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A Method for Determining Relative Amounts of Combined and Uncombined Radon Daughter Activity in Underground Uranium Mines

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A method for determining relative amounts of "uncombined" radon daughter decay products has been developed. Because of the different diffusion coefficients of atomic-size radon daughter products compared with those combined with particulates, a diffusion tube will separate out a calculable fraction of the atomic-size daughter products and permit larger particulates to pass through the tube. Data obtained from samples collected at several active underground working faces indicate substantial amounts of highly mobile radon daughter alpha activity.

Introduction

THE SHORT-LIVED decay products of radon-222 (RaA through RaC') create such significant health hazards in uranium mining that continued study is warranted to elucidate information on factors that might influence the deposition and retention of these elements in the respiratory system. Several investigators have proposed lung models based on various assumptions and have calculated the radiation doses that would be delivered to the respiratory tract as a consequence of inhaling air contaminated with radon daughters. Reports by Shapiro,[1] Chamberlain and Dyson,[2] Hultqvist,[3] Simpson *et al.*,[4] Altshuler,[5] and Jacobi[6] give details of the various models used.

There is general agreement that radon daughters may exist in an atmosphere in two forms — as individual or "uncombined" atoms, or as atoms "combined" with particulate matter. The relative importance of these two forms as sources of radiation dose to the lung has been assessed differently. For example, Chamberlain and Dyson assumed that only the uncombined RaA atoms delivered an effective radiation dose to the bronchi and trachea. On the other hand, Altshuler and Jacobi calculated that these atoms contributed only a minor portion of the total radiation dose to the critical lung tissue. Several different estimates have been used by workers considering the fraction of uncombined daughters that may exist. Shapiro calculated that in low-nuclei air containing 50 nuclei/cc, 99% of the RaA would be uncombined; in fairly clean air containing 5000 nuclei/cc, 43%; and in air containing 100,000 nuclei/cc, the fraction of uncombined RaA atoms would be negligible. Some measurements have been reported[7,8] of combined-uncombined radon daughter relationships in uranium mines, but these data are too fragmentary to permit of any general conclusions.

Radon decay products will become attached readily to particulates commonly present in air. Wilkening[9] used an electrostatic precipitator to measure the mobility of the carriers of the radioactivity in the general air. By calculation from Stokes' law he found that 90% of the radioactivity was associated with particles of effective diameters of less than 0.5 micron, and that most of the particles in this fraction were in the size range of 0.001 to 0.04 micron. As the

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mobilities of freshly formed radon daughter atoms are similar to those of positive ions where the charge is caused by the stripping of shell electrons, it is reasonable to expect that the daughter atoms will be attached to particulates soon after formation.

Wellisch[10] reported the diffusion constant of RaA to be 0.045 cm^2 sec^{-1}. In contrast, Nolan and Keenan[11] reported diffusion coefficients for condensation nuclei produced by a hot platinum wire ranging from 5×10^{-4} to 8×10^{-7} cm^2 sec^{-1}. As these values for nuclei are less than that for RaA atoms by more than two orders of magnitude, it should be feasible to develop diffusion methods that would separate atomic-size ions from the larger forms. The mathematical basis for such separations was developed by Townsend[12] in 1900, and redeveloped and modified by Gormley and Kennedy[13] in 1948. Nolan and Guerrine[14] and Thomas[15] extended the method to characterize monodisperse aerosols by determining the related diffusion coefficients. Gallimore and Mercer[16] used diffusion theory to develop diffusion coefficients from which particle sizes could be estimated for I^{131}, Te^{132}, and Ru^{103}. Chamberlain and Dyson[2] studied the deposition of uncombined Rn^{222} and Tn^{220} daughters in a model of a human trachea and bronchus. They also investigated aerosols containing Rn^{222} and Tn^{220} daughters, using a diffusion tube and flow rates designed to simulate the conditions existing in a bronchus during respiration.

To determine the relative amounts of combined and uncombined radon daughter atoms existing in mine atmospheres, it was necessary to devise a method that would separate efficiently the two species of atoms and permit the measurement of each fraction. The study reported here was undertaken to develop such a procedure and to measure conditions existing in active uranium mines.

Theory

When an aerosol is passed through a tube under conditions of laminar flow, particulates are deposited on the surface of the tube at rates and distances determined by the length of the tube, the rate of flow, and the diffusion constants of the various species of particles composing the aerosol. Thus, diffusion tubes can be used to separate an aerosol into fractions if the diffusion constants of the particles differ markedly. The equations of Gormley and Kennedy[13] which describe the process are given below:

$$F = 0.8191e^{-7.314h} + 0.0975e^{-44.6h} + 0.0325e^{-114h} + \ldots + \quad \text{when } h > 0.0156$$

$$F = 4.07h^{2/3} + 2.4h + 0.446h^{4/3} + \ldots + \quad \text{when } h < 0.0156$$

where

$h = \pi DL/2Q$.

