EMISSION OF RADON FROM DECORATIVE STONE

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Abstract

Over 35 samples of decorative stone, imported into the United States for use as countertops and mantels, were measured for the emission of radon using electrets and continuous radon monitors. The 14 engineered stones emitted little or no measurable radon (≤2 pCi/lb; <0.2 Bq/kg), while the natural stones emitted up to 42 pCi/lb (3.4 Bq/kg). Pieces of a granite countertop, removed from a local home, emitted an average 170 pCi/lb (14 Bq/kg) of radon. It is estimated that in most cases the contribution of the decorative stone to the indoor radon concentration will be <1 pCi/L, but may exceed 4 pCi/L in rare cases.

Introduction

Radon (^{222}Rn) is a gaseous decay product of radium, a naturally occurring radionuclide found in all rocks and soils. Inhalation of radon, a Class A carcinogen, and its radioactive decay products have been linked by epidemiological studies to an increased risk of lung cancer. Most indoor radon typically enters buildings at the soil-foundation interface, and a small amount enters the home by emanation from groundwater use, building materials, and outdoor air. The contribution from these sources to indoor radon levels varies widely, although building materials seldom becoming a significant source.

Recently, the use of decorative stone (e.g., granite and marble) as a material for the interior of homes has increased dramatically. Stone-based materials contain widely varying levels of the radionuclides, though igneous rocks, such as granite, are commonly associated with a relatively higher content of radioactivity than sedimentary rock. Studies of radon exhalation from building materials, such as bricks and concrete, have been published, but little information currently exists on the contribution from natural-stone countertops to indoor radon levels. A preliminary study (Kitto and Green, 2005) showed that radon and gamma-rays are emitted from natural stone used in homes, with similar results being reported worldwide (e.g., Anjos et al., 2005; Al-Jarallah et al., 2005; Righi and Bruzzi, 2006). Due to the health implications associated with radon and the lack of radiological data regarding the decorative stone that is imported into the US, the goal of the present study was to measure radon emanation from decorative-stone material using radioanalytical techniques.

Methodology

The engineered and natural-stone slab samples analyzed in the study are representative of decorative stone commonly manufactured and imported, respectively, into the US for use as countertops, mantels, and hearths. Most of the samples included in the study had a single polished surface (1 ft^2; 930 cm^2), similar thickness (~0.75 inches; 2 cm), and densities of 2.5-3.0 g/cm^3. A granite countertop, removed from a local home, was trisected and included in the study. Radon emanation from the samples was determine in a laboratory environment containing ~0.3 pCi/L of airborne radon and a radiation background of 7 µR/hr. The indirect determination of radon emanation was accomplished by the encapsulation of each sample and measurement of the radon in the enclosed air. Direct determination of radon flux from a sample can be accomplished through absorption onto activated charcoal, etched alpha-track detectors, or discharged electrets.

Direct measurement of radon exhalation from the samples was accomplished using a 960-mL hemispherical device (H-chamber; Rad Elec Inc., Frederick, MD) containing a charged Teflon disc (electret). Radon entered the H-chamber through an attached Tyvek sheet in contact with the surface of the samples. The edge of the chamber was sealed to each sample using putty to eliminate leakage for measurement periods of 24-74 h. The flux of radon from the samples was calculated based on a formula and calibration supplied by the manufacturer (Stieff and Kotrappa, 1996). The voltage reader was calibrated using electrets exposed at an accredited radon chamber. Seven duplicate measurements were conducted using this method.

The indirect measurement of radon exhalation was accomplished by sealing the samples inside radon-impermeable bags (Associated Bag Co., Milwaukee, WI) to allow the progeny of the emanating radon to ingrow to a steady state in the enclosed air. An aliquot (125 mL) of radon was transferred from the bag into each of two evacuated "Lucas" cells using a 0.75-inch (2-cm) piece of tubing to connect each evacuated cell to a vent nipple installed in the bag. After >3 h, the cells were measured at least five times with an alpha-scintillation counter (Randam Electronic Inc., Cincinnati, OH) and the radon concentrations determined from an average of the decay-corrected measurements. Background count rates averaged 1.8 cpm and the efficiency of the alpha-scintillation counter was ~2.3 cpm/dpm for ^{222}Rn and its short-lived alpha-emitting progeny. The scintillation counter was calibrated using ^{226}Ra standards traceable to the National Institute of Standards and Technology (NIST).

Results and Discussion

Radon that entered the H-chamber device through the Tyvek membrane discharged the electrets from 0.2 to 9.5 V/hr, including discharge attributable to background (0.2 V/h) radiation. Over half of the samples had net discharges below 2 V/hr, and only six samples had net discharge rates above 4 V/hr. To determine flux, we used a calibration factor, supplied by the manufacturer, that was not confirmed in this study. Calculated radon fluxes showed that two-thirds of the samples emitted <50 pCi/ft^2-hr (<20 Bq/m^2-hr) of radon, but the fluxes ranged up to 770 pCi/ft^2-hr (310 Bq/m^2-hr). Results of duplicate measurements of seven samples agreed well (r^2=0.99). For this study, the polished side of the samples were measured. As the unpolished side may emanate radon at an increased rate due to the lack of polish resin, the fluxes could be greater than measured.

The concentration of radon gas from the samples sealed inside the bags was determined using Lucas cells and was found to range from 0.3 to 480 pCi/L (~10 to 17,800 Bq/m^3). Since the volume of air remaining in each bag (typically 2-4 L) was determined after the filling of both scintillation cells, the total activity of radon in the bags could be determined for the samples. The equilibrium activity of radon was <30 pCi (1 Bq) from over half of the samples, and only three samples produced >270 pCi (10 Bq) of radon in the bag. Measurement of the mass of the samples allowed determination of the mass-activity concentration; this ranged from <0.1 to 170 pCi/lb (0.01 to 14 Bq/kg). Results of the duplicate scintillation cells were highly correlated (r^2=0.99).

