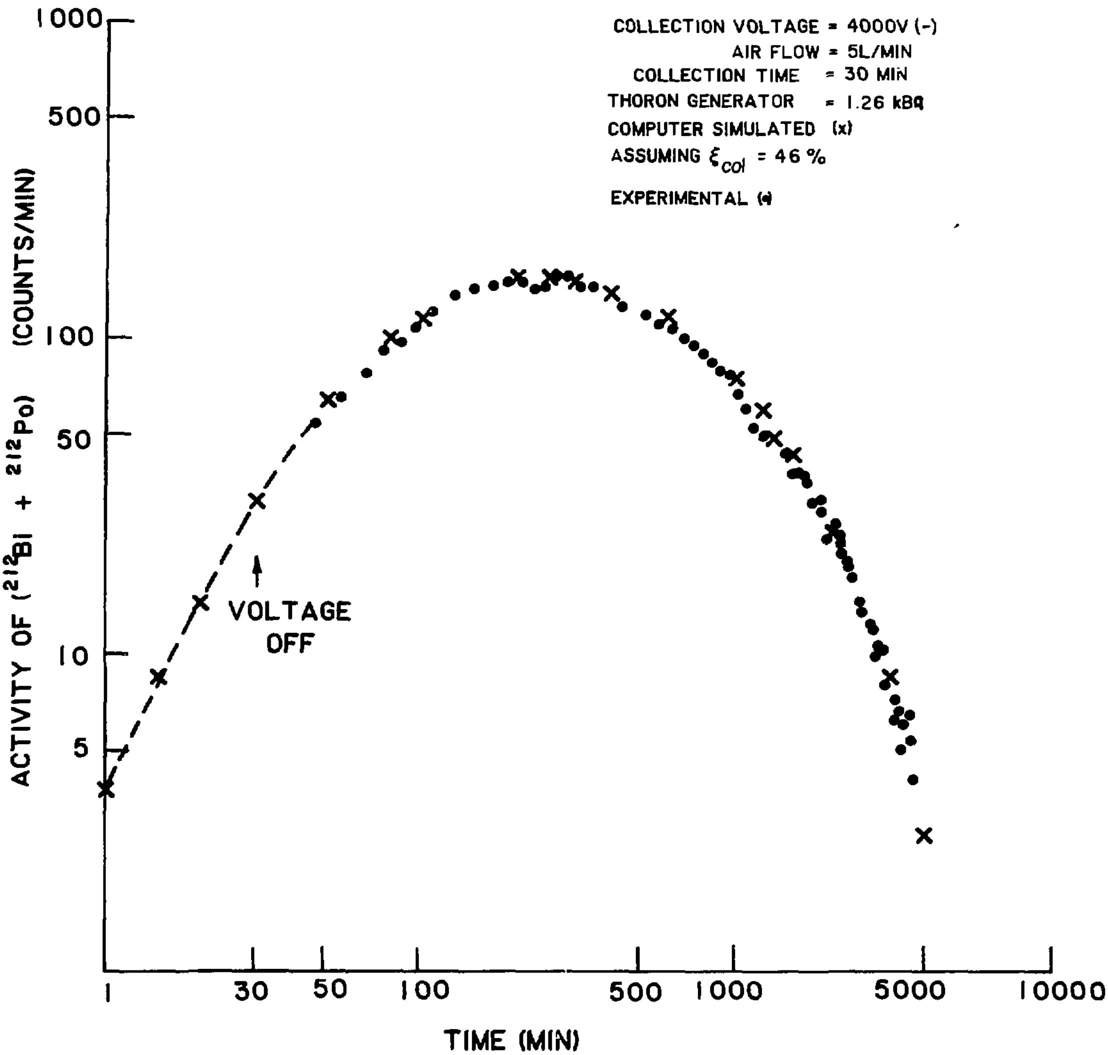


FIGURE 5 Comparison between the measured alpha activity (Bi-212 and Po-212) (·) and the computer generated curve (X)

