J. SCI. INSTRUM., 1963, VOL. 40

A scintillation counter for the measurement of radon concentration in air

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MS. received 24*th June* 1963, *in revised form* 26*th July* 1963

A method is described for measuring the radon content of three litre samples of atmospheric air without pretreatment. The apparatus used consists of a 12 in. photomultiplier with ZnS(Ag) phosphor and counting chamber. A measured efficiency of 38%, when the phosphor is negatively charged, is in good agreement with theory. The ultimate sensitivity is governed by the background which is equivalent to a concentration of 5×10^{-14} c l^{-1}.

1. Introduction

This paper describes a method developed for an investigation into the radon concentrations in dwelling houses, offices and industrial premises in the Borough of Finsbury. For this survey it was essential that the sample volume be small (not more than two or three litres) and that the sampling method be simple and unobtrusive. In addition it was required that activities as low as 10^{-14} c l^{-1} should be measured with reasonable accuracy.

Methods involving the immediate separation of the decay products by filter paper (Dawson 1952) and their subsequent assay by a suitable detector are still used but cannot be regarded as accurate because the efficiency of separation varies with particle size (Anderson, Mayneord and Turner 1954). This difficulty can be surmounted if, say, electrostatic precipitation is used but, in any case, inaccuracies are likely because the method is based on the assumption of equilibrium between atmospheric radon and its products which has been shown to be unjustified even in closed rooms by Běhounek and Majerová (1956). Consequently, in order to measure the radon concentration accurately it is necessary to take a given sample of air and wait until equilibrium has been attained before measuring the activity of the radon and its decay products or the latter alone if these are to be separated.

A number of workers have employed scintillation counting methods. Bryant and Michaelis (1950 unpublished) and Simpson and Cowper (1953 unpublished) admitted the air sample into a chamber of two or three litres capacity containing a small negatively charged aluminium foil on which the decay products were deposited and counted using a ZnS phosphor and photomultiplier mounted behind the foil. The overall efficiency achieved by this method is rather low, the radon activity making a negligible contribution and the collection of the two alpha emitting daughter products being about 50% with a further 50% counting loss. Taysum and van Dilla (1955) and Lucas (1958) describes more efficient methods in which the volume of the air sample is small ($\simeq$ 100 cm^3) necessitating the initial separation of the radon over cooled charcoal and remixing with a smaller volume of air.

The scintillation method offers the advantages of simplicity and comparatively trouble-free electronics, the main difficulties apart from low efficiency being associated with background. The arrangement described in this paper employs a scintillation detector in which samples of 2·7 l. of air can be assayed without pretreatment except for simple filtering and drying. The efficiency has been improved by employing a large (12 in.) photomultiplier and both daughter products and radon are counted.

2. Construction and method

The photomultiplier (EMI Type 9545, window diameter 300 mm, cathode diameter 250 mm) is enclosed in a light-tight cubical box (figure 1) one end of which is a $\frac{1}{2}$ in. brass plate

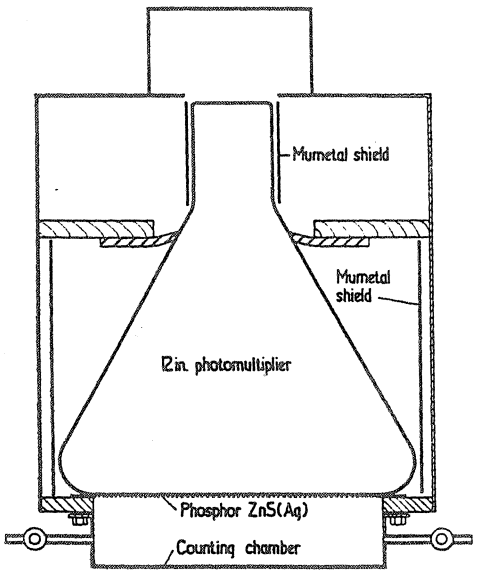

Figure 1. Arrangement of photomultiplier and counting chamber.