D = diffusion coefficient, cm^2/sec

L = length of diffusion column, cm

Q = flow rate, cm^3/sec

F = fraction of particles penetrating the column.

The smallest condensation nuclei reported by Nolan and Keenan[11] had diameters of 0.01 micron with an associated diffusion constant of 5×10^{-4} cm^2 sec^{-1}. Condensation nuclei studied by Chamberlain and Dyson[2] had a diameter of 0.15 micron and a diffusion constant of 3×10^{-6} cm^2 sec^{-1}. This latter size is probably more representative of the condensation nuclei found in mine atmospheres. Radon daughter atoms are assumed to have the same diffusion constant as RaA, which is 0.045 cm^2 sec^{-1}. Table I lists the calculated percentages of penetrations of particles with these specified diffusion constants through a 63.5-cm cylindrical tube at various flow rates. This table shows that at the lowest flow rate about 90% of the smaller and 99.5% of the larger condensation

TABLE I

Calculated Percentage of Penetration for Particles with Three Diffusion Coefficients at Various Flow Rates through a 63.5-Cm Tube

Q	Percentage Penetrating the Tube		
		Condensation Nuclei	
(Liters/min.)	RaA $D=0.045$	$D=5\times10^{-4}$	$D=3\times10^{-6}$
0.6	3.06	89.29	99.62
1.0	11.4	91.62	99.73
1.5	22.2	94.00	99.79
2.0	30.6	95.00	99.98

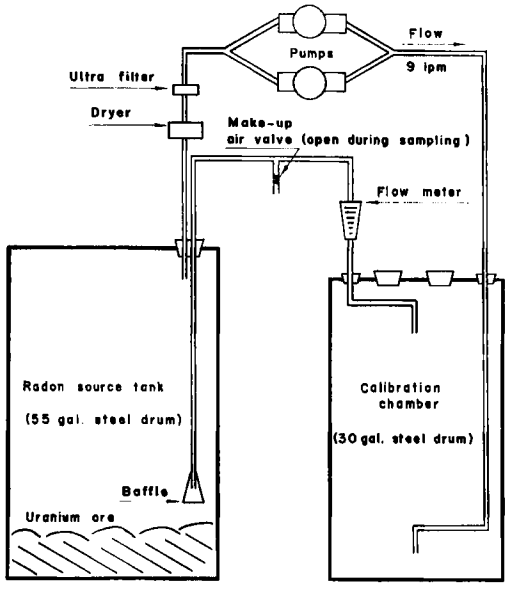

FIGURE 1. Closed-circuit system.

nuclei penetrate the diffusion tube. At higher flow rates the penetration is even greater. In contrast, the penetration of atomic-size nuclei ($D = 0.045$ cm^2 sec^{-1}) ranges from about 3% to about 30% over the indicated flow rates.

Laboratory Studies

Apparatus

Diffusion tubes must be individually calibrated to adjust for factors, such as entry losses, that cannot be calculated from diffusion theory. To perform such calibrations a system is required in which all the variables that might affect diffusion process are controlled. A series of preliminary experiments showed that it was essential that the test atmosphere be practically free of nuclei; that the humidity be maintained at a constant low level; that the radon daughter concentration be held constant; that primarily only one isotopic species be present; and that electrostatic effects be controlled. With these considerations in mind, the closed-circuit system shown in Figure 1 was designed.

In this system the source of radon was uranium ore contained in a steel drum. Air was pumped from this drum through a $CaSO_4$ drying column, through a membrane filter (0.8-micron pore size) to the sampling chamber, and then back to the source chamber. Two diaphragm pumps in parallel maintained a flow rate of 9 liters/min. Since the membrane filter was very efficient in collecting radon daughters[17] (both uncombined and combined), the air discharged into the sampling chamber contained almost daughter-free radon. After a circulation time of one hour (4.6 air changes), the radon daughter concentrations in the tank reached a steady state. Decay curve analyses[18] of typical samples taken from the sampling chamber showed that the RaA:RaB:RaC activity ratios were 1:0.1:0.01. Thus, essentially all the alpha activity in the sampling chamber was produced by freshly formed RaA atoms in an atmosphere free of particulates to which they might attach. This system provided a source of uncombined atoms for calibrating the diffusion tube.