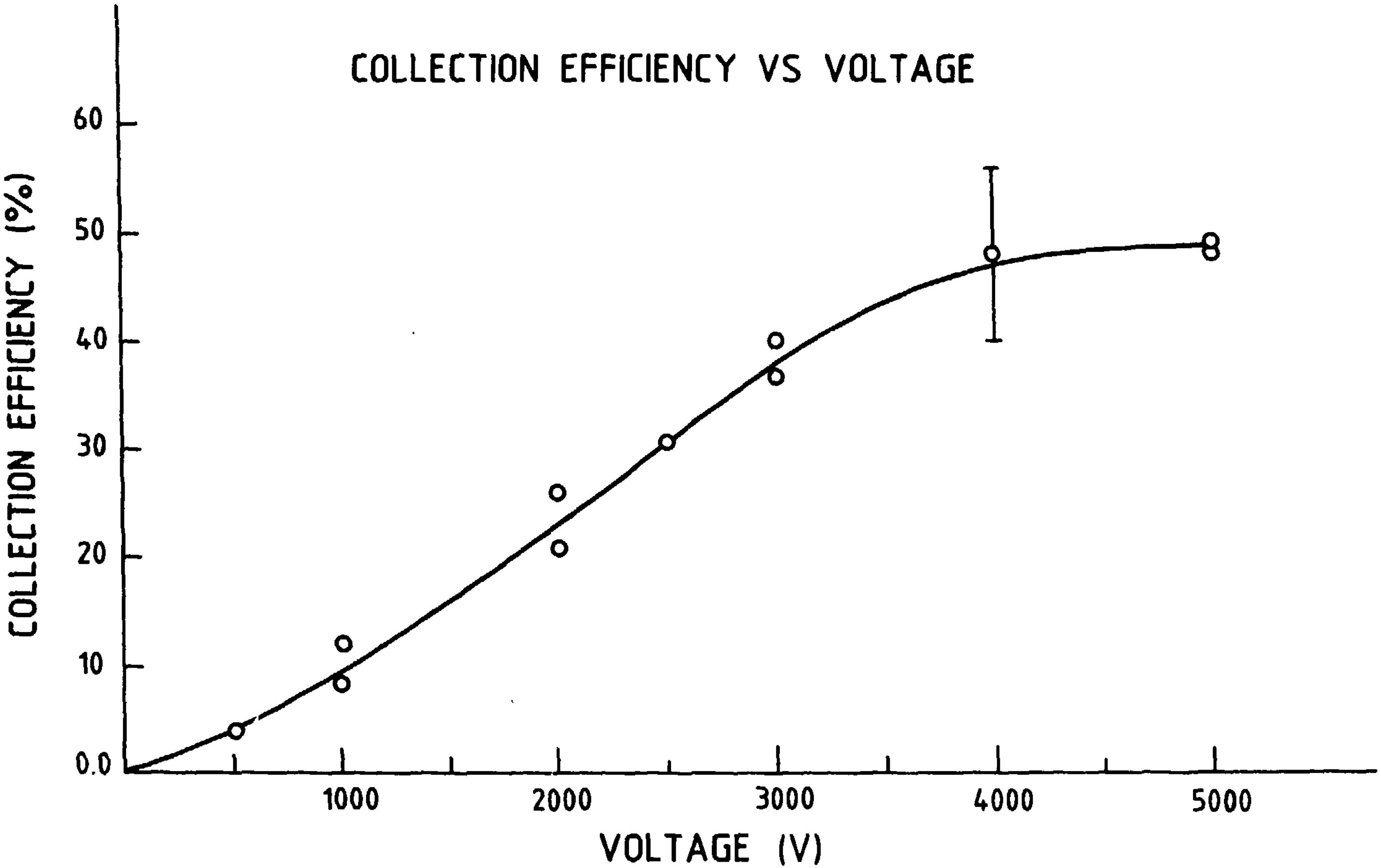


FIGURE 6 Graphic presentation of the relationship between the collection efficiency and collection voltage