accurately machined flat with a 10 in. hole. The photomultiplier window, ground flat, rests on this plate and overlaps the hole by $\frac{1}{2}$ in. all round. The flanged cylindrical counting chamber of $\frac{1}{8}$ in. brass and 4 cm deep is bolted to the underside of the plate. The phosphor ZnS(Ag) is deposited on a Perspex disk held against the photomultiplier window. The effective depth of the chamber is 5·4 cm and its volume 2·7 l. The seal of the photomultiplier face against the brass plate is maintained by the weight of the multiplier together with slight pressure applied through a thick ring of foam rubber at the neck of the tube. Some difficulties were experienced with this seal at atmospheric pressure so that all measurements were made with the chamber at 700 torr pressure.

The air samples were collected in three litre flasks. The

procedure for measuring the radon concentration was to evacuate the chamber and admit the air sample from the flask through a drying tube and glass wool filter, bringing the pressure up to 700 torr by using a Neoprene balloon, fitted inside the flask and connected to a separate outlet. After waiting about two hours for the radon to come into equilibrium with its products, the count rate due to the α-particles from radon and its two α-emitting products RaA and RaC' was measured over a period of two to four hours. Corrections due to the background and decay of the sample after collection were necessary.

Conventional electronics, an amplifier (gain = 90) followed by a voltage discriminator and scaler were employed.

3. Background

As it was necessary that the background be stable, the amplifier and discriminator settings were chosen to ensure that the contribution from cosmic rays and noise pulses was negligible.

An interesting effect was observed when investigating what were thought to be purely photomultiplier noise pulses, in that at sufficiently high gain the count rate with the face of the photomultiplier pointing upwards was more than double that with the face downwards. This effect was present whether the phosphor was in position or not. In addition, a shielding effect was observed when layers of lead were placed above the counter (figure 2). This increase in count rate could only be

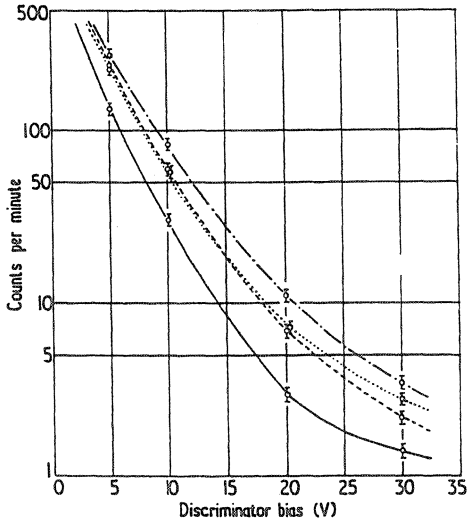

Figure 2. Discriminator bias curves of noise and cosmic ray pulses at high photomultiplier gain, (e.h.t. 2000 v).
———— Photomultiplier face downwards (no shield)
—·—·—· Photomultiplier face up (no shield)
····· Photomultiplier face up (1in. lead shield)
– – – – – Photomultiplier face up (5in. lead shield)

ascribed to the Čerenkov effect produced by cosmic ray particles on the glass face ($\simeq$ 15 mm thick) of the multiplier. The cone of light produced by cosmic rays travelling mainly in the downward direction would move towards the photocathode in one case and away from it in the other. In practice, the contributions due to noise pulses and cosmic rays were measured for the working region of the counter by removing the phosphor when it was found to be negligible (less than 0·5 counts per hour) compared with the main background.

The background was determined by measuring count rates with the chamber evacuated and also filled with aged air. The backgrounds obtained were found to be the same with both methods of measurement indicating that the source was primarily α-contamination of the phosphor.