The diffusion tubes were made of aluminum tubing, 60 cm long and 37 mm in diameter. Each tube was fitted with a plastic filter paper holder containing a membrane filter (0.8-micron pore size) at the exit end. Diffusion tube B, used as the reference tube (see calibration section), was also fitted with a similar holder and paper at the entrance. Sampling tube A was equipped with an open plastic collar at the entrance end to make the over-all lengths of each tube 63.5 cm. In use, both tubes were grounded to the sampling chamber to ensure that equipotential conditions existed.

Calibration

The diffusion tubes were calibrated by measuring the fractions of uncombined atoms that penetrated the tubes at various flow rates. These values were compared with the theoretical values calculated by Gormley and Kennedy's diffusion equation. The theoretical values for flow rates of 0.6, 1.0, 1.5, and 2.0 liters are presented in Table I.

Air was drawn, by means of a vacuum system, through each of two diffusion tubes vertically positioned side-by-side in the sample chamber (see Figure 2). The alpha activity collected on filter paper S attached to the exit end of tube A comprised that fraction of the chamber activity that was

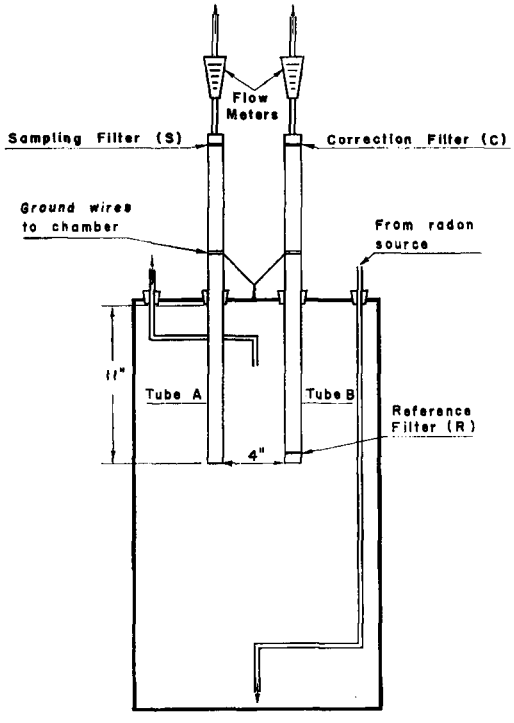

FIGURE 2. Calibration apparatus.

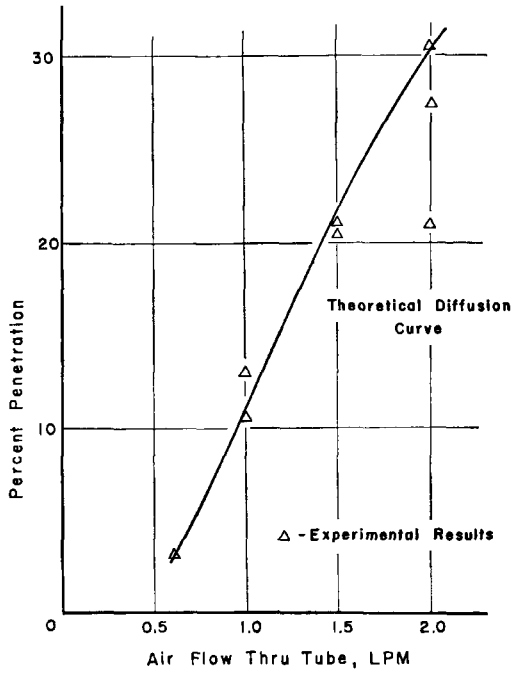

FIGURE 3. Experimental results versus theoretical diffusion values.

able to penetrate the tube plus the radon daughter activity produced by decay of radon in the tube. The activity collected on filter R at the entrance end of tube B represented the radon daughter activity in the air entering the tubes (reference activity). Since this filter removed the radon daughters, only radon gas entered the tube. The purpose of filter C at the exit end of tube B was to provide a correction for the decay products that formed from radon in the tube during the sampling period. It was thereby determined that about 16% of the activity penetrating the sampling tube at a flow rate of 1 liter/min resulted from the radon decay products formed during passage through the tube.

The observed fraction (F) of radon daughter activity penetrating the tube can be calculated as follows:

$$F = \frac{\text{Activity on filter S—activity on filter C}}{\text{activity on filter R}}$$

The results of several tests are presented in Figure 3, along with a plot of theoretical values determined by the Gormley and Kennedy diffusion equation. Since the experimental results at a flow rate of 1 liter/min show good agreement with theory, and since 1 liter/min is a practical flow rate for field application, that rate was selected for field sampling.