Based on the H-chamber measurements, the concentration of radon that would accumulate in a room containing the samples can be estimated using general assumptions. For a 20 m^3 (10 ft • 10 ft • 7 ft) room containing 3 m^2 (32 ft^2) of a granite that emanates 770 pCi/ft^2-hr (310 Bq/m^2-hr), the ingrowth of radon would resemble Fig. 1. Included in the estimate is a 0.5 air change/hr and instant mixing in the room. The granite would continually increase the indoor radon level in the room to 1.2 pCi/L (47 Bq/m^3). Based on the Lucas-cell measurements, a 500-lb countertop (32 ft^2 at 2.6 g/cm^3) emanating 170 pCi/lb (14 Bq/kg) of radon, may produce over 4 pCi/L (148 Bq/m^3) in the 20 m^3 room. As noted above, the unpolished may emanate a greater amount of radon than the polished side, which would explain the lower radon concentrations associated with the H-chamber in Fig. 1. The alpha-scintillation method measured radon emitted from all sides of the samples. If the radon-laden air is dispersed throughout the entire house, the radon level will decrease, and if the air exchange is less, the radon level will increase.

Conclusions

Emission of radon from 36 types of countertop material produced for home interiors was determined using direct and indirect radio-analytical methods. The methods demonstrated that radon emanates from the surface of most natural stone, but rarely from engineered stone. The radon emanations measured in this study spanned three orders of magnitude, with fluxes > 300 pCi/ft^2-hr (120 Bq/m^2-hr). As the samples represent a small fraction of the selection available, decorative stones of greater emanating potential inevitably exist. The contribution from 3 m^2 of the granite that emitted the most radon in this study to indoor radon concentrations was estimated to be at or below the EPA recommended-action level.

Acknowledgments

The flux chambers and electrets were generously contributed by Dr. Paul Kotrappa of Rad Elec Inc. This study was sponsored in part by C&C North America, Inc.

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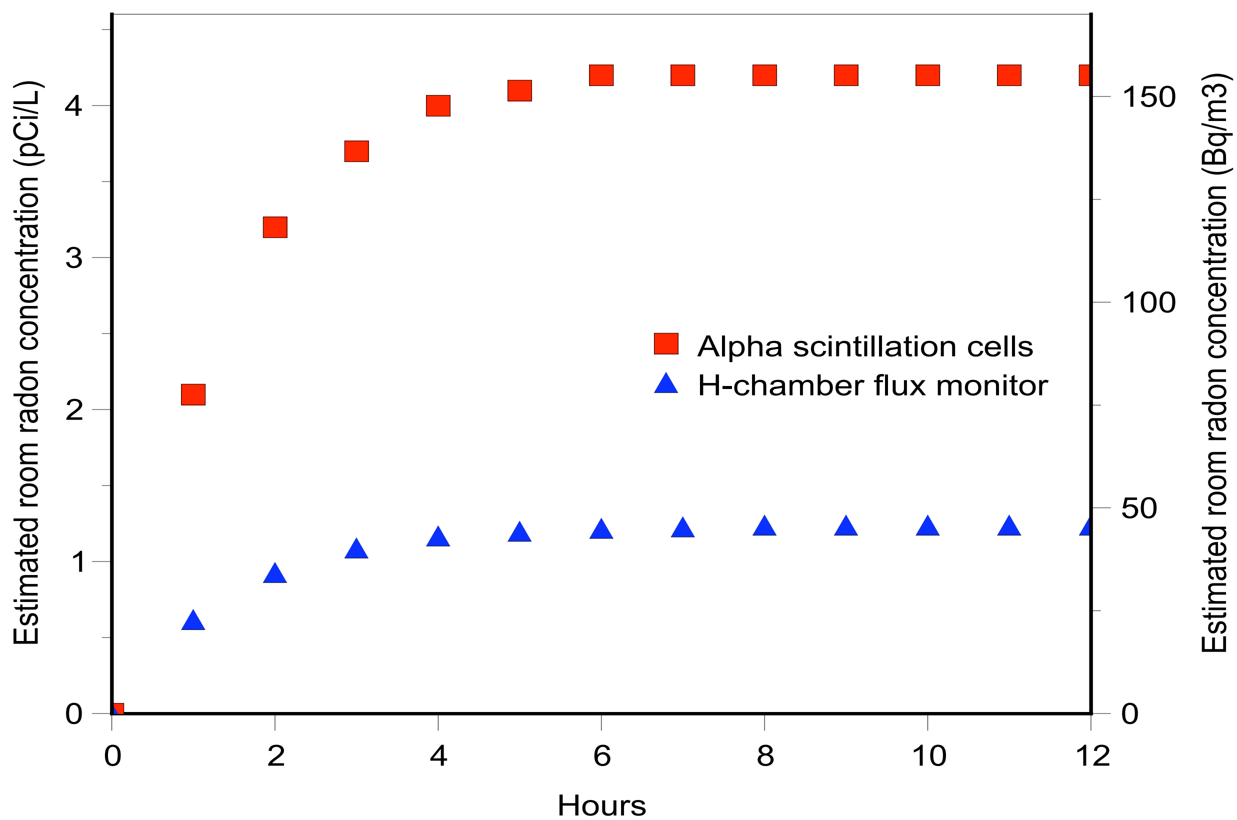

Figure 1. Estimate of radon ingrowth in a 20 m^3 room from granite countertop material based on measurements conducted using a H-chamber flux monitor and alpha-scintillation cells.