Chapter 21

Plate-Out of Radon and Thoron Progeny on Large Surfaces

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The radon and thoron progeny plate-out characteristics of several materials have been investigated in a large (26 m^3) radon/thoron test facility (RTTF). Discs (about 0.5 mm thickness, and 25 mm diameter) were made up of different materials such as metals, plastics, filter materials, fabrics, and powders. The discs were placed on the wall, and/or horizontal trays, of the RTTF for 24 hours. After exposure the materials were removed from the RTTF and their surface α-activity was measured (differential α-count, gross α-count and α-spectrometry). Experiments were also conducted using specially designed Ra-226 and Th-228 reference sources. Plate-out studies were conducted preferentially at low relative humidity and low aerosol concentrations. Significant differences in the measured surface α-activity were found between different materials. This work is relevant to the determination of radon and thoron progeny deposition velocities on, and attachment rates to, large surfaces.

The short-lived decay products of radon and thoron are initially formed in a positively charged atomic state of great diffusivity. These decay products readily attach themselves to small surfaces such as aerosols, and to large surfaces such as walls. Attachment of particles to large surfaces is commonly referred to as plate-out. The degree of plate-out depends on environmental conditions including aerosol concentration, air moisture content, the presence of tracer gases, and most probably, on the properties of the surface of the material. Although theoretical and experimental data are available on the attachment of the radon and thoron progeny to aerosols, less is known on the attachment to walls with regards to the chemical nature and physical characteristics of the wall surface.

Data on the rate of attachment or deposition, i.e., plate-out of radioactive particles on walls can be used to calculate the particle deposition velocity. Deposition rates can be determined experimentally by measuring the surface activity on some samples

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designed for the purpose. These samples usually assume the form of small 'discs' which are attached to the surface where plate-out is to be determined. The samples are used because plate-out measurements carried out directly on the surface of interest may not be practical, and in most cases quite difficult. Furthermore, because of practical considerations, a limited number of materials are used for this purpose under the assumption that the mechanism, or extent, of deposition is independent of the type of material used. As there is insufficient evidence to support or deny this view, work in this area is relevant.

This paper deals with the plate-out characteristics of a variety of materials such as metals, plastics, fabrics and powders to the decay products of radon and thoron under laboratory-controlled conditions. In a previous paper, the author reported on measurements on the attachment rate and deposition velocity of radon and thoron decay products (Bigu, 1985). In these experiments, stainless steel discs and filter paper were used. At the time, the assumption was made that the surface α-activity measured was independent of the chemical and physical nature, and conditions, of the surface on which the products were deposited. The present work was partly aimed at verifying this assumption.

Experimental Procedure

Experiments were conducted in a large (~26 m^3) radon/thoron test facility (RTTF) designed for calibration purposes and simulation studies (Bigu, 1984). A number of different materials were exposed in the RTTF to a radon/radon progeny or thoron/thoron progeny atmosphere. Exposure of the materials was carried out under laboratory-controlled conditions of radiation level, aerosol concentration, air moisture content and temperature. The materials used were in the form of circular 'discs' of the same thickness (~0.5 mm) and diameter (~25 mm), and they were placed at different locations on the walls of the RTTF at about 1.6 m above the floor. Other samples were placed on horizontal trays. Samples (discs) of different materials were arranged in sets of 3 to 4; they were placed very close to one another to ensure exposure under identical conditions. Exposure time was at least 24 hours to ensure surface activity equilibrium, or near equilibrium, conditions.

After a given exposure time, the discs were removed from the RTTF and their surface α-activity was measured as a function of time, up to 2 hours, using standard radiation instrumentation and methods. Counting of samples began 2 min after their removal from the RTTF. For the thoron progeny, samples were counted for 15 min followed by 5 min counts with 1 min interval between counts. For the radon progeny two different counting routines were used, namely: 2-min counts with 1-min interval, and 5-min counts with 1-min interval. The former counting routine was intended for more precise Po-218 measurements. In addition, gross α-count (30 min counts) and α-spectrometry measurements were also done.

