any configuration of source container. They provide sealing capability unaffected by temperatures as low as 5°C, can be applied quickly and safely at remote distances from the sources and can be easily removed for source retrieval. Gel sealing is not recommended as an alternative to the disposal of leaking radium sources. Such sources however, can be safely handled prior to and during disposal in containers sealed by gel.

PAUL H. BEDROSIAN

EPA Region I
John F. Kennedy Federal Building
Boston, Massachusetts 02203

EDWIN L. SENSINTAFFAR

National Institute for Occupational Safety & Health
1014 *Broadway Street*
Cincinnati, Ohio 45202

GERALD L. GELS

Radiochemistry & Nuclear Engineering
Research Division
National Environmental Research Center
5555 *Ridge Avenue*
Cincinnati, Ohio 45213

DAVID L. NORWOOD
AVIS A. CULVER

Eastern Environmental Radiation Facility
P.O. Box 3009
Montgomery, Alabama 36109

Reference

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Preliminary Studies of the Effects of Sealants on Radon Emanation from Concrete*†

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EXPERIMENTAL studies of the emanation of radon from concrete were conducted with a type of concrete chosen because of its similarity to that used in forming concrete from shales, cinders and other low density aggregates (AUXIER, 1972). In order to provide sufficiently high radon yield without unmanageably large surfaces, some preliminary studies were made with commercially available building blocks and with samples of uranium ore. The size and configuration of the concrete elements constructed for use were 3-ft square slabs of 6-in. thickness. The aggregate was a commercially available shale preparation (Shalite) to which uranium ore was added. The uranium ore was carnotite from the Grand Junction area of Western Colorado and assayed approximately 0.2% uranium by weight. The densities of the finished blocks were 90–95 lb/ft^3. The total weight of each block was approximately 400 lb of which either 50 or 100 lb were due to the uranium ore. Both the density and appearance of the blocks closely approximated that of shale or cinder blocks.

The experiments were conducted in a relatively airtight enclosure as shown in Fig. 1. The walls of the

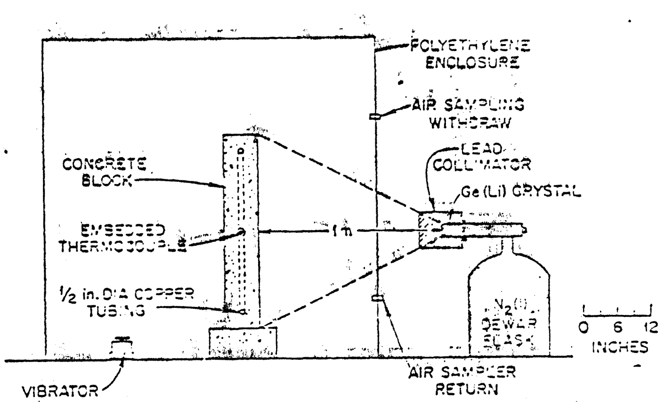

FIG. 1. Diagram of airtight enclosure used for radon emanation studies with concrete block and Ge(Li) detector in place.

enclosure were polyethylene (0.004 in.) supported on a wooden frame. A 55 cm^3 Ge(Li) detector was mounted so as to measure the gamma spectrum from outside the plastic wall. The enclosure was chosen to provide a stable temperature, humidity, and dust distribution. These quantities as well as the barometric pressure were measured as the temperature of the blocks was varied in the course of the experiment.

The blocks were cast with 0.5-in. o.d. copper tubing woven through the midplane parallel to the large faces. A water heating and pumping system was provided for heating the blocks uniformly by means of the copper tubing. Temperatures were recorded by six thermocouples located in, on and outside the block. Air sampling for the enclosure was accomplished by a closed loop sampling system; the air was withdrawn through the four-stage cascade impactor, a millipore filter, a flow-rate meter, and returned to the enclosure at a flow rate of 17 l/min.

