A NEW APPROACH TO DETERMINING ^{222}Rn IN AIR USING LIQUID SCINTILLATION COUNTING

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ABSTRACT. I have developed a new approach to determining ^{222}Rn in air using liquid scintillation counting (LSC). With this method, special charcoal canisters absorb the ^{222}Rn. When the canister is connected to a standard Teflon® vial, radon is transferred to the scintillation cocktail. Exposure time is 3–7 days, with desorption and measurement taking up to 20 h. A complete set consists of 3–5 charcoal canisters per vial. An ordinary vial cap is used to close the vial after desorption for storage and counting with an LS spectrometer. A discrete charcoal canister allows up to 20 ml of cocktail per sample, which decreases the radon elution time. High-energy resolution and low background with Teflon® vials allow improved sensitivity of the method compared to commercially available disposable vials. Only Teflon® vials are used in the laboratory, which prevents evaporative losses. A low detection limit down to 1.0 Bq m^{-3} for a 6-day exposure and 10-min counting time are easily obtained (background: 0.1 cpm; efficiency: 1.0 cpm per Bq m^{-3}).

INTRODUCTION

I have developed a new approach to determining ^{222}Rn in air by liquid scintillation counting (LSC) using special charcoal canisters. After exposure, the charcoal canister is connected to a standard Teflon® vial filled with scintillation cocktail. After desorption, the vial is sealed with a standard cap and the radioactivity measured with a Wallac Quantulus 1220™ LS spectrometer. Exposure time is 3–7 days and desorption takes up to 8–15 h (depending on the scintillator used). Transportation to the site and back to the lab will take up to 4 days. The counting time for a batch of 10–20 samples does not exceed 3–5 h, which means that 3–5 charcoal canisters can be used for one vial. A discrete charcoal canister allows up to 20 ml of cocktail per sample, which increases elution speed. High-energy resolution improves the sensitivity of the method compared to commercially available (Packard) disposable plastic vials. Re-use of Teflon® vials reduces the cost of each measurement. This new approach utilizes the following advantages of the standard Teflon® vials: low background, high output and high-energy resolution.

METHODS

The widely used charcoal-based LSC method for determining ^{222}Rn in air includes several steps: 1) transportation to the site; 2) exposure; 3) transportation back to the lab; 4) desorption of Rn into cocktail; and 5) LSC. In our laboratory, we have designed our own standard Wallac-type aluminium Teflon® 7-, 15- and 20-ml vials. Sorption speed and efficiency depend on scintillator volume, so large-volume vials are preferred. We used an aluminium canister with a charcoal volume of 3.2 cm^2.

Figure 1 shows a schematic diagram of the charcoal canister, which allows air (*i.e.*, Rn) to access the charcoal through a mesh on the top and bottom of the charcoal holder. The holder is filled with freshly prepared charcoal, placed into a canister, and is then fixed using a specially designed spring inside the canister. The canister is closed with a screw cap; it may be packed into a hermetically sealed package. The canister is then transported to the sampling site, opened and exposed for 3–7 days, after which it is recapped and returned to the laboratory.

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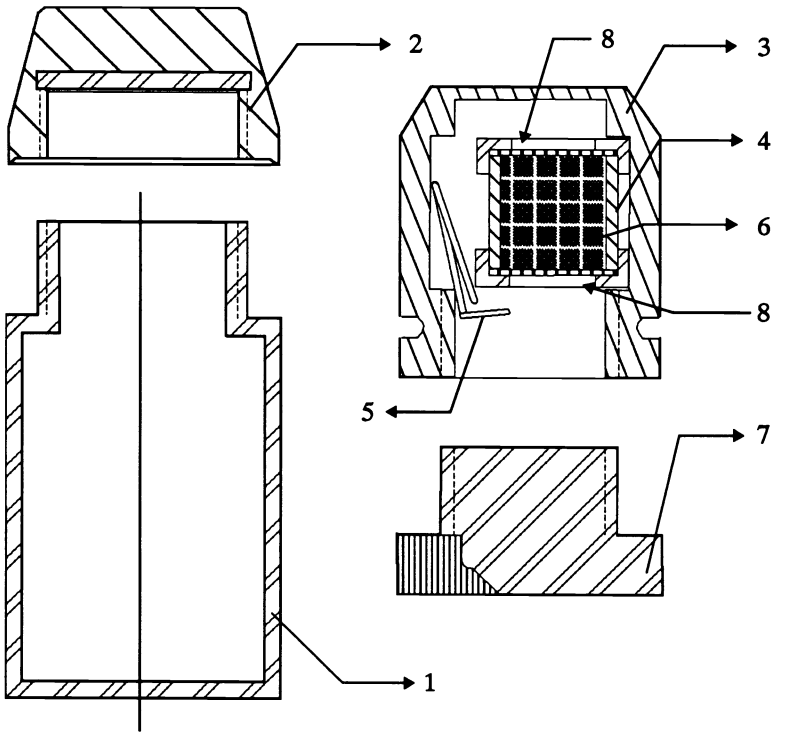

Fig. 1. Diagram of a Teflon® 20-ml vial and charcoal canister sections: 1. 20-ml Teflon® vial; 2. vial cap; 3. charcoal canister; 4. charcoal holder; 5. metal spring; 6. charcoal; 7. screw cap; 8. mesh sieve.

Rn desorption is carried out using standard Teflon® vials filled with LS cocktail. Desorption time depends on the LS cocktail solvent used; it takes up to 8 h with toluene-based cocktails and 15 h with dioxane-based ones. Toluene-based cocktails are more efficient. Rn is completely removed from the charcoal during desorption. Sealed with its standard screw-cap, the vial is ready for measurement. After charcoal replacement, the canister is then ready for another cycle. After measurement and LS cocktail replacement, the vial is also ready for another cycle with another exposed canister. This time-sharing principle allows for recycling vials simultaneously with the charcoal canister. The charcoal itself may be used only once because of residual LS cocktail.