The surface α-activity on the samples for each exposed set of samples was compared. Each set of samples was exposed, and measured, a minimum of about five times to improve overall statistics. Experiments were conducted in the temperature range 19-22^{o}C and at low relative humidity (10 to 40%). The aerosol concentration, mainly

natural atmospheric aerosols, was kept low, i.e., 5×10^2 to 9×10^3 cm^{-3}. No attempt was made to maintain the aerosol concentration at any particular level at these low aerosol concentrations. The values attained were those due to controlled make-up unfiltered air, purposely leaking into the test facility used for the experiments. The aerosol concentration varied with barometric pressure.

The range of radiation conditions in the RTTF was as follows. Radon gas concentration, [Rn-222]: 40-200 pCi/L (1.48×10^3 - 7.4×10^3 Bq/m^3; radon progeny Working Level, WL(Rn): 20-400 mWL; thoron gas concentration [Rn-220]: 350-1700 pCi/L (1.295×10^4 - 6.29×10^4 Bq/m^3); thoron progeny Working Level, WL(Tn): 0.15-14 WL. (The square brackets are used here to denote activity concentration.)

The materials investigated included several metals (copper, Cu; aluminum, Al; stainless steel, SS; and galvanized steel, GS); filter materials (Glass Fiber, Millipore and Varsopor); plastic materials (Plexiglas); 'powders' (activated carbon, talcum); and other materials such as sandpaper, emery cloth and several fabrics. Activated carbon and talcum powder samples were prepared by depositing a thin layer (~0.5 mm) of the powder on filter paper on which some binding material (glue) had been added to its surface to retain the powder.

Further plate-out studies were conducted using radon progeny and thoron progeny reference sources, models Rn-190 and Th-190, respectively, manufactured by Pylon Electronic Development (Ottawa), hereafter referred to as Pylon sources, for simplicity. These are small cylindrical containers (<40 cm^3 volume) provided with a Ra-226 source or Th-228 source. The containers can be opened at their base and some suitable material can be placed in it for exposure purposes (Vandrish et al., 1984). The Ra-226 and Th-228 sources decay, respectively, to Rn-222 and Rn-220 which in turn, decay into their progeny. In this respect, the above sources can be considered miniature RTTFs quite suitable for plate-out studies, in which air flow pattern effects are minimized.

Radioactivity measurements in the above case were similar to the measurements indicated for the test facility, except that only one sample per source could be investigated at a time. In order to facilitate the study, four sources were used, two Ra-226 sources and two Th-228 sources. The strengths of the two Ra-226 sources were different. The same applies to the two Th-228 sources.

Because the reproducibility of α-particle counting with the Pylon sources was quite good, this study was conducted as follows. Four identical samples, i.e., same material, were placed in the decay product standard sources for a period of 24 h at a time. The four samples were then removed and measured. Gross α-particle count and α-spectrometry analysis of the samples were done.

The bulk of the data presented here was obtained in a follow-up to a plate-out study reported earlier elsewhere (Bigu and Frattini, 1985). It should be noted that some qualitative and quantitative differences between earlier studies and the most recent follow-up may be observed depending upon experimental conditions. Although the conditions of the surface of the material, environmental conditions and air flow patterns are believed to play a significant role in plate-out phenomena, their effect on the latter, and their relationships, are far from being understood. Furthermore, the chemical nature of the material, and of the radon and thoron progeny, could play a significant role in plate-out phenomena.

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Results and Discussion

The data obtained in this study are presented in Figures 1 to 5, and Tables 1 to 3. For simplicity, the data will be presented according to the location where they were obtained, namely the RTTF, and the Pylon reference sources.

Large Radon/Thoron Test Facility (RTTF). Figures 1 to 5 show the surface α-activity measured on several materials exposed to a radon progeny atmosphere (Figures 1 and 2), and to a thoron progeny atmosphere (Figures 3 to 5).

Figure 1 shows no significant difference between the three metals, namely: aluminum (Al), copper (Cu), and galvanized steel (GS), exposed to a radon progeny atmosphere. Figure 2 shows a significant plated-out activity difference between Fiberglas filters, and coarse sandpaper and emery cloth exposed to a radon progeny atmosphere. No appreciable difference was found between sandpaper and emery cloth.