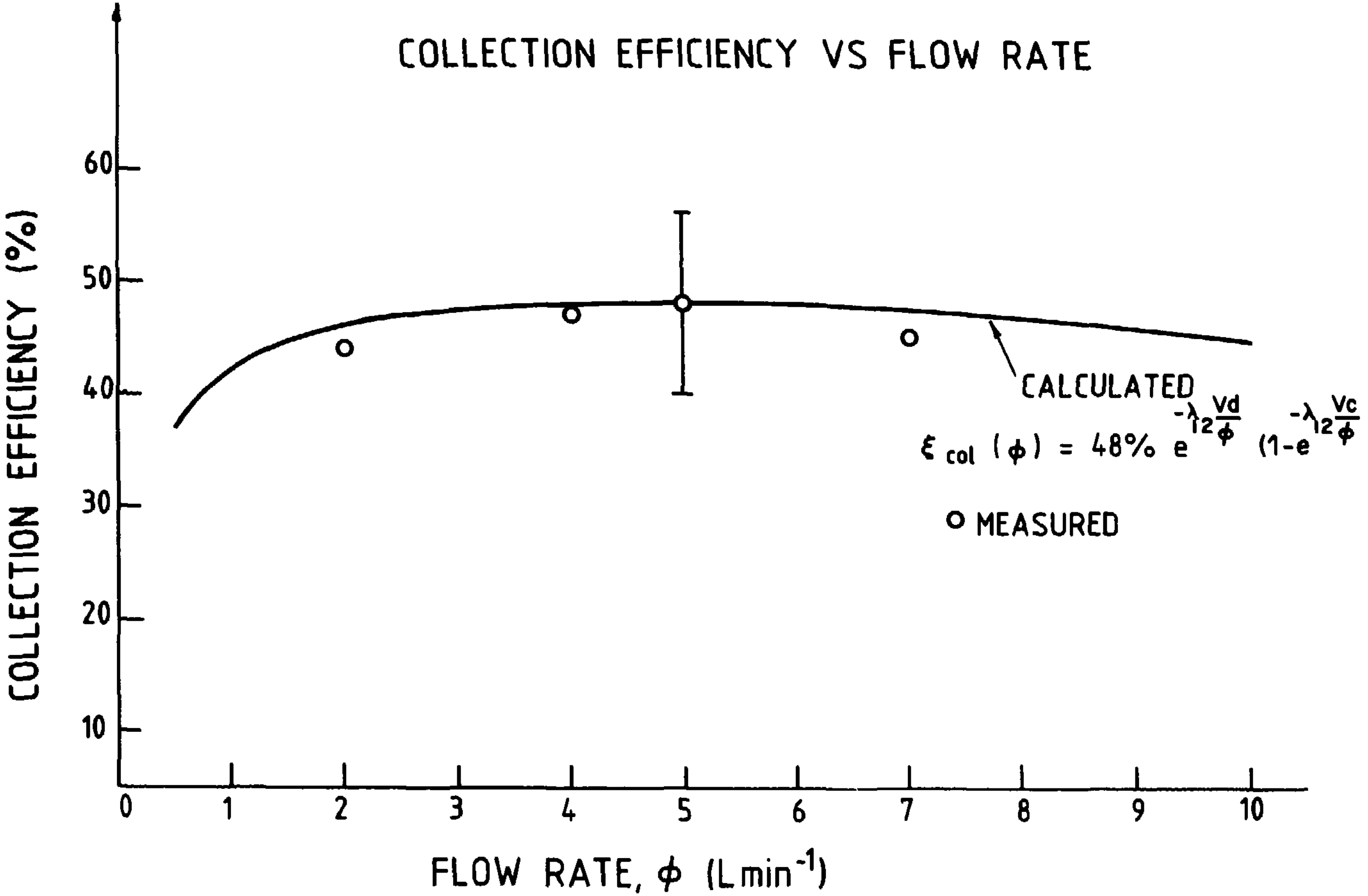


FIGURE 7 Graphic presentation of the relationship between the collection efficiency and flow rate through the chamber

APPENDIX A

In the simulation of thoron-in-breath experiment, it was assumed that the air which passes through the Th-228 solution brings into the chamber $\gamma\, I_o$ thoron atoms per minute where γ is the correction factor due to the decay of thoron in the dead volume (connecting tubing) defined by

$$\gamma = e^{-\lambda_{12} \frac{V_d}{\phi}}$$

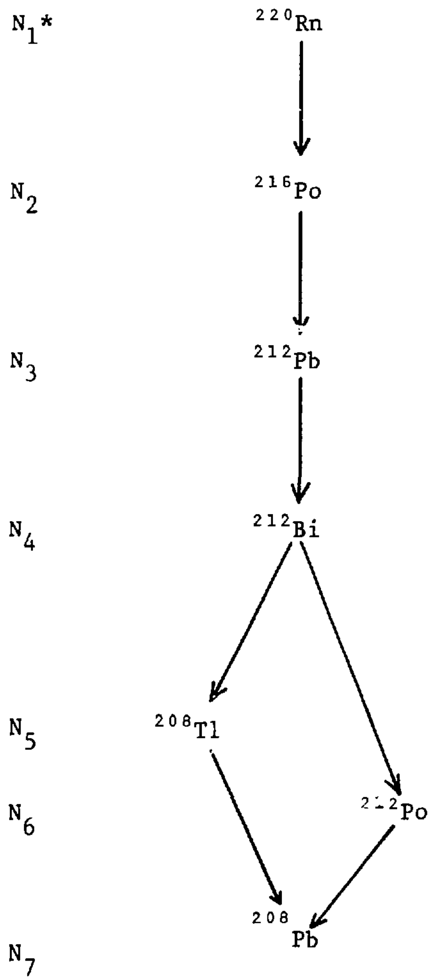
and I_o is the activity of Ra-224 in solution (solution of Th-228 was prepared and calibrated about ten months before it was used for the experiments and therefore the equilibrium between Th-228 and Ra-224 was assumed). Furthermore, it was assumed that thoron is completely removed from the Ra-224 solution.

