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DIRECT MEASUREMENT OF THE FRACTION OF RADON LOSS IN CERAMICS BY GAMMA-RAY SPECTROSCOPY

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We have re-examined the problem of radon loss in ceramics by direct measurement of gamma-rays originating in the uranium and thorium decay series. Data from a series of unaltered as well as crushed sherds and one clay sample produced results that indicate no radon loss to within the experimental accuracy (<±2%)

Thermoluminescence (TL) dating has as its goal the determination of the time that has passed since a ceramic material has been fired. The procedure, conceptually, is to divide the measured thermoluminescence effect, expressed as an equivalent radiation dose, by the annual dose effective in producing the TL. In order to calculate the annual dose, the amounts of uranium and thorium with their daughter-series, plus the potassium and rubidium present in the ceramic and ambient cosmic rays must all be considered.

In this paper we consider, as has been done by other authors (Tite and Waine, 1962; Aitken and Alldred, 1972; Desai and Aitken, 1974; Aitken, 1976), the effect of possible escape from the ceramic of the inert gas (radon) radionuclides, on the calculation of the annual dose. These inert gas radionuclides, Rn-219, 3.96-second half life and Rn-222, 3.82-day half life, occur about half way through the natural uranium decay series. Thoron (Rn-220, 55.3-second half life) is similarly produced in the natural thorium decay chain.

The short half lives of Rn-220 and Rn-219 would seem to preclude their diffusion to any appreciable degree before transformation to the respective polonium isotopes (Tanner, 1964), leaving these and all subsequent daughter products bound in the system. It has however been assumed in TL research and dating studies that the much longer half life of Rn-222 may allow some of it to diffuse out and not decay in the ceramic. This effect would upset conditions of secular equilibrium in the object, decrease to some extent the post-radon activity, and introduce serious errors into calculations of annual dose rate.

This problem has been discussed by Tite and Waine (1962) and Desai and Aitken (1974), who report measurements of radon loss using alpha scintillation counters: from this Aitken has proposed a method of radon-loss dose correction (Aitken and Alldred, 1972; Aitken, 1976).

The work reported here deals with an alternative method of measuring the amount of radon loss in ceramics and clays and results from several types of sherds are presented. Our philosophy was to measure directly the gamma rays emitted by all the radioisotopes naturally present in sherds, to calculate the gamma emission rates from radioisotopes preceding and following the radon in the natural series, and to compare these rates in cases where the escape of radon was (a) prevented and (b) favored.

DISCLAIMER

To do this, a collection of sherds of one pottery type was placed in a specially designed brass and aluminum counting cell that could be reproducibly positioned to partially enclose a solid state, Ge(Li), detector. The detector was manufactured by Ortec, of 12% efficiency and 1.80 keV resolution and coupled to a Nuclear Data 4096 channel analyzer. The cell could be operated sealed off or could be evacuated with a mechanical pump. Details of the five samples used in the radon loss test are included in Table 1.

The samples were counted for periods ranging from 25 to 60 hours in a sealed cell configuration at atmospheric pressure: the resulting count rates, reduced to counts per hour, indicated the amount of activity in a steady state condition. The spectra were recorded, not merely at the end, but also at various times during the full counting period. The results indicated no change in count rate in a steady state condition, of any of the radioisotopes present, within the experimental limits of uncertainty due to counting statistics.

The cell was then evacuated and the spectra were re-measured at various times after pumping commenced. The sherds were carefully secured in the cell so that, during pumping and counting periods, any differential absorption effects brought about by changing geometry would be held to a minimum.

Peaks from transitions in Th-234, Pa-234, Ra-226, Pb-214, Bi-214, Th-232, Ac-228, Ra-224, Pb-212, Bi-212, and Tl-208 were recorded. Because of differences in branching ratios, some peaks had accumulated many more counts than others, with accompanying smaller statistical uncertainties. Table 2 lists the isotopes and gamma ray energies used in the radon loss analysis. To further improve the counting statistics, the measured activities of two post-radon isotopes, Pb-214 and Bi-214, were added, it being assumed that they could not diffuse or change geometry.