Figure 3 shows a significant difference in thoron progeny activity plated-out on laboratory coat cotton cloth samples and Millipore filters (0.8 μm), and emery cloth. Differences between the two latter sample materials are not clear in these measurements.

Figures 4 and 5 show significant differences in thoron progeny activity plated-out on cotton cloth and emery paper (Figure 4), and cotton cloth and Fiberglas filters (Figure 5). Differences between the other pair of materials were less pronounced or difficult to ascertain (<7%).

As expected, Figures 1 and 2 show that the surface α-activity on the materials examined decreased with increasing time after exposure. The approximate half-life corresponding to the surface α-activity was in the range of 35 to 50 min. These values roughly agree with the 'combined' half-life of the short-lived decay products of radon. The differences in the decay curves for different materials and for a given material under different environmental conditions, i.e., radiation level, temperature, relative humidity and aerosol concentration are mainly due to the different radioisotope make-up about the surface of the materials investigated. The radioisotope make-up very much depends on the radon progeny disequilibrium ratios, i.e., [Pb-214]/[Po-218] and [Bi-214]/[Po-218]. The radon progeny disequilibrium ratios are highly dependent on the air flow characteristics, relative humidity and aerosol concentration in the RTTF.

Figures 1 and 2 also show a rapid decrease in α-activity during the first few minutes after removal of the samples from the RTTF followed by a less pronounced decrease in the activity. This behaviour is due to the rapid decay of Po-218 with a half-life of about 3 min.

Figures 3 to 5 show that the surface thoron progeny α-activity remained fairly constant during the counting period, i.e., about 100-min. This result indicates that the α-activity measured on the surface of the materials was mainly due to Bi-212, and Po-212 in equilibrium with Bi-212, which was in equilibrium with the relatively long-lived, 10.6 hour half-life, Pb-212. Hence, Bi-212 decayed with the half-life of Pb-212.

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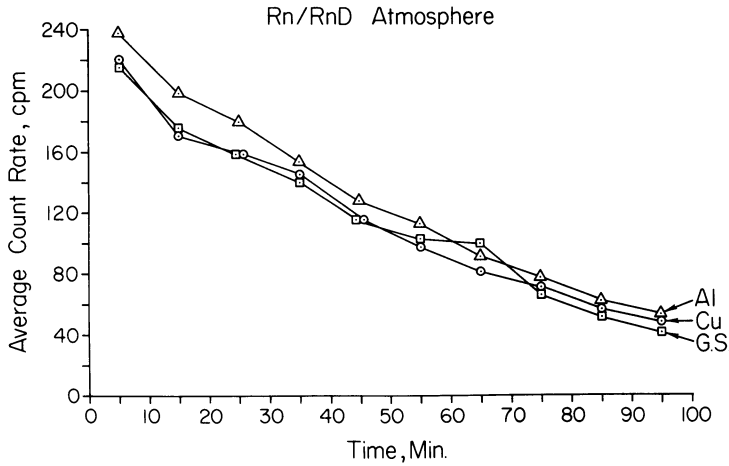

Figure 1 Alpha activity versus time from radon progeny plated-out on several materials.

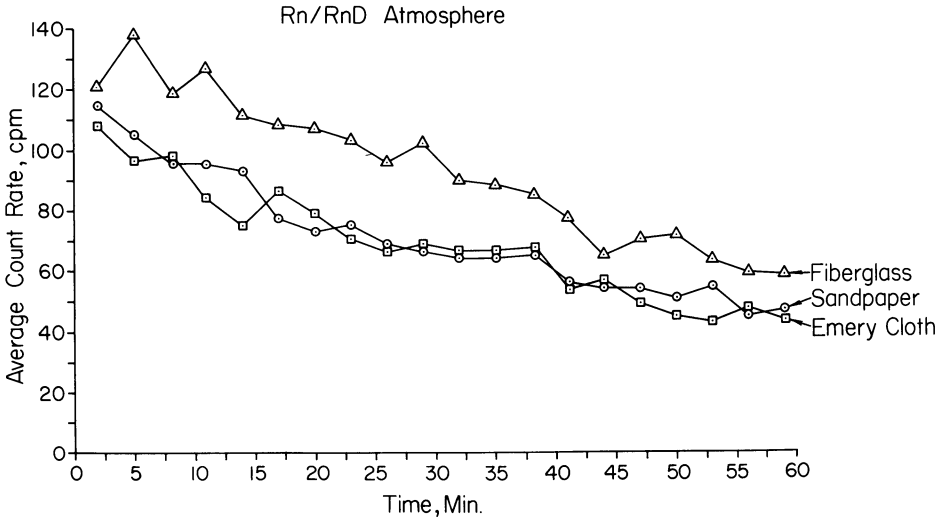

Figure 2 Alpha activity versus time from radon progeny plated-out on several materials.

