

RADON EMANATION FROM HOUSEWARES AND CONSTRUCTION MATERIALS

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ABSTRACT

Emanation of radon and thoron gas from souvenir housewares and construction materials was determined using alpha-scintillation cells. Though gamma-ray spectroscopy noted significant uranium concentrations in the housewares, no radioactive gas emanation was observed. Radioactive disequilibrium of the super ^{238}U and ^{235}U decay series exists in the housewares. The ^{232}Th decay series is in radioactive equilibrium in the housewares at concentrations several orders of magnitude lower than ^{238}U levels. The decay series are in radioactive equilibrium in the construction materials, but only the cement block emanated a significant amount of radon.

INTRODUCTION

Many housewares (*e.g.*, dishes, ashtrays, vases) manufactured using colorful uranium salts and marketed prior to 1972 are held as private souvenirs. Though producing considerable surface exposure, the housewares are not a significant health hazard as the uranium is bound in the glaze (Landa and Councell 1992). Using only gamma-ray spectroscopy measurements, it was claimed (Zeman and Hon 1994) that the dishes emanated radon (^{222}Rn), a gaseous radioactive decay-product of uranium, to produce indoor concentrations in excess of the 150 Bq m^{-3} recommended by the U.S. Environmental Protection Agency, but emanation measurements were not reported. References were made to the study (Valenti 1994; Johnson 1994). However, another study (Kitto et al. 1996) detected no radon emanation from a single plate. Considering the emanation discrepancy an investigation of the magnitude of radon and thoron emanation from additional housewares was essential for an assessment of public exposure.

EXPERIMENTAL

Seven uranium-glazed tablewares from various manufacturers, an ashtray enriched in uranium, and three common home-construction materials (Table 1) were enclosed in individual glass desiccators at atmospheric pressure to allow radioactive ingrowth of the radon and thoron gases. A small-volume vacuum system connected each desiccator to a 125-mL alpha-scintillation (Lucas) cell. Following evacuation of the Lucas cell and connected tubing, the desiccator valve was opened and gases transferred to the Lucas cell.

For thoron determinations the cells were immediately (<30 s delay) measured using 30-s counting times on a Randam[#] SC-5 alpha-scintillation counter with an absolute efficiency of 77% for alpha particles. Three hours after the gas transfer, counting times were increased to one hour for radon measurements. Cells were repeatedly measured to monitor radioactive decay.

High-resolution gamma-ray spectroscopy of the housewares and construction materials was conducted using a vertical, intrinsic-Ge detector ($\sim 95 \text{ cm}^3$) with an efficiency of 22% and resolution of 2.0 keV at 1332 keV. The unstandardized geometries were counted for 1000 min each directly on the detector face to examine radioactive equilibrium of the ^{238}U , ^{235}U , and ^{232}Th decay series. The construction materials were sealed in polyethylene prior to counting. Spectra were collected and integrated using a Genie Spectroscopy System^{*} and PEAK, respectively. Photopeaks with >50% counting error were eliminated. Identical samples were measured for emanation and gamma-ray spectroscopy, except that only a portion of the cement block was used for the latter.

[#] Randam Electronics Inc., 591 Northland Blvd., Cincinnati, OH

^{*} Canberra Industries, Meriden, CT

RESULTS AND DISCUSSION

Gamma-ray spectroscopy revealed approximate radioactive equilibrium of ^{232}Th decay products in all the samples (Fig. 1). The decay products would attain secular equilibrium in the few decades since the housewares were produced. The ^{232}Th decay-product concentrations are similar in the tablewares, brick, and block, but about 10 times lower in the ashtray and sheetrock. The ^{232}Th concentrations in the housewares, being over three orders of magnitude less than ^{238}U (Fig. 1), are insufficient to produce thoron above the Lucas-cell detection limit (300 Bq m^{-3}). There was no measurable thoron emanation from the construction materials, partly as a result of the fact that most thoron decays (half-life of 55 s) before emanating from the samples.

The housewares emanated no radon above the Lucas-cell detection limit of 15 Bq m^{-3} . Gamma-ray spectroscopy showed that some ^{238}U decay-product (^{234}Th , $^{234\text{m}}\text{Pa}$ and ^{234}U) concentrations are in equilibrium in the housewares (Fig. 1), but radon decay-product (^{214}Pb and ^{214}Bi) concentrations are roughly three orders of magnitude lower. While the disequilibrium suggests that radon emanates from the housewares, the 186-keV photopeak often attributed to ^{226}Ra , a decay product of ^{238}U and the parent isotope of radon, contains an interfering contribution from ^{235}U . Though the concentrations of ^{226}Ra appear to be elevated in the housewares, other photopeaks (e.g., 144, 163, and 205 keV) confirm that nearly all (>95%) of the 186-keV photopeak is accounted for by ^{235}U in the housewares. The application of gamma-ray spectroscopy without a correction for the ^{235}U interference is probably responsible for the report (Zeman and Hon 1994) of radon emanation from the tableware. Insufficient time has elapsed for ingrowth of ^{226}Ra (half-life= 1600 y) in the housewares. Similarly, while ^{235}U concentrations in the housewares are greater than those of its decay products (Fig. 1), insufficient time has elapsed for ingrowth of ^{231}Pa (half-life~33000 y). Ratios of $^{235}\text{U} / ^{238}\text{U}$ in the housewares reflect natural abundances.

Concentrations of radon decay products in the brick are equivalent to those in the housewares, while their concentrations in the sheetrock and cement block are significantly lower. The desiccators holding the brick and sheetrock contained about 40 Bq m^{-3} of radon while the cement block produced 1600 Bq m^{-3} . The latter's porosity may be responsible for the contrast. Gamma-ray spectroscopy showed the ^{238}U , ^{235}U , and ^{232}Th decay series to be in radioactive equilibrium in the construction materials (Fig. 1). Sealing the construction materials in polyethylene prior to gamma-ray spectroscopy apparently retained sufficient radon for its short-lived decay products to re-establish equilibrium.