The experiment was conducted in the following sequence: (1) set up a block in enclosure; (2)

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measure gamma ray spectrum; (3) measure radon concentrations and particulate size distributions and concentrations, at room temperature (maintained at 23°C) and a fixed humidity, as a function of time; (4) vary temperature or humidity and remeasure Step 3, repeating until temperature and humidity range are covered insofar as is practical; and (5) remove block, coat with a sealant chosen to be of practical interest and repeat Steps 2 and 3, including measurements of the gamma spectrum as a function of time. From the spectral data, it was possible to assess the buildup of the specific radon daughters in the concrete behind the sealant; however, because the gamma buildup was small compared to the total, no further discussion of this part of the study is given here.

The radon activity was determined by using a high resolution silicon diode detector system described by AUXIER (1972). Dust particle size distributions on the various impactor stages were measured, principally with the aid of electron micrographs from the scanning electron microscope (S.E.M.). The size range of particles on the fourth stage of the cascade impactor was 1–5 μm mean linear dimension with a monotonic decrease in the particle count with increasing dimension. As expected from previous studies, the greatest activity was on the particles having a mean linear dimension of 0.3 μm. Generally, less than 10% of the activity was on particles having a mean linear dimension equal to or greater than 3 μm. From these data and the work of HARLEY (1971) and GEORGE *et al.* (1970), it appears that an effective particle dimension for lung exposure of 0.1–0.3 μm is representative of the atmosphere, generally. Blocks were sealed by stuccoing with cement plaster, the stucco sealant evaluated on the basis of widespread utilization in home construction, and then heavily painted with epoxy paint. A second sealant type was the commercially available asphalt used to cement asphalt or vinyl floor tiles in place.

Results and Analysis

For the concrete with high moisture content, the relative humidity in the air of the enclosure varied from 84% at a block temperature of 23°C to 86% at a block temperature of 43°C. Because these humidity factors were larger than those normally encountered in buildings, the block was dried by heating. For low moisture in the block, the relative humidities of the air in the enclosure varied from 58% at a block temperatue of 23°C to 72% at a block temperature of 43°C. In the case of a relative humidity of 80% at a block temperature of 23°C, the humidity was artificially increased by placing trays of water in the enclosure. In all other cases, the humidity of the air was allowed to come into a natural equilibrium with the moist concrete blocks. The total moisture in the low moisture concrete was 2–4% by weight and in the high moisture concrete was 6–8% by weight. The uncertainties in these estimates are large because of the limited space in the available vacuum oven.

Results of these studies indicate a negligible temperature effect on the emanation of radon from concrete, between 23° and 43°C, and a relatively strong dependence of radon emanation on the moisture content of the concrete. However, this is not contrary to the temperature effect reported by GABRYSH and DAVIS (1955) from studies using crushed samples of uranium bearing rocks; their temperature ranges were much greater.

In the measurements of radon emanation from low moisture concrete, the concentration of radon daughters in air has an apparent correlation with the relative humidity of the air in the enclosure as shown in Fig. 2. This apparent correlation is shown in the figure by the straight lines whose equations were obtained by a least squares fit to the experimental data. If the concentration of radon daughters in the air of the enclosure for the low moisture state of concrete is normalized to the same relative humidity using the equations given in Fig. 2, the lack of dependence of radon emanation on the temperature of the concrete is obvious. The dependence of radon emanation on the moisture content of the concrete is emphasized by normalizing the concentration of radon daughters in air from a low moisture block to a relative humidity of 80%.

