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Measurement of ^{222}Rn by absorption in plastic scintillators and alpha/beta pulse shape discrimination

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HIGHLIGHTS

- Common polystyrene- or polyvinyltoluene-based plastic scintillators absorb ^{222}Rn.
- The pulse decay times of alpha and beta particles in plastic scintillators are different.
- Alpha-/beta-pulse shape discrimination of the pulses of ^{222}Rn and its progeny is possible.
- Potential applications of plastic scintillators for ^{222}Rn measurements are suggested.
- The applications are based on ^{222}Rn absorption and pulse shape discrimination with plastic scintillators.

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ABSTRACT

This work demonstrates that common plastic scintillators like BC-400, EJ-200 and SCSF-81 absorb radon and their scintillation pulse decay times are different for alpha- and beta-particles. This allows the application of pulse shape analysis for separation of the pulses of alpha- and beta-particles emitted by the absorbed radon and its progeny. It is shown that after pulse shape discrimination of beta-particles' pulses, the energy resolution of BC-400 and EJ-200 alpha spectra is sufficient to separate the peaks of ^{222}Rn, ^{218}Po and ^{214}Po and allows ^{222}Rn measurements that are unaffected by the presence of thoron (^{220}Rn) in the environment. The alpha energy resolution of SCSF-81 in the experiments degrades due to imperfect collection of the light emitted inside the scintillating fibers. The experiments with plastic scintillation microspheres (PSM) confirm previous findings of other researchers that PSM have alpha-/beta-discrimination properties and show suitability for radon measurements. The diffusion length of radon in BC-400 and EJ-200 is determined. The pilot experiments show that the plastic scintillators are suitable for radon-in-soil-gas measurements. Overall, the results of this work suggest that it is possible to develop a new type of radon measurement instruments which employ absorption in plastic scintillators, pulse-shape discrimination and analysis of the alpha spectra. Such instruments can be very compact and can perform continuous, real-time radon measurements and thoron detection. They can find applications in various fields from radiation protection to earth sciences.

1. Introduction

Several new radon (^{222}Rn) measurement techniques have emerged in the last two decades. These techniques are based on absorption in polycarbonates or on absorption in polystyrene.

The high absorption ability of polycarbonates to ^{222}Rn is discovered by Möre and Hubbard (1997). In combination with the track-etch properties of the polycarbonates, it is the basis of perhaps the currently most accurate retrospective ^{222}Rn measurement method (Pressyanov et al., 2000, 2001, see also Pressyanov et al., 2012, Chapter 4 and the references therein). Tommasino et al. (2009) proposed a new generation of passive ^{222}Rn monitors, referred to as radon-film badges which are formed by thin film plastic radiators which face an appropriate detector (Tommasino et al., 2010, Tommasino and Tokonami, 2011a,b). Independently, the polycarbonate material is proposed as a sampler of ^{222}Rn and other radioactive noble gases (RNG) like ^{85}Kr and ^{133}Xe (Pressyanov et al., 2004a,b, 2007; Mitev et al., 2009a). Appropriate methods for the measurement of RNG activity absorbed in polycarbonates like alpha track-etching (Pressyanov et al., 2000, 2004a; Dimitrova et al., 2011), gross-beta counting (Pressyanov et al., 2004b; Georgiev et al., 2012) or gamma-spectrometry (Pressyanov et al., 2007; Mitev et al., 2009a) are developed. More recently, the liquid scintillation counting (LSC) of polycarbonates is

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proposed as a sensitive technique for measurement of ^{222}Rn (Mitev et al., 2012, 2014a,b) or ^{85}Kr (Mitev et al., 2009b) concentrations in the environment. Owing to the high RNG absorption ability and the high refractive index of polycarbonates, Mitev (2013) proposed the Cherenkov counting technique for the measurement of RNG absorbed in the polycarbonate.

Similar ^{222}Rn measurement technique, based on polystyrene foam absorption and LSC, is proposed by Saito (1999). In this approach, termed absorptive polymer dissolved liquid scintillation counting (APDLS), the polystyrene foam is used as a sampler and after the sampling is dissolved in LS cocktail to measure the absorbed ^{222}Rn activity (Saito and Takata, 2000; Saito et al., 2003). Further, Saito et al. (2006) proposed the absorption of ^{222}Rn in a polystyrene-based plastic scintillator (PS) as a method for the measurement of ^{222}Rn activity concentration in water.

Generally, the plastic scintillators can be used as alpha-particle detectors (Morishita et al., 2014). They are also proposed as an alternative to the liquid scintillation (LS) cocktail in measurements with LS spectrometers (Tarancón et al., 2002a,b, 2003). Polystyrene-based PS beads can be used for routine determination of the activity of low-energy beta emitters (Tarancón et al., 2004), and as a radio-chemical sensor with application in measurement of liquid effluents (Tarancón et al., 2005a,b). The application of PS beads to radionuclide mixture characterization is demonstrated by Tarancón et al. (2007) and Bagán et al. (2009). Bagán et al. (2010) demonstrate that it is possible to perform alpha/beta pulse shape discrimination (PSD) with the PS. Successive works are devoted to the synthesis of plastic scintillation microspheres (PSM) (Santiago et al., 2013) and to the demonstration and evaluation of their PSD capabilities (Santiago et al., 2014).