Field Studies

The percentage of uncombined radon daughters was measured, by using the diffusion techniques described, in various working areas of several underground uranium mines in southern Utah and Colorado. The size of mine and the types of mining operations were selected to represent a cross section of uranium mines in the Colorado Plateau.

Sampling was accomplished in the field in much the same manner as in the laboratory. Portable battery pumps were used to draw air through the diffusion tubes and membrane filters at a flow rate of 1 liter/min. A series of counts of each filter (R, S, and C) were made by a gas proportional counter. These counts were used to plot decay curves on semilog paper which permitted compari-

TABLE II
Summary of Field Studies Results

Mine	Sample Number	Conditions RH	Conditions T (°F)	Working Level[a]	Percentage of Uncombined Activity	Remarks
A	1	80%	—	0.3	54.6	Active stope. Moderate visible dust. Air velocity 30 fpm.
A	2	87%	—	5.6	48.6	One mucker operating. Little visible dust.
A	3	56%	—	2.7	3.7	Loading ore. Little air movement. Diesel smoke.
A	4	76%	—	1.8	33.4	Slusher slot, after blasting. Heavy powder smoke.
B	5	57%	72°	6.4	60.8	Haulageway. Moderate visible dust. Air velocity 30 fpm.
B	6	66%	65°	4.6	13.2	One drill operating. Moderate visible dust. Air velocity 20 fpm.
C	7	—	72°	0.5	0	Drilling stopped shortly before. Moderate visible dust. Diesel smoke.
D	8	81%	72°	7.0	24	Face. Little air movement. Little visible dust.
E	9	70%	66°	4.3	33	One man shoveling ore. Little visible dust. Air velocity 15 fpm.
E	10	74%	66°	3.1	73	One drill operating. Moderate visible dust.
E	11	90%	65°	9.6	10.2	Moderate visible dust.
E	12	90%	65°	8.4	6.6	Moderate visible dust.
F	13	—	66°	6.6	5.8	Diesel smoke. Oil mist. Little air movement.
F	14	—	67°	0.9	0	Active stope. Moderate dust. Mucking ore prior to sample. Little air movement.
G	15	80%	70°	0.6	12	Dead end drift. Two men drilling. Oil and H_2O mist visible. Little air movement.
G	16	—	—	18.6	20.6	Face. Ventilation system not operating. Moderate visible dust.

[a] 1.3×10^5 Mev of potential alpha energy per liter of air is equivalent to one working level.

son of the activities at similar times. From these data the percentages of uncombined daughter activity were calculated by the following technique:

Let X = uncombined activity
Y = combined activity.
F = the fraction of uncombined activity that will penetrate the tube. For the dimensions of the tube used and at a flow rate of 1 liter/min, $F = 0.114$.
C = daughter activity formed in the tube (count on correction filter C).

Then

$$X + Y = R \text{ (activity on reference paper R)} \quad (1)$$

$$F(X) + Y + C = S \text{ (activity on sampling filter S)} \quad (2)$$

$$[F(X) + Y + C] - C = \text{(corrected activity on filter S)} \quad (3)$$

Equations 1 and 2 solved simultaneously for X give

$$X = \frac{R - S + C}{1 - F}$$

$$\frac{X}{R} \times 100 = \% \text{ uncombined activity}$$

A summary of field study results is presented in Table II. Temperature and relative humidity measurements were made at most sample locations. Radon daughter working levels as measured by the standard method are also listed, along with descriptions of sample location, dust conditions, air velocity, and type of operation.

Discussion

The amount of uncombined radon daughter activity ranges from almost zero up to 73%. This wide range indicates that it would be impossible to select any particular fraction of uncombined atoms as representative of actual mine conditions. It is apparent that several factors affect the percentage of radon daughter atoms that exist in the uncombined state. Some of these are the atmospheric concentration of particulates, the amount of water vapor in the air, and the rate of air change in the area (the longer the residence time, the greater is the opportunity for any particular daughter atom to become combined). An extensive study would be required to evaluate these factors and determine their significance. One general observation was that in areas where diesel smoke was present the fraction of uncombined daughters was low.

The biological meaning of these findings is uncertain. As was pointed out earlier, several different lung models have been proposed which assess the role of uncombined daughters quite differently. The information developed by this study does not help to resolve those differences but only shows that the fraction of the daughters that exist in a highly mobile form varies over wide ranges and is high in many mine areas. The method of collection and counting will not permit calculation of the relative amounts of each of the daughters present in a given mine atmosphere, but the deposition rates of these elements in the respiratory tract should be quite similar and should be affected similarly by the same factors.