A typical plot of count rate vs time after pumping is shown in Figure 1. Four radioisotopes are used: pre-thoron (Ac-228) and post-thoron (Tl-208) and post-radon (Bi-214 and Pb-214). The thorium chain daughters are used as a monitor in the experiment, since these are not expected to change with pumping. The remaining post-radon points should be greatly influenced by pumping. Note that the longest half life affecting these activities is 26.8 minutes and that most pumping times are orders of magnitude greater than this. The half life of Rn-222, 3.82 days, does not enter into the interpretation of these results, provided the diffusion mean life of free radon atoms in the cell is shorter than this (Harbottle, et al, 1956).

The reduced data from the other samples are listed in Table 3. As in Figure 1, pre- and post-thoron peaks were used as monitors. These results clearly indicate that there was no radon loss within the accuracy limits of the experiment for any of the samples during a pumping period of up to 24 days. This rules out the possibility of appreciable radon diffusion with a diffusion mean life less than an order of magnitude greater than the half life of Rn-222 for these sherds. In addition, two samples of powder were specifically included to see if crushed sherds or unfired clay would influence radon loss. There was no observable radon loss with these samples.

We have described the results of a direct test of radon loss using widely varying samples; results indicate negligible radon loss over a period of approximately 6 half lives of Rn-222. It is suggested that

future radon loss measurements be made using this technique because of its simplicity and directness. If radon losses are found, corrections to the dose rate as proposed by Aitken may be applied.

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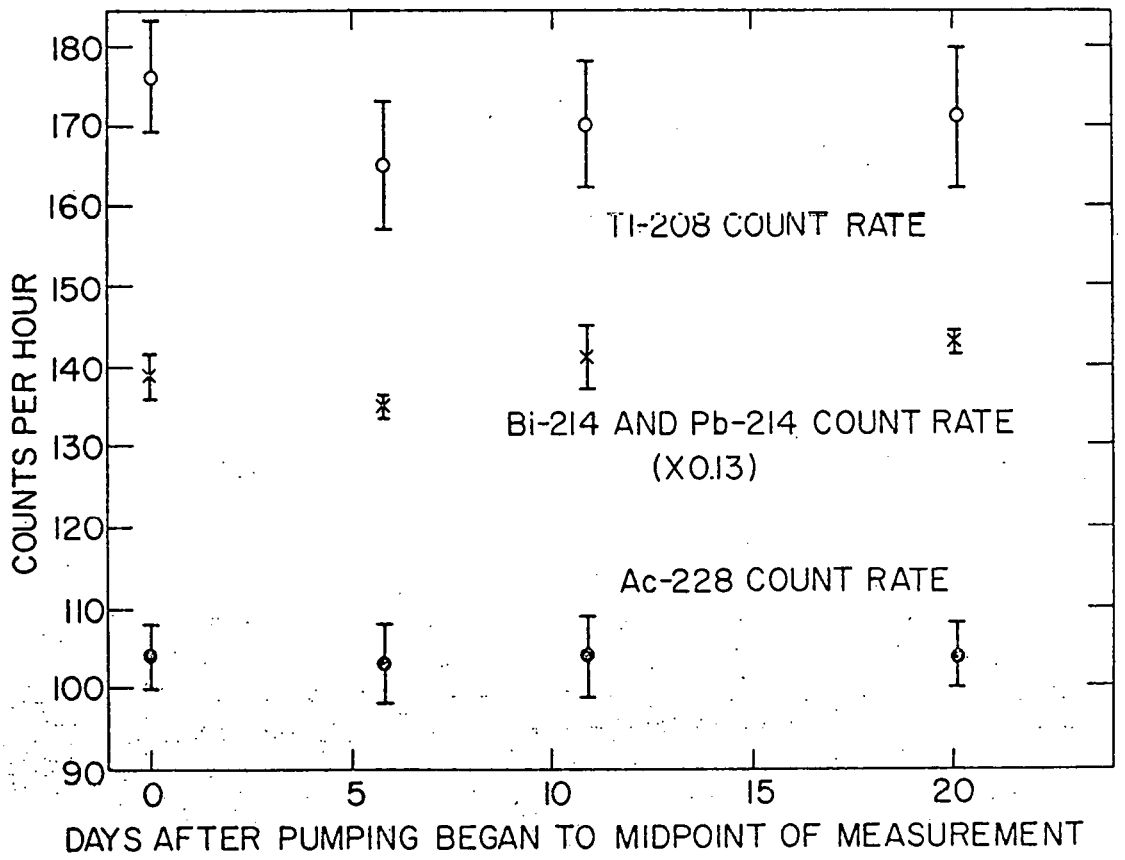