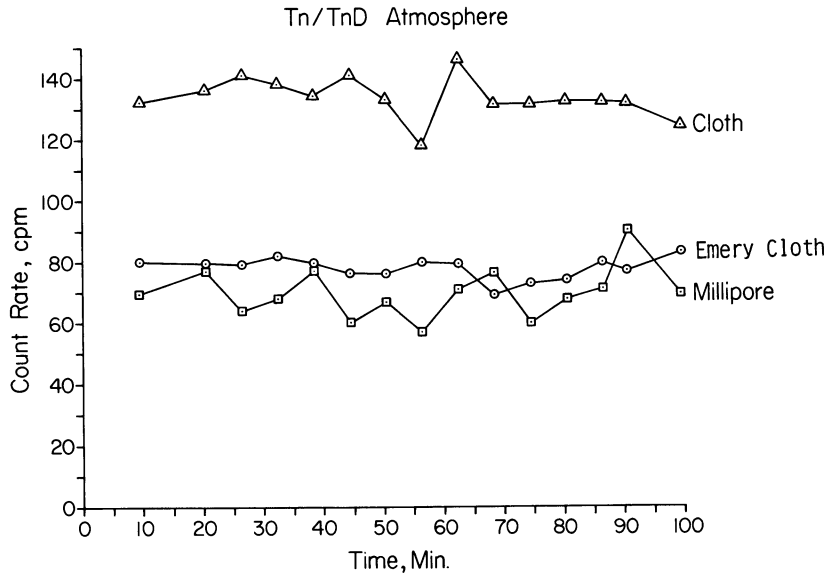

Figure 3 Alpha activity versus time from thoron progeny plated-out on several materials.

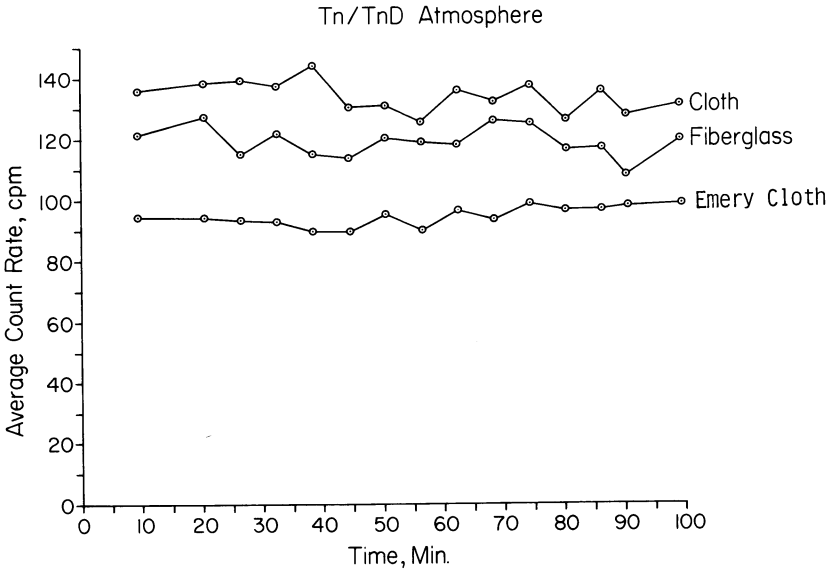

Figure 4 Alpha activity versus time from thoron progeny plated-out on several materials.