If the fraction of radon daughter atoms that exist in either a combined or uncombined state does influence the effective radiation dose delivered to the critical tissue in the lungs, the present method of measuring atmospheric concentrations of radon daughters is only poorly correlated with the true exposure of the miners. For example, the fraction of combined daughter atoms found in the field tests varied from 1.0 to 0.27. If it is assumed that only the combined atoms are the important item, the working level values shown in Table II should be multiplied by factors varying from 1 to 0.27 before these numbers are used to calculate exposure indices. On the other hand, if the uncombined atoms are all-important, the correction factor would vary from 0 to 0.73. Studies of the lung deposition and retention of known mixtures of uncombined and combined radon daughter atoms are needed to properly assess the radiation exposure of uranium miners.

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References

1. Shapiro, J.: *An Evaluation of the Pulmonary Radiation Dosage from Radon and Its Daughter Products*, UR-298, the University of Rochester Atomic Energy Project (1954).
2. Chamberlain, A. C., and E. D. Dyson: The Dose to the Trachea and Bronchi from the Decay Products of Radon and Thoron. *Brit. J. Radiol. 29:* 317 (1956).
3. Hultqvist, B. K.: Studies on Naturally Occurring Ionizing Radiation. *Svenska Uetensk. Akad. Handl. 6:* No. 3 (1956).
4. Simpson, S. D., C. G. Stewart, and E. W. Vogt: The Airborne Radiation Hazard in Ventilated Uranium Mines. Unpublished memorandum (1958).
5. Altshuler, B.: Estimation of Lung Tissue Dose from the Inhalation of Radon and Daughters, Hanford Symposium. *Health Phys. 10:* 1136 (1964).
6. Jacobi, W.: *Die Naturlicke Radioaktivitat der Atmosphase and ihre Bedeutung fur die Strahlenbelastung des Menchen*. Hahn-Meitner-Institut fur Kernforschung, Berlin, HMI B21 (1962).
7. *Experimental Environmental Study of AEC Leased Mines*, Health and Safety Laboratories, NYOO, USAEC, HASL-91 (1960).
8. Bellard, F., J. Miribel, G. Madeleine, and J. Pradel: Methodes de mesure du Radon et de Dosage dans les Mines d'Uranium. *Radiological Health and Safety* in *Mining and Milling of Nuclear Materials*, Vol I, pp. 411-423, International Atomic Energy Authority, Vienna, Austria (1964).
9. Wilkening, M. H.: Natural Radioactivity as a Tracer in the Sorting of Aerosols According to Mobility. *Rev. Sci. Instr. 23:* 13 (1952).
10. Wellisch, E. M.: Experiments on the Active Deposit of Radium. *Phil. Mag.* [6] *28:* 417 (1913).
11. Nolan, P. J., and E. L. Keenan: Condensation Nuclei from Hot Platinum: Size, Coagulation Coefficient and Charge Distribution. *Proc. Roy. Irish Acad. 52:* 171 (1949).
12. Townsend, J. S.: The Diffusion of Ions into Gases. *Phil. Trans. Roy. Soc. London A193:* 129 (1900).
13. Gormley, P. G., and M. Kennedy: Diffusion from a Stream Flowing through a Cylindrical Tube. *Proc. Roy. Irish Acad. 52:* 163 (1949).
14. Nolan, J. J., and V. H. Guerrine: The Diffusion Constants and Velocities of Fall in Air of Atmosphere Condensation Nuclei. *Proc. Roy. Irish Acad. 43:* 4, 1935.
15. Thomas, J. W.: *The Diffusion Battery Method for Aerosol Particle Size Determination*. ORNL 1648 (1954).
16. Gallimore, J. C., and T. T. Mercer: *The Particulate State of Fission Products Released from Irradiated Uranium When Heated in Air. I. Preliminary Results for I^{131}, Te^{132}, and Ru^{103}*. Lovelace Foundation for Medical Education and Research, USAEC, Lf-9 (1963).
17. Anderson, D. E.: Efficiencies of Filter Papers for Collecting Radon Daughters, *Amer. Ind. Hyg. Assoc. J. 21:* 428 (1960).
18. Tsivoglou, E. C., H. E. Ayer, and D. A. Holaday: Occurrence of Nonequilibrium Atmospheric Mixtures of Radon and Its Daughters, *Nucleonics 11:* No. 9, 40 (1953).