In experimental measurements of the ratio of radon concentration to its daughter concentration in the air of the enclosure with blocks of 100 and 50 lb

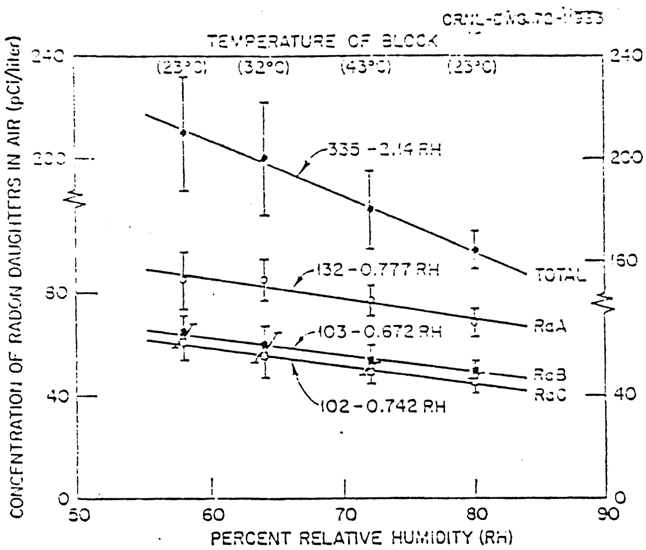

FIG. 2. Concentration of radon daughters in emanation enclosure as a function of humidity.

of carnotite, respectively, a value of 1.60 ± 0.11 was obtained, the block containing 100 lb of carnotite yielding the larger concentrations of radon daughters. A value of 1.61 ± 0.05 was obtained also for the ratio of the radium concentration in the blocks of different uranium content using gamma-ray-emission spectra. This average value was obtained by weighing according to the uncertainties in the ratios of the counts in peaks of the emission spectra from the 186, 839 and 2448 keV gamma rays of ^{226}Ra.

In order to estimate the fraction of the radon escaping from the concrete, the value of 80 pCi/l. in the air of the enclosure from the block containing 100 lb of carnotite was used. This was the value measured for low moisture concrete at a block temperature of 23°C and a relative humidity of 58% in the air of the enclosure as shown in Fig. 2. For these conditions, the fraction escaping from the concrete was calculated to be approximately 1%.

In studies on the effects of sealants, no reduction in the concentration of radon daughters in the air of the enclosure and no increase in gamma-ray emission from radon daughters in the concrete were observed from blocks that were either stuccoed with cement plaster or coated with asphalt. For a similar stuccoed block coated heavily with epoxy paint, both a reduction in the radon daughter concentration in air and an increase in gamma-ray emission for radon daughters in the concrete were observed.

The concentration of radon daughters in the air of the enclosure before and after the stuccoed block was painted is given in Table 1. The data show that radon escaping from the painted block is less by a factor of four.

Table 1. The effects of epoxy paint on the escape of radon from a stuccoed concrete block

Radon daughter nuclide	Concentration in air in pCi/l. Before painting	After painting	Ratio
RaA	47.5 ± 1.9	12.1 ± 2.0	3.93 ± 0.66
RaB	35.5 ± 2.8	9.1 ± 1.7	3.90 ± 0.79
RaC	29.9 ± 0.7	7.3 ± 0.8	4.10 ± 0.46
Total	113.0 ± 4.6	28.6 ± 3.3	3.95 ± 0.48

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J. A. AUXIER
W. H. SHINPAUGH
G. D. KERR
D. J. CHRISTIAN

Health Physics Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830

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Comparative Distribution and Excretion of ^{237}Pu and ^{239}Pu Nitrates in Beagle Dogs*

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THE toxicity of ^{238}Pu and ^{239}Pu has largely prevented the use of these isotopes in human studies. Even in cases of accidental contamination of human beings, the data obtained are usually quite unsatisfactory because the amount deposited is small and the radiation characteristics of these plutonium isotopes are such that *in vivo* measurements are difficult or impossible. Therefore, our knowledge of the metabolic behavior of plutonium has come largely from animal experiments, and its extrapolation to human being is fraught with considerable uncertainty.

Studies by TODD and LOGAN (1966) and LOGAN and TODD (1968) suggest that ^{237}Pu may be a suitable isotope for studies of the metabolic behavior of plutonium in human beings. More recently, WAITE (1972) studied inhaled ^{237}Pu labeled ferric oxide particles in two human volunteers. Plutonium-237

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