Figure 1. Count rate vs time after pumping began for sherds from Texoloc, Mexico. Error bars represent $\pm 2\sigma$ determined from counting statistics. See Table 2 for gamma-ray energies.

Table 1. Samples Used in the Radon Loss Test

Sample Description	Weight (gm)	UO_3 (ppm)	ThO_2 (ppm)
Amphorae sherds from Marseille, France	975	0.59*	14.6*
Pottery sherds from Texoloc, Mexico	785	1.1*	3.0*
Tuyere sherds from Itezhitezhi, Zambia	750	22	111
Crushed Tuyere sherd from Itezhitezhi, Zambia	91	22	111
Unfired Tennessee** Ball clay powder	73	8.3	23.8

* A representative average from several sherds in the group.
** Obtained from Stewart Clay Co., New Brunswick, NJ 08902.

Table 2. Isotopes and Gamma-Ray Energies Used in Radon Loss Tests

	Isotope	Energy (keV)
Pre-thoron	Ac-228	911.2
Post-thoron	Tl-208	583.14
Post-radon	Pb-214	295.22
	Pb-214	351.99
	Bi-214	609.30
	Bi-214	1120.40

Table 3. Background corrected count rates (counts per hour) ± two standard deviations for radon loss test. The value in parentheses, under the count rate, is the time in days to the mid-point of the measurement, after pumping commenced. The background was taken with the cell empty. The gamma-ray lines are described in Table 2.

Tuyere Sherds				
Post-radon	5546 ± 60 (0.0)	5538 ± 88 (8.2)	5525 ± 66 (19.2)	
Pre-thoron	981 ± 15 (0.0)	973 ± 15 (8.2)	963 ± 46 (19.2)	
Post-thoron	1601 ± 20 (0.0)	1585 ± 50 (8.2)	1543 ± 24 (19.2)	
Tuyere Powder				
Post-radon	1482 ± 34 (0.0)	1479 ± 33 (0.7)	1532 ± 30 (12.8)	
Pre-thoron	248 ± 18 (0.0)	238 ± 9 (0.7)	230 ± 16 (12.8)	
Post-thoron	380 ± 20 (0.0)	384 ± 11 (0.7)	365 ± 7 (12.8)	
Amphorae				
Post-radon	1717 ± 45 (0.0)	1690 ± 41 (1.8)	1755 ± 20 (24.8)	
Pre-thoron	209 ± 8 (0.0)	203 ± 7 (1.8)	208 ± 7 (24.8)	
Post-thoron	357 ± 11 (0.0)	346 ± 8 (1.8)	348 ± 20 (24.8)	
Mexican Sherds				
Post-radon	1067 ± 24 (0.0)	1038 ± 12 (5.8)	1086 ± 30 (10.9)	1100 ± 12 (20.1)
Pre-thoron	104 ± 4 (0.0)	103 ± 5 (5.8)	104 ± 5 (10.9)	104 ± 4 (20.1)
Post-thoron	176 ± 7 (0.0)	165 ± 8 (5.8)	170 ± 8 (10.9)	171 ± 9 (20.1)
Tennessee Ball Clay				
Post-radon	424 ± 16 (0.0)	463 ± 41 (1.9)	437 ± 15 (10.9)	
Pre-thoron	43 ± 7 (0.0)	48 ± 14 (1.9)	45 ± 2 (10.9)	
Post-thoron	67 ± 4 (0.0)	70 ± 7 (1.9)	69 ± 8 (10.9)	