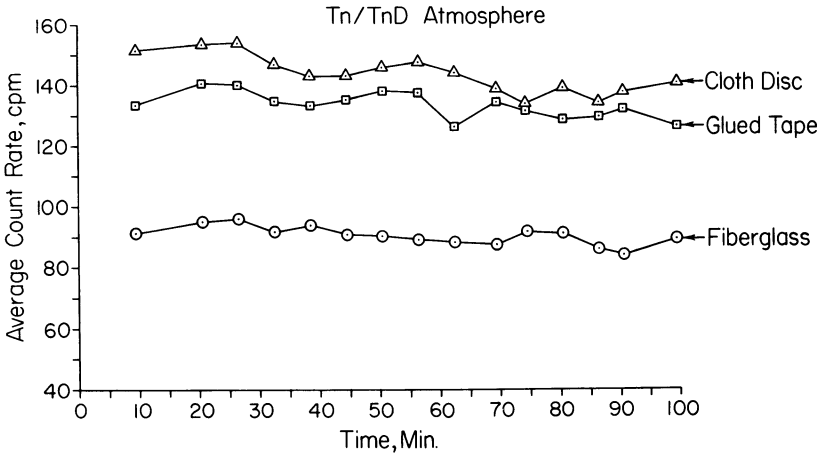

Figure 5 Alpha activity versus time from thoron progeny plated-out on several materials.

Table I Thoron progeny activity plated-out on different materials

Material	Gross α-particle Count	R*	Qualitative Agreement with Previous Data	Remarks
Copper	9521±1216	0.947	Yes	N/A
Galvanized steel	10051±1972	0.999		
Aluminum	10052±2093	1.000		
Copper	9469±3393	0.976	Partial	N/A
Plexiglas	9508±3580	0.980		
Aluminum	9701±2822	1.000		
Plexiglas	8914±2330	0.898	Marginal	N/A
Aluminum	9854±3286	0.993		
Galvanized steel	9926±3297	1.000		
Millipore	5199±1101	0.730	Yes	See Fig. 3
Emery cloth	5208±721	0.731		
Cotton cloth	7122±903	1.000		
Fiberglas	5820±447	0.818	Yes	See Fig. 5
Tape	7041±748	0.989		
Cotton cloth	7115±750	1.000		
Emery cloth	5414±445	0.667	Yes	See Fig. 4
Fiberglas	6973±714	0.860		
Cotton cloth	8110±230	1.000		
Carbon powder	5436±519	0.786	Yes	N/A
Cardboard	5823±628	0.842		
Talcum powder	6918±77	1.000		

* R represents the ratio of the activity of the material to the maximum activity measured within each set of three materials.

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Table II- Radon progeny plate-out on different materials using Pylon source Standards model Rn-190

Material	N_1	N_2	$N_{1,2}$ +	N_3	N_4	$N_{3,4}$ +	$N_{1,2,3,4}$ +	S.R.*
Carbon	49198	24735	73933	115678	60133	175801	249734	-
Copper	20127	11056	31183	42966	23537	66503	97686	4.39
Cotton cloth	20763	11025	31788	40024	21789	61813	93601	2.94
Nuclepore	21102	10954	32056	40901	21687	62588	94644	4.02
Fiberglas	19248	10201	29449	39238	21348	60586	90035	3.70
Emery cloth	18486	9872	28358	35357	19479	54836	83194	3.37
Sandpaper	16539	8834	25373	32701	17405	50106	75479	3.30

* S.R. stands for spectral ratio, i.e., the α-particle count ratio between $^{214}Bi(\equiv {}^{214}Po)$ and ^{218}Po.

+ $N_{1,2} = N_1 + N_2$; $N_{3,4} = N_3 + N_4$; and $N_{1,2,3,4} = N_{1,2} + N_{3,4}$

Note: the symbol N denotes gross α-count. S.R. refers to N_1 and N_3 only.

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Table III- Thoron progeny plate-out on different materials using Pylon source standards model Th-190

Material	N_1	N_2	$N_{1,2}^{+}$	N_3	N_4	$N_{3,4}^{+}$	$N_{1,2,3,4}^{+}$	S.R.*
Carbon	16620	16024	32644	431120	422542	853662	886306	-
Fiberglas	11586	11495	23081	262294	261282	523576	546657	1.673
Copper	11258	10993	22251	259513	259678	519191	541442	1.769
Nuclepore	11543	11485	23028	258653	257761	516414	539442	1.768
Cotton cloth	11056	11025	22081	243801	242296	486097	508178	1.704
Emery cloth	10123	9982	20105	224460	224178	448638	468743	1.697
Sandpaper	9807	9781	19588	215702	214943	430645	450233	1.678

* S.R. stands for spectral ratio, i.e., the α-particle count ratio between ^{212}Po and ^{212}Bi.