Thus far, it is known that polycarbonate and polystyrene absorb radon and that it is possible to perform PSD with polystyrene-based plastic scintillation microspheres. However, the combination of the absorption and the PSD properties of plastic scintillators to be applied to radon measurements has not been proposed and studied yet. The application of commercially available plastic scintillators (either polystyrene- or polyvinyltoluene-based) for such type of radon measurements is also not studied. Moreover, a recent systematic compilation of earthquake precursors by Cicerone et al. (2009) shows that changes in radon gas emissions (radon anomalies) can be regarded as earthquake precursors. The survey reports the finding of 125 observations of changes in radon gas emissions from 86 earthquakes. Other researchers point out that radon anomalies can be regarded as earthquake and volcano precursors (see, for example, Cigolini et al., 2007). In regard to the research in the applied earth sciences, it is reasonable to study the possibilities which plastic scintillators provide for in situ radon-in-soil gas measurements and radon monitoring.

The objective of this work is to investigate the possible application of PS to the measurement of radon with the study of the alpha and beta PSD signals as well as the radon absorption in the PS volume. A new radon measurement technique, which is based on ^{222}Rn absorption in plastic scintillators, alpha/beta pulse shape discrimination and analysis of the alpha spectra is proposed and studied. The ^{222}Rn diffusion in common plastic scintillators, their alpha/beta pulse shape discrimination capabilities and energy resolution in the alpha channel are studied. Pilot experiments on exposure of plastic scintillators to radon-in-soil-gas are performed and they demonstrate the applicability of this approach. Potential applications for continuous radon monitoring and thoron detection are being proposed and discussed.

2. Experimental

The first step of the experimental studies was laboratory exposure of PS specimens to ^{222}Rn in air. It was followed by PSD studies with the PERALS® spectrometer (ORDELA, USA). Next, the stability of counting of ^{222}Rn absorbed in PS is studied by RackBeta 1219 (Wallac, Finland) and PERALS follow-up measurements of PS samples immersed in glycerin or water in closed LS vials. Further, RackBeta follow-up measurements were performed in order to study the desorption of radon from the PS specimens. The potential for practical applications was studied by radon-in-soil gas exposure of PS specimens with subsequent PERALS and/or RackBeta measurement of the absorbed activity. Five common plastic scintillators were studied:

- PSW-51 polyvinyltoluene-based PS, produced in the past by TESLA;
- BC-400 polyvinyltoluene-based PS, produced by Saint-Gobain Crystals;
- EJ-200 polyvinyltoluene-based PS, produced by Eljen Technology;
- SCSF-81 polystyrene-based PS fibers produced by Kuraray;
- PSM – polystyrene-based plastic scintillation microspheres, produced at the Department of Analytical chemistry, University of Barcelona.

Specimens with plate (BC-400, EJ-200) or cylindrical (PSW-51, SCSF-81) shape with mass below 1 g were prepared from each PS. The sizes of the specimens and the continuous-slowing-down approximation (CSDA) ranges of the alpha- and beta-particles of ^{222}Rn and its progeny in these materials are given in Table 1. As it can be seen, the dimensions of BC-400, EJ-200, PSW-51 and SCSF-81 are much larger than the alpha-particle ranges in these materials and are comparable to the maximal ranges of the beta-particles. The latter is not true for the PSM, which have different diameters forming a particle size distribution which depends on the production process (Santiago et al., 2013, 2014). The particle size distribution of the batch of PSM used in this work has a mean radius $\bar{R}$ = 0.0343 mm (median radius 0.0342 mm) with 10% of the PSM having R < 0.020 mm and 91.6% of the PSM having R < 0.050 mm. Thus, the PSM particle sizes are comparable to the alpha-particle ranges and much smaller than the beta-particle ranges.

The specimens were exposed to different, high ^{222}Rn

Table 1
Characteristics of the plastic scintillators used in the PERALS experiments. The CSDA ranges are calculated with the NIST ASTAR and ESTAR database (Berger et al., 2010).

PS	Density g/cm^3	Dimensions $X \times Y \times Z$ mm	$R_{CSDA,E_{\alpha 1}}$, mm $E_{\alpha 1}$ = 5.489 MeV	$R_{CSDA,E_{\alpha 2}}$, mm $E_{\alpha 2}$ = 6.002 MeV	$R_{CSDA,E_{\alpha 3}}$, mm $E_{\alpha 3}$ = 7.686 MeV	$R_{CSDA,E_{\beta,max\ 1}}$, mm $E_{\beta,max\ 1}$ = 1.019 MeV	$R_{CSDA,E_{\beta,max\ 2}}$, mm $E_{\beta,max\ 2}$ = 3.270 MeV
PSW-51	1.023[a]	h = 10.8 mm, R = 3.4 mm	0.0409	0.0471	0.0701	4.33	15.1
BC-400	1.032	17.3 × 6.8 × 1.69	0.0414	0.0477	0.0710	4.34	15.1
EJ-200	1.023	16.3 × 7.0 × 2.27	0.0409	0.0471	0.0701	4.33	15.1
SCSF-81	1.05 (core)	h = 4 mm, R = 0.5 mm	0.0407	0.0468	0.0698	4.27	14.9
PSM	1.05	$\bar{R}$ = 0.0343 mm	0.0407	0.0468	0.0698	4.27	14.9

[a] Density not specified by the producer. The same density as EJ-200 is assumed, due to the same polyvinyltoluene base of both scintillators.