ZnS(Ag) screens of different thicknesses, 5 mg cm^{-2}, 8 mg cm^{-2}, 10 mg cm^{-2} and 12 mg cm^{-2}, were tried initially to determine which thickness would give the best sample to background ratio. The grain sizes of these screens were graded according to thickness in order to give a layer of single crystals. It was found that a thickness of 10 mg cm^{-2} represented the optimum. Figure 3 shows the discriminator

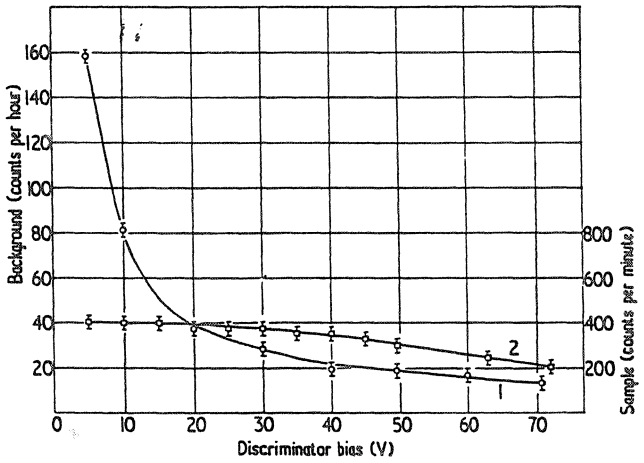

Figure 3. Discriminator bias curves for background (curve 1) and sample (curve 2). Photomultiplier e.h.t. 1700 v.

bias curves for sample and background for this screen, the bias chosen for all subsequent measurements being 40 v. At this setting the background was 20 $\pm$ 1 counts per hour, and this had been redetermined regularly over the past two years remaining at this figure within the limits stated.

4. Calibration

The counter was calibrated in terms of known radon activity using the de-emanation method described by a number of authors, e.g. Evans (1933). A known amount of radon was boiled off from a standard solution of RaCl, mixed with aged air and then passed through a needle valve and drying tubes at a pressure of about 100 torr to the chamber. Further air was then added to bring the pressure up to 700 torr. The calibration was repeated with different quantities of radon, obtained by allowing the activity in the solution to grow for different periods of time.

Some difficulties were experienced in obtaining consistent values of the calibration, successive determinations yielding lower values of the count rate due to the same quantity of radon. This was found to be due to silicon oil, used between the Perspex disk and the photomultiplier face, seeping out and causing either a deterioration in the light collecting efficiency or possibly absorbing radon. When the use of oil was discontinued consistent results were obtained. The calibration was found to be independent of temperature between 15° c and 25° c and also of total pressure between 700 torr and 760 torr.

The mean value of the calibration figure was 150 counts per hour for 1pc in the chamber, that is for a concentration of 0·37 pc l^{-1}. The sample count rate is equal to the background at 0·05 pc l^{-1}. At this concentration the accuracy for a two hour count is to $\pm$20%.

5. Efficiency

Taking the efficiency as 100% when the three α-particles from Rn, RaA and RaC′ are recorded, then the calibration figure obtained experimentally corresponded to an efficiency of 37·4%.

In order to check this with the counter geometry, assume that the radon is uniformly distributed throughout the gas and that the two daughter products are distributed uniformly over the walls and roof of the chamber and over the phosphor.

Considering first the radon, for which the α-particle range R_1 is slightly less than the depth H of the chamber, the α-particles reaching the phosphor and originating at a height h above it are those contained within a solid angle $2\pi(1 - h/R_1)$. The efficiency for radon alone is given by

$$\frac{1}{4\pi H}\int_0^{R_1} 2\pi\left(1 - \frac{h}{R_1}\right)dh = \frac{R_1}{4H}. \qquad (1)$$

For the RaA and RaC′ deposited on the walls at a height h, the α-particles reaching the phosphor are those contained within the solid angle $\pi(1 - h/R)$. The mean probability of an α-particle from RaA deposited on the walls, range $R_2 < H$, reaching the phosphor is then

$$\frac{1}{4\pi H}\int_0^{R_2} \pi\left(1 - \frac{h}{R_2}\right)dh = \frac{R_2}{8H}.$$

The similar figure for RaC′ α-particles, range $R_3 > H$, is given by

$$\frac{1}{4\pi H}\int_0^{H} \pi\left(1 - \frac{h}{R_3}\right)dh = \frac{1}{4}\left(1 - \frac{H}{2R_3}\right).$$

The corresponding probability for α-particles from RaA and RaC′ deposited on the phosphor is 0·5 in both cases, for RaA deposited on the roof it is zero and for RaC′ on the roof it is $\frac{1}{4}(1 - H/R)$.

