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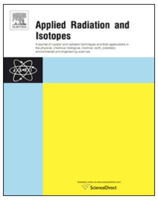

Liquid scintillation counting of polycarbonates: A sensitive technique for measurement of activity concentration of some radioactive noble gases

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HIGHLIGHTS

- This work explores the application of the liquid scintillation counting of polycarbonates for measurement of the activity concentration of radioactive noble gases.
- Results from experimental studies of the method are presented.
- Potential applications in the monitoring of radioactive noble gases are discussed.

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ABSTRACT

This work explores the application of the liquid scintillation counting of polycarbonates for measurement of the activity concentration of radioactive noble gases. Results from experimental studies of the method are presented. Potential applications in the monitoring of radioactive noble gases are discussed.

1. Introduction

In the last years the high absorption ability of polycarbonates to radon (Möre and Hubbard, 1997) and other noble gases became the basis for development of a new method for measurement of activity concentrations of some radioactive noble gases (RNGs) (Pressyanov et al., 1999, 2004). In this method, hereafter referred to as the polycarbonate method, polycarbonate specimens are exposed to RNG containing environment and are used to absorb (i.e. sample) the RNG. The activity of the absorbed RNG is proportional to the ambient RNG concentration and could be measured by different techniques: track etching of the polycarbonate (^{222}Rn measurements), measurement of the beta- particles emitted from the polycarbonate (gross beta counting for measurement of ^{222}Rn, ^{133}Xe and ^{85}Kr) or by gamma-spectrometry (^{222}Rn and ^{133}Xe measurements). The applications of the polycarbonate method include measurements of the activity concentration of ^{222}Rn (see Pressyanov et al. (2012) and the references therein), ^{85}Kr and ^{133}Xe in air (Pressyanov et al., 2004) and ^{85}Kr in air or water (Mitev et al., 2009a,b).

Recently, the liquid scintillation counting (LSC) was proposed as a sensitive technique within the polycarbonate method for measurement of the RNG alpha- or beta-radiation, emitted by the polycarbonates (Mitev et al., 2009b, 2012; Dimitrova et al., 2011). In this approach the polycarbonate samples are placed in a liquid scintillation (LS) vial which is then filled with LS cocktail and measured by LS counter. The first studies showed that RNG measurements by LS counting of polycarbonates are more sensitive than other traditional methods like gross beta-counting or gamma-spectrometry (Mitev et al., 2012). One advantage of this technique over the other methods for measurement of radioactive noble gases is that it does not require air sampling. Since the sampling of the noble gases from the environment is performed by absorption in the polycarbnonates, the sampling procedure can be unattended and performed simultaneously in a large number of points. This feature is particularly useful for monitoring of ^{85}Kr and ^{133}Xe in nuclear facilities during normal operation or accidents. The LSC counting of polycarbonates offers an efficient method for monitoring of these and other RNG isotopes with equipment that is typically available in nuclear installations. The technique can be

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considered as a complementary to the sophisticated stationary monitors that monitor the RNG concentrations in fixed locations in the nuclear installations. Moreover, the absorption in polycarbonates is not influenced by the humidity of the air (Pressyanov et al., 2003, 2004a), which has an adverse effect in the methods based on adsorption on activated charcoal (Scarpitta and Harley, 1990).

The objective of this communication is to present recent results about the application of LSC within the polycarbonate method. Results of experimental studies of the transparency of the polycarbonates to the liquid scintillation cocktail light as well as studies of the dependency of the measurement efficiency on the amount of the counted polycarbonate material are presented. A calibration of a RackBeta 1219 LS spectrometer is performed and estimates of the achievable minimum detectable activity concentrations (MDACs) are given. Potential applications of the proposed method for measurement of activity concentrations of RNGs are discussed.

2. Experimental

The conducted experiments are focused on studies of the properties of the LS counting of polycarbonate grains. The shapes of the LS spectra of ^{222}Rn and ^{85}Kr absorbed in grains and their peculiarities are investigated. A calibration of RackBeta 1219 LS spectrometer is performed and minimum detectable activities (MDAs) are estimated. The response of the counter to different amounts of counted polycarbonates and the transparency of the polycarbonates to the LS cocktail light are investigated.

2.1. Sampling radioactive noble gases from the environment by absorption in polycarbonates

When polycarbonates are exposed to air or water containing RNG, some of the RNG is absorbed in the polycarbonate material. The polycarbonates are known to have remarkably high absorption ability to radon (Möre and Hubbard, 1997) and, under certain circumstances, can concentrate radon and other RNG in their volume (Pressyanov et al., 2011b). The processes of sorption and desorption of radioactive noble gases in polycarbonates are studied in detail in Pressyanov et al. (2009) and are described by the diffusion equation, taking into account the radioactive decay:

$$\frac{\partial C(x,y,z,t)}{\partial t} = D\left(\frac{\partial^2 C}{\partial x^2}+\frac{\partial^2 C}{\partial y^2}+\frac{\partial^2 C}{\partial z^2}\right) - \lambda C, \tag{1}$$

where $C(x,y,z,t)$ is the RNG atom concentration in a moment t at location (x,y,z) within the polycarbonate. D is the diffusion coefficient of the atoms in the polycarbonate material and λ is the decay constant of the RNG. The solution of Eq. (1) depends on the exposure conditions (which are in fact boundary conditions for this equation) and on the polycarbonate's shape and thickness. For example, the solution of Eq. (1) for the RNG activity in a thin polycarbonate plate with thickness L and volume V after desorption for time t_d, which follows exposure to constant RNG concentration A_V for time t_s is (Pressyanov et al., 2009)