+ $N_{1,2} = N_1 + N_2$; $N_{3,4} = N_3 + N_4$; and $N_{1,2,3,4} = N_{1,2} + N_{3,4}$

Note: the symbol N denotes gross α-count.

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In order to verify the results reported above, an independent series of measurements in the large RTTF was carried out. Samples were exposed to a thoron progeny atmosphere, and surface gross α-particle activity was measured this time for periods of 30 min. The environmental conditions during this experimental phase were in the following range. Temperature: 24-27°C, relative humidity: 40-55%, aerosol concentration: $1.2 \times 10^3 - 3.4 \times 10^3$ cm^{-3}. Some of the data obtained are reproduced in Table I.

Table I shows that, in most cases, qualitative agreement with previously reported data is attained (Bigu and Frattini, 1985). Absolute values, of course, change according to experimental conditions. The large standard deviations quoted in Table I arise from experiments carried out under significantly different radon (or thoron) progeny concentrations. The values quoted in the Table are average values calculated from several measurements conducted on different days under different experimental conditions. Data for each set of materials have been arranged according to increasing surface α-particle activity measured on the material. The variable R represents the ratio of minimum to maximum surface activity measured on each set of three different materials. Gross α-particle 30-min counts are given as absolute α-counts by dividing the instrument reading by its α-particle counting efficiency. Each set of materials was exposed and measured several times on different days, and an average value (shown in the Table) was calculated, as indicated above.

The effect of surface electrostatic charge on a material on the attachment of the decay products of radon has been known since pioneering work on atomic structure by Rutherford. Extensive research into this area for the radon and thoron progeny has been conducted in this laboratory for environmental monitoring purposes. Several authors have reported on the effect of electrostatic charge on the collecting characteristics of copper for the radon progeny for exploration purposes (Card and Bell, 1979).

In a series of recent thoron progeny experiments in a small and large RTTF, dramatic differences in the α-activity collected on copper discs were observed when a positive charge, a negative charge, or a zero charge, i.e., discs at ground potential, was applied. Discs positively charged (~+270 V) collected 1.4 to 2.8 (ave.:2.0) times more α-activity than discs at zero potential. Discs negatively charged (~-270 V) collected 11 to 21 (ave.:17) times more α-activity than discs at zero potential. The α-activity ratios between discs charged at -270 V and +270 V were in the range 4.5 to 15.5 (ave.: 9.5). The above figures, and figures for the radon progeny obtained in the past, varied quite substantially with environmental conditions in a rather complex and little understood fashion.

The data on the electrostatic effect discussed above is relevant in the context of this paper because certain materials can esily acquire electrostatic charge which could, therefore, alter or mask their plate-out characteristics to the decay products of radon and thoron.

Pylon Radon (Thoron) Progeny Reference Sources. Plate-out studies using the Pylon reference sources are shown in Tables II and III. In this case the materials (discs) were placed in the reference sources

where they were exposed for 24-h, after which they were analyzed by gross α-particle count and α-spectrometry. Two counts, and spectra, were taken (1000 s counting time) on each sample exposed to each reference source. Measurements were carried out 1-min and 40-min after exposure. The first count and spectrum using the radon progeny reference source were intended to study the deposition of the short-lived radioisotope Po-218. Ambient temperature and relative humidity at the time the samples were placed in the reference sources were carefully noted. The range of values was as follows: 20.4-23.1°C (temperature), and 10.7-33.7% (relative humidity).

Tables II (radon progeny) and III (thoron progeny) present some of the data obtained. In these Tables N_1 and N_2, obtained with the weak reference sources, stand for the gross α-particle count measured 1-min and 40-min after exposure, respectively. The symbols N_3 and N_4 represent the same as N_1 and N_2, respectively, but for the strong references sources. Furthermore, in the above Tables: $N_{1,2} = N_1 + N_2$, $N_{3,4} = N_3 + N_4$, and $N_{1,2,3,4} = N_{1,2} + N_{3,4}$.