CONCLUSIONS

Contrary to a previous report no measurable radon and thoron emanated from the housewares. Though the ^{232}Th decay series has established radioactive equilibrium in the housewares, low concentrations and emanation precluded the detection of thoron. Significant levels of uranium exist in the housewares, but insufficient ingrowth of ^{226}Ra is responsible for the absence of radon and its decay products. Radioactive equilibrium existed in the construction materials. Radon emanation from the cement block was 40 times that from the brick and sheetrock.

Acknowledgements - The authors wish to thank Peter Rosenberg and Craig McNulty for technical assistance.

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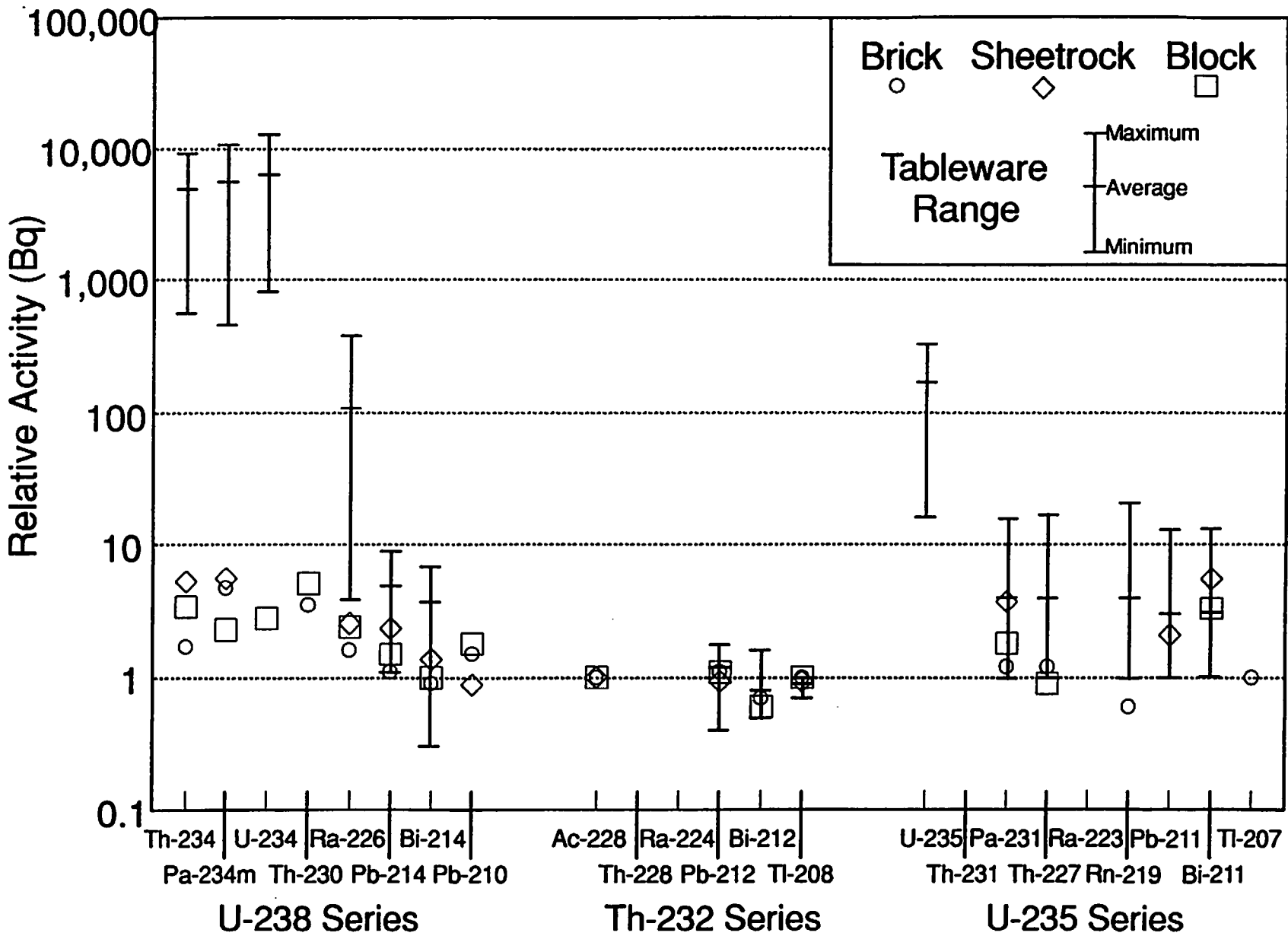


Fig. 1. Concentrations, relative to ²³⁸Ac, of ²³⁸U, ²³⁵U, and ²³²Th decay-products observed in housewares and construction materials by gamma-ray spectroscopy.

Table 1. Description of materials analyzed.

| Tableware Brand ^a | Diam (cm) | Houseware and Construction Material | Volume (cm ³) | | |
|------------------------------|-----------|--|---------------------------|--------------|------|
| Vistosa | 15 | ashtray (broken) ("vaseline" or "canary") | 93 | | |
| Caliente | 21 | | | | |
| Vitreous | 17 | | | | |
| Fiesta | 18 | red brick | 1030 | | |
| Radar City | 19 | | | | |
| GMB | 21 | | | sheetrock | 470 |
| Caliente | 23 | | | cement block | 2440 |
| Fiesta | 21 | | | | |
| No name | 15 | | | | |

^aObtained from insignia on bottom of tableware.