Some of the thoron atoms will decay in the chamber $(\lambda_{12}\, T_n)$ and some will be lost from the chamber $(\phi/V_c)T_n$. After 10 minutes of passing air containing thoron through the chamber the number of thoron atoms in the chamber will reach the equilibrium value of

$$T_n = \frac{\gamma\, I_o}{\lambda_{12} + \phi/V_c}$$

which represents the initial condition for the collection period. When the voltage is turned on the collection of the charged thoron daughters will begin. As the collection proceeds the collected thoron daughters will also decay according to the scheme in Figure A1.

FIGURE A1



$\lambda^{\dagger}_{12} = 0.76 \text{ min}^{-1}$

$\lambda_{23} = 263 \text{ min}^{-1}$

$\lambda_{34} = 0.0011 \text{ min}^{-1}$

$\lambda_{45} + \lambda_{46} = 0.0115 \text{ min}^{-1}$

$\lambda_{57} = 0.223 \text{ min}^{-1}$

$\lambda_{67} = 1.38 \times 10^{8} \text{ min}^{-1}$

* N_i represents the ith radionuclide

† λ_{ij} represents the rate of decay ($\lambda = \ell n2/T_{1/2}$)

Equations describing the collection are as follows:

$$\frac{dN_1}{dt} = \gamma\, I_o - (\lambda_{12} + \phi/V_c)\, N_1 \qquad \text{(A1)}$$

$$\frac{dN_2}{dt} = \lambda_{12}\, N_1 - (\lambda_{23} + \phi/V_c)\, N_2 \qquad \text{(A2)}$$

$$\left(\frac{dN_3}{dt}\right)_n = \lambda_{23}\, N_2 - (\lambda_{34} + \phi/V_c)\, N_3 \qquad \text{(A3)}$$

$$\left(\frac{dN_3}{dt}\right)_e = \varepsilon_{col} \left(\frac{dN_3}{dt}\right)_n \qquad \text{(A4)}$$

$$\left(\frac{dN_4}{dt}\right)_e = \lambda_{34}\, N_{3,e} - (\lambda_{45} + \lambda_{46})\, N_{4,e} \qquad \text{(A5)}$$

where N_i's represent different radionuclides as defined in Figure A1, e denotes the radionuclide deposited on the electrode, and λ_{ij}'s are the rate constants. In the above equations, it was assumed that, due to the short half-life, Po-216 will decay before it is collected and before it escapes out of the collection chamber (the ratio of ϕ/V_c to λ_{23} is of the order of 0.0005). Therefore we neglected the term $\phi/V_c\, N_2$ in equation (A2). In order to estimate the losses of Pb-212 from the collection chamber ($\phi/V_c\, N_3$) another chamber of 0.5 L capacity was attached to the exhaust of the main chamber. The activity on the mylar disc of the smaller chamber was found to be less than 2% of the activity collected on the mylar disc of the larger chamber. (The activity is probably due to the decay of thoron escaping from the chamber.) Similar results were obtained when Pb-212 atoms were collected by a filter mounted at the exhaust of the collection chamber. Thus, the experiments indicate that most of the available charges are collected on the electrode (only about one half of the electrode surface is covered by the mylar disc). Therefore, the term $N_3 \phi/V_c$ was neglected in further calculations. The equations (A2) and (A3) are then reduced to:

$$\frac{dN_2}{dt} = \lambda_{12} N_1 - \lambda_{23} N_2 \qquad \text{(A6)}$$

$$\frac{dN_3}{dt} = \lambda_{23} N_2 - \lambda_{34} N_3 \qquad \text{(A7)}$$

The quantity (dN_3/dt) represents the net rate of generation of Pb-212 atoms available for collection.

The values for N_3 and N_4 at the end of the collection period are then used as the initial values for the decay of the collected thoron daughters described by the following equations:

$$\frac{dN_{3,e}}{dt} = - \lambda_{34} N_{3,e} \qquad \text{(A8)}$$

$$\frac{dN_{4,e}}{dt} = \lambda_{34} N_{3,e} - (\lambda_{45} + \lambda_{46}) N_{4,e} \qquad \text{(A9)}$$

$$A_{\alpha}(t) = \eta_{det} (\lambda_{45} + \lambda_{46}) N_{4,e}(t) \quad \text{(A10)}$$

The last equation (A10) describes the time dependence of the gross alpha activity and the parameter η_{det} represents the detector efficiency. Both systems of the differential equations were solved numerically by means of a computer. To find the efficiency of the collection of the Pb-212 atoms, ε_{col} was varied until the measured activity agreed with $A_{\alpha}(t)$.

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