Tables II and III show that the α-activity plated-out on the materials investigated can roughly be divided into three groups, namely, low activity (emery cloth and sandpaper), medium activity (Fiberglas filter, Nuclepore filter, cotton cloth and copper), and high activity (activated carbon deposited on suitable material).

The only data from Tables II and III that can be compared directly with Figures 1 to 5, and Table I are that corresponding to the materials emery cloth, Fiberglas filter and cotton cloth. It is seen that the data corresponding to the radon progeny in Table II agree, qualitatively speaking, with Figure 4, for the thoron progeny. There is agreement between the thoron progeny data of Figure 4, Table I and Table III for emery cloth as having the lowest plated-out activity. However, there is disagreement between cotton cloth and Fiberglas. This difference shows with both the weak and strong reference sources (see Table III). This experimental observation seems to rule out spurious or systematic errors.

The spectral ratios (S.R.) given in Tables II and III show that Nuclepore filters and copper discs have the highest S.R., indicating that less degradation of the spectra, and hence, highest energy resolution, occurs. It should be noted, that except for cotton cloth (Table II), there seems to be a correlation between the S.R. and the activity plated-out on the material.

The α-particle activity measured on activated carbon samples is much higher than that corresponding to any other material (see Tables II and III). This result was expected because of the well known adsorptive properties of carbon for radon and thoron. However, the activated carbon data on Tables II and III is in sharp disagreement with previous, and numerous, data obtained in the large RTTF (see Table I). This topic is under investigation. It should be noted that there are substantial quantitative differences between the behaviour of the radon and thoron progeny relative to activated carbon.

The data presented here suggest some qualitative and/or quantitative differences in behaviour between the radon progeny and thoron progeny relative to some materials, and for some materials relative to either the radon progeny or the thoron progeny.

Other data, not presented here, also suggest that the plate-out characteristics of some materials depend on air moisture content,

aerosol and dust concentration and size distribution and other variables in a rather complex, not clearly understood, fashion. Hence, substantial differences may arise when the same set of materials are investigated under different environmental conditions.

At present, only some speculation can be offered regarding the cause for the observed plate-out characteristics of different materials, as estimated from surface α-activity measurements. Some factors that might account for the observed data have been indicated elsewhere (Bigu and Frattini, 1985).

Not previously considered in the above reference is α-particle self-absorption phenomena. However, attempts are at present being made to estimate radon and thoron progeny deposited on the surface of materials by γ-spectrometry and gross γ-count in order to eliminate self-absorption effects.

Conclusions

Significant differences in the radon and thoron progeny plate-out characteristics of different materials have been found. As radon and thoron progeny deposition velocities are normally determined from surface α-activity measurements using some material as a collector of activity, the choice of collecting surface is important in view of our results. The arbitrary choice of a material as an 'activity' collector may lead to substantial errors in the determination of deposition velocities, and other variables, if the data obtained for this particular material are to be used as representative of the deposition velocity on a large surface made of a different material. It should be noted that there are some practical difficulties in determining radon and thoron progeny activity directly on the surface of interest.

Some of the data reported here, and unpublished data, seem to indicate that there might be some qualitative difference between the behaviour of the radon progeny and thoron progeny towards a given material.

The underlying physical and/or chemical mechanisms responsible for the differences observed between the radon progeny and the thoron progeny as related to different materials are not clearly understood. Finally, it should be pointed out that the main thrust in this paper was to determine differences in surface α-activity measured on different materials with the same geometrical characteristics exposed to identical radioactive atmospheres. The calculation of deposition velocities and attachment rates, although it follows from surface α-activity measurements, was not the intent of this paper. This topic is dealt with elsewhere (Bigu, 1985).

The nature of the data discussed here is in part consistent with recent work by other authors who suggested a sticking coefficient of less than unity, under certain conditions, for the attachment of Po-218 to monodisperse aerosols (Ho et al., 1982), and to surfaces (Holub, 1984).

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