$$A(t_s,t_d) = \frac{8\lambda K A_V V L_D^2}{L^2}\sum_{k=0}^{\infty}\frac{(1-e^{-\lambda_{2k+1}t_s})}{\lambda_{2k+1}}e^{-\lambda_{2k+1}t_d}, \tag{2}$$

where $\lambda_j = \lambda(1+(j\pi L_D/L)^2)$, $j=2k+1$ and $L_D=\sqrt{D/\lambda}$. K is the partition coefficient at the polycarbonate border (see Pressyanov et al., 2011b for details). Similarly, the activity in a polycarbonate grain with cylindrical shape with height H and radius R after desorption for time t_d, following a sorption for time t_s is (Pressyanov et al., 2009)

$$A(t_s,t_d) = 32\lambda K A_V V \sum_{k=0}^{\infty}\sum_{m=1}^{\infty}\left(\left(\frac{L_D}{H\nu_m}\right)^2 + \left(\frac{L_D}{\pi R(2k+1)}\right)^2\right)\frac{(1-e^{-\lambda_{2k+1,m}t_s})}{\lambda_{2k+1,m}}e^{-\lambda_{2k+1,m}t_d}, \tag{3}$$

where $\lambda_{j,m} = \lambda(1+(\nu_m L_D/R)^2+(j\pi L_D/H)^2)$, $j=2k+1$ and ν_m is the mth zero of the Bessel function of zero order. The infinite sums in (Eqs. (2) and 3) are convergent and can be computed numerically (see Georgiev, 2012 for details). The physical parameters which determine the absorption ability of the polycarbonates are the partition coefficient K and the diffusion length L_D. These are physical properties of the polycarbonate material and were determined in dedicated experiments. Estimated values of the diffusion length L_D of ^{85}Kr, ^{133}Xe and ^{222}Rn in polycarbonates are published in Pressyanov et al. (2009). The partition coefficients for exposure of polycarbonates to ^{85}Kr, ^{133}Xe and ^{222}Rn in air and ^{85}Kr and ^{222}Rn in water are published in Pressyanov et al. (2011b). A method for simultaneous determination of the diffusion length and the partition coefficient K is published in Pressyanov et al. (2011a). The diffusion length and the partition coefficient depend on the temperature. Estimates of the temperature dependence are published in Pressyanov (2009) and Dimitrova et al. (2012).

To illustrate the potential of the above model Fig. 1 shows calculations of the spatial and time dependence of the activity in polycarbonate plates for t_s=24 h exposure in air at $A_V = 100$ Bq/m^3 of ^{85}Kr (Fig. 1(a)), ^{133}Xe (Fig. 1(b)) or ^{222}Rn (Fig. 1(c)). The width and height of the polycarbonate plates (with thickness 0.25 mm) are 1.6×4 cm and are very convenient for placing the plate in a standard LS vial. The temporal variations of the absorbed activities are shown in Fig. 1(d). The results in the figures show that among the three RNGs ^{222}Rn is mostly absorbed in the polycarbonates, followed by ^{133}Xe and ^{85}Kr.

Thus far, the developed model allows to estimate quantitatively the RNG activity absorbed in the polycarbonate during the exposure. It also allows to relate the absorbed activity in the polycarbonate to the ambient RNG activity concentration (if the exposure conditions are known) which, combined with the high absorption ability, makes the polycarbonates very appropriate RNG samplers. For the purposes of LSC measurements, a sampling efficiency (ε_s) can be defined (Mitev et al., 2012):

$$\varepsilon_s = \frac{A_{PC}}{A_{amb}}, \tag{4}$$

where A_{PC} is the RNG activity absorbed in the polycarbonate. A_{amb} is the ambient RNG activity contained in a volume, equal to that of the polycarbonate sample:

$$A_{amb} = VA_{V,amb}, \tag{5}$$

where $A_{V,amb}$ is the ambient RNG activity concentration during the exposure and V is the volume of the polycarbonate.

Obviously, the sampling efficiency is a measure of the ability of polycarbonate samplers to concentrate RNG in their volume. To facilitate the application of the above model in Figs. 2 and 3 we have plotted the dependence of the sampling efficiency on the sampling (exposure) time t_s for exposure of 15 g polycarbonate grains in air with constant ^{222}Rn concentration at 10 °C and 20 °C. The dependence of the sampling efficiency on the sampling time for exposure of 15 g polycarbonate grains in water with constant ^{222}Rn concentration at 10 °C is shown in Fig. 4. The above dependencies are fitted with a simple exponential function to provide rough estimates of the dependence $\varepsilon_s(t_s)$:

$$\varepsilon_s = \varepsilon_0 - \varepsilon_1 e^{-t_s/\tau}. \tag{6}$$

The parameters of the exponential fit (Eq. (6)) are as follows: exposure to ^{222}Rn in air at $T=10$ °C: $\varepsilon_0 = 2.248 \pm 0.034$, $\varepsilon_1 = 1.795 \pm 0.033$ and $\tau = 53.1 \pm 3.4$ (h); Exposure to ^{222}Rn in air at $T=20$ °C: $\varepsilon_0 = 2.944 \pm 0.044$, $\varepsilon_1 = 2.345 \pm 0.043$ and $\tau = 52.8 \pm 3.3$ (h); Exposure to ^{222}Rn in water at $T=10$ °C: $\varepsilon_0 = 9.81 \pm 0.15$, $\varepsilon_1 = 7.83 \pm 0.14$ and $\tau = 53.1 \pm 3.4$ (h). This fit approximation is valid for exposure times in the interval 6–168 h. The differences between the exponential fit and the sampling efficiency calculated by the model are within 10%