For the charcoal preparation, we heat canisters filled with charcoal at a temperature of 120°C for 60 min. To determine Rn concentration, we used a comparative calibration method. The calibration factor was determined after exposure of the charcoal canister in the Rn chamber with known Rn-specific activity. Thus, we exposed a set of calibration canisters as well as the canisters at sites for Rn-specific activity in air determination. Rn decay due to any delay after exposure and after desorption should be corrected as follows:

$$A_0 = Ae^{\lambda Td} \tag{1}$$

where

A_0 = the initial activity
At = measured activity
Td = total delay time
λ = decay constant = $0.693/T_{½}$

To estimate how tightly the vial is attached to the charcoal canister, we continuously cycled 7-ml vials with the attached charcoal canister in the LS counter. Only vials of this size with attached charcoal canisters may be installed directly into an LS spectrometer. We estimated Rn leakage for 5-day storage to be <10%. At the same time, Rn leakage is important only during the 15-h desorption time. Our estimate for our three 20-ml aluminum Teflon® vials filled with LS cocktail and bubbled ^{85}Kr yield values of 60.6, 77.4 and 93.6 days for the time that the count rate decreases to 50% (storage for 3 months, see Fig. 2).

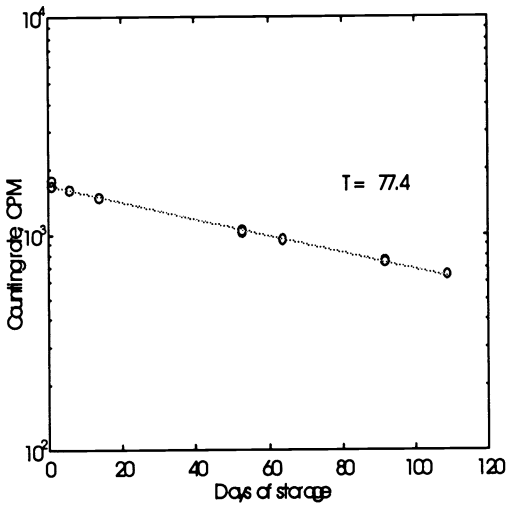

Fig. 2. ^{85}Kr count rate decrease for Teflon® 20-ml vial

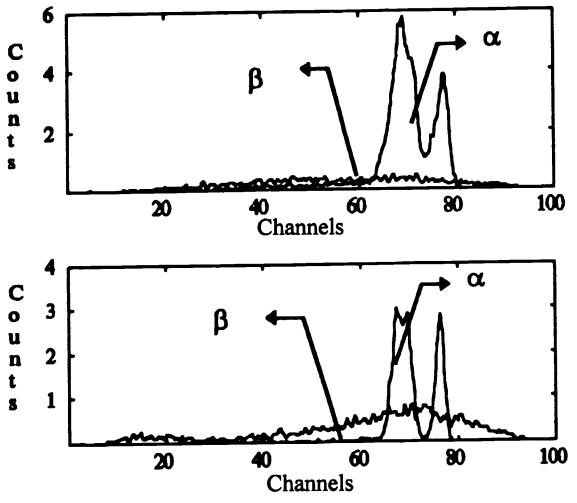

Fig. 3. α and β spectra of ^{222}Rn with daughters in plastic (upper) and Teflon® (lower) vials

RESULTS AND CONCLUSION

To compare measurement parameters of the vial, we used dioxane-based LS cocktail GS-8[1] and alpha/beta discriminator mode of the spectrometer. We used the optimal pulse-shape analysis setting of 85 for both Teflon® and plastic vials. Figure 3 shows relative α and β spectra of ^{222}Rn with daughters for plastic and Teflon® vials, respectively. The spectrum shape and energy resolution for the Teflon® vial are much better than for plastic. We calibrated the three above-mentioned types of vials and designed canisters in a Rn atmosphere (Zelensky and Buzinny 1993). Reproducibility of the measurements is shown in Table 1. The standard deviation (SD) for "calibration" is better when exposure time is increased. Relative SD for "measurement" depends only on counting time (*ca.* 15% for 20 min). The following minimum detectable activity level was obtained for a 6-day exposure and for 20-ml vials: 1.0 Bq m^{-3} (background: 0.1 cpm; efficiency: 1.0 cpm per Bq m^{-3}). Future charcoal selection and silica gel additions are planned for improving Rn trapping.

TABLE 1. Count-Rate (cpm) Reproducibility in Charcoal Canister Exposure

Exposure time (h)	Comments	Beta	SD	Alpha	SD	No. of canisters
12	Measurement	7.5	0.8	6.9	0.7	10
24	Calibration	60.7	12.6	107.7	23.2	5
24	Measurement	7.2	0.7	5.6	1.2	10
36	Measurement	7.7	1.1	7.0	1.0	11
48	Calibration	56.4	2.8	98.7	4.2	5
48	Measurement	7.8	1.1	6.4	1.7	9

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REFERENCE

Zelensky, A.V. and Buzinny, M. G. 1993 Optimization of ^{222}Rn specific activity in air determination method based on charcoal and liquid scintillation counting. Actual problems of liquidation of medical consequences of Chernobyl accident (Abstract). In *Proceedings of the Ukrainian Scientific-Practical Conference.* Part I. Kiev, Health Ministry of the Ukraine, Ukrainian Research Center for Radiation Medicine: 124–125 (in Russian).

[1]Manufactured by Monokristalreactiv, Kharkov, Ukraine