Taking into account the relative areas of roof, walls and phosphor, the efficiency for RaA is given by

$$\frac{(R_2/2 + a)}{4(H + a)} \qquad (2)$$

and for RaC′ is

$$\frac{(2a + H - aH/R_3 - H^2/2R_3)}{4(H + a)} \qquad (3)$$

where $2a$ is the diameter of the chamber. Substitution of numerical values in (1), (2) and (3) gives 18·8% for radon, 21·0% for RaA and 25·8% for RaC′, yielding an overall efficiency of 21·5%. This was so low compared with the experimental figure that it seemed the original assumptions were in error. If the daughter products are assumed to be deposited only on the phosphor, then the overall efficiency, assuming 50% efficiency for RaA and RaC, is 39·6%, much closer to the experimental value. The efficiency for RaC was measured separately by filling the chamber with a known amount of radon mixed with air, allowing it to come to equilibrium, evacuating and immediately refilling with aged air. The count rate was then determined over a four hour period and from the decay curve obtained, the count rate due to the RaC′ in equilibrium with the radon was calculated. The efficiency was found to be nearly 50% indicating almost complete deposition on the phosphor. The only explanation for this appeared to be that the phosphor and Perspex were negatively charged, the daughter products being collected immediately after formation.

The existence of this negative charge was verified by inserting an insulating probe into the chamber, the potential it attained being measured by a Lindemann electrometer. From this measurement it was deduced that the field in the chamber was between 3 and 4 v cm^{-1} which should be adequate to ensure deposition on the phosphor.

The question naturally arises as to the origin of the charge on the screen. It is well known that Perspex readily acquires a charge, particularly during cleaning or polishing (Forrest 1953). Alternatively, it may have been acquired when the ZnS(Ag) layer was deposited by spraying. There seems to be no doubt that the charge was held over a considerable period as long as the screen was in position in the chamber. During the two years that the equipment has been in use the calibration has been checked at intervals of about six weeks with no appreciable change. The fact that only dry air was admitted would help in the retention of the charge. Recently, the screen has been removed and exposed to air in the laboratory. After a period of two weeks the charge fell to a very low figure which fluctuated from day to day. When the screen was replaced and the calibration repeated, it was found that the overall efficiency had fallen to 21%, while the efficiency for RaC′ was 25%, both of these values being close to those calculated on the assumption of uniform deposition of daughter products on roof, walls and phosphor. When a freshly deposited screen was used however, the efficiency was restored to its original value.

6. Conclusion

The method described enables samples of about three litres of air to be assayed for radon content without pretreatment, the efficiency being of the order of 38% and in agreement with the theoretical figure. This figure is only attained when the phosphor is negatively charged and this could have been achieved by placing a thin foil over the phosphor and connecting it to an appropriate negative potential. However, the electrostatic charge on the phosphor and Perspex backing has been found to be adequate.

The counter suffers from the limitation of rather a high background, equivalent to a radon concentration of 5×10^{-14} c l^{-1}, so that the accuracy at concentrations lower than this figure is rather poor. The background is attributable almost entirely to contamination in the ZnS(Ag) phosphor. It is possible that a thin plastic phosphor might have a lower contamination figure. On the other hand, the light output is considerably less, so that the cosmic ray pulses due to the Čerenkov effect may add to the background.

Acknowledgments

The authors wish to thank Professor J. Rotblat of St. Bartholomew's Hospital Medical College, in whose laboratories part of this work was carried out and Dr. C. O. S. Blyth Brooke, Medical Officer of Health for Finsbury, for his help and interest. Also Mr. E. R. Kerkin who constructed the apparatus and was responsible for most of the mechanical design.

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