



Measurement of ^{222}Rn and ^{226}Ra in water by absorption of radon in polycarbonates and etching alpha-tracks

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ABSTRACT

This work describes a new method for measurement of the activity concentrations of ^{222}Rn and ^{226}Ra in water by exposure of a polycarbonate detector in the water and etching it for alpha-tracks. A theoretical model that gives the in-depth distribution of tracks inside polycarbonate detectors exposed in water is developed and used to optimize the method. The presented experimental results demonstrate the feasibility of the method for measurement of activity concentrations of both ^{222}Rn and ^{226}Ra in water. The lower detection limit is estimated at 0.06 kBq m^{-3} for 30 days exposure time and is achieved without preconcentration of the water or radiochemical separation of ^{226}Ra . The method allows radon measurements directly in the water source, which could be carried simultaneously in a large number of points.

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1. Introduction

The concentrations of ^{222}Rn and ^{226}Ra in water are of interest for radiation protection, geology, seismology and environmental science. A lot of attention is focused on ^{222}Rn (radon) due to its major contribution to the effective dose received from natural radioactive sources (UNSCEAR, 2000). The risk from exposure to ^{226}Ra is also acknowledged and monitoring ^{226}Ra in water supplies is required in some countries (Bulgarian Council of Ministers, 1986; Environmental Protection Agency, 2000). Radium-226 in water is usually measured by its product ^{222}Rn . Although, there are methods that directly measure ^{226}Ra and could be applied for measurement of very low activity concentrations (below 1 Bq m^{-3}) they typically involve radiochemical procedures (Eikenberg et al., 2001; Jia et al., 2005; Karamanis et al., 2006). Most of the methods for measurement of radon activity concentration in water are based on laboratory analysis of grab samples. The methods that could be used “on-sight” involve transfer of radon to air (or some other carrier gas) and apply radon-in-air measurement techniques (Freyer et al., 2003; Lee and Kim, 2006; Schmidt et al., 2008; Schubert et al., 2006).

In this work we propose a method for measurement of the activity concentration of ^{222}Rn and ^{226}Ra in water by exposure of

a polycarbonate detector directly in the water and etching it for alpha-tracks. The method is based on the high absorption ability of some polycarbonates to radon (More and Hubbard, 1997). When Makrofol¹ or equivalent polycarbonate is exposed to a radon-containing media, radon is absorbed inside the polycarbonate (see Fig. 1). The distribution of ^{222}Rn in the polycarbonate is governed by diffusion and radioactive decay (a quantitative description of these processes is given by Pressyanov et al., 2009). The activity of ^{222}Rn and its short-lived progenies inside the polycarbonate is proportional to the ambient activity concentration of radon. On this basis a method for express measurement of radon in water has been developed, in which the beta- or gamma-radiation of the short-lived progenies of ^{222}Rn (Fig. 1) in the polycarbonate is registered (Pressyanov et al., 2007). In contrast, the method proposed in this work employs a different approach – Makrofol is used as a nuclear track detector inside which the alpha-particles emitted by the absorbed radon and its progenies (Fig. 1) form tracks. This approach has first been applied for measurements in air (Pressyanov et al., 2000) and has shown potential for precise retrospective measurements of ^{222}Rn in dwellings (Pressyanov et al., 2001, 2003, 2004). For the case of exposure in air, it has been shown that the track density (number of tracks per unit area) at a given depth beneath the surface of the polycarbonate detector is proportional to the

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¹ Makrofol® is a trademark of Bayer.

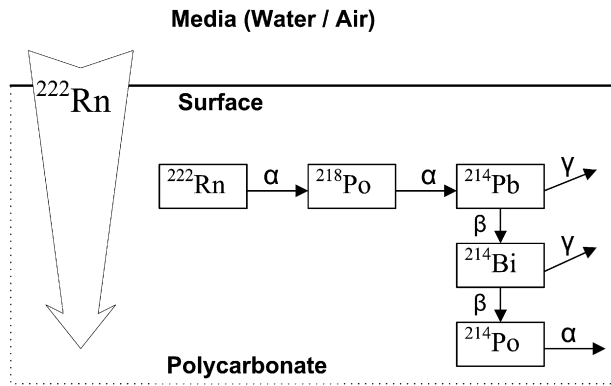


Fig. 1. Illustration of the basic idea behind the methods for measurement of ^{222}Rn by absorption in polycarbonates: the activity of the absorbed radon and its short-lived progenies is related to the activity concentration of ^{222}Rn in the ambient media.

activity concentration of radon in the air integrated over the exposure time (Pressyanov et al., 2000, 2001, 2003, 2004).

The main goal of this work is to present a method for measurement of the activity concentration of ^{222}Rn in water by alpha-track etching of polycarbonates. The feasibility of this approach is tested by dedicated experiments, in which polycarbonates were exposed to known concentrations of radon in water. A theoretical model which gives the dependence of the track density on the depth beneath the surface of polycarbonate detectors is developed. The results of the model are compared with the experimental results. The model is further used to optimize the method. Calibration coefficients for measurement of the activity concentration of radon in water by alpha-track etching of polycarbonates are obtained. The measurement range and the achievable lower detection limit are estimated. Results from pilot experiments on measurement of the activity concentration of ^{226}Ra in water by the proposed method are also presented.

2. Theoretical model

2.1. Basic steps

The track density distribution in polycarbonate detectors has been studied experimentally (Pressyanov et al., 2003) and theoretically (Pressyanov, 2009) for exposures in air. The theoretical model agrees well with the experimental results (Pressyanov, 2009) and both show that the track density decreases for depths greater than $79\ \mu\text{m}$. However, neither experiments nor modeling has been performed for depths smaller than $70\ \mu\text{m}$. That is because at smaller depths part of the tracks can be formed by progenies of ^{222}Rn and ^{220}Rn plated-out on the polycarbonate's surface or present in the air. Since the activity of these progenies cannot be easily correlated to the activity concentration of radon in the air, etching has been performed at depths which their alpha-particles could not reach (i.e., $d > 79\ \mu\text{m}$).

The developed theoretical model describing the in-depth track density distribution in plate detectors exposed to radon in water is presented in detail in the Appendix. It is based on the model used for detectors exposed in air (Pressyanov, 2009). However, it is expanded to include depths in the range $0 \div 79\ \mu\text{m}$. The principle steps in the model are:

- Modeling the source of alpha-particles
 - Three nuclides emit alpha-particles – ^{222}Rn , ^{218}Po and ^{214}Po . Inside the polycarbonate it can be considered that radon and its short-lived progenies are in radioactive equilibrium and their activities follow the same volume distribution.

- The distribution of ^{222}Rn in the polycarbonate detector is found by solving the diffusion equation in which a term is added to account for radioactive decay. The solution is obtained for a given temperature, since the diffusion coefficient of radon in polycarbonates (D) is temperature dependent.
- Water is considered a homogenous source, in which radon and its progenies are in equilibrium.
- Modeling the detector response.
- Deriving calibration coefficients. A quantitative estimate of the calibration coefficient for a given depth below the surface can be made if the solubility (K) of radon from water to polycarbonate is known. The solubility is defined as the ratio between the concentration of radon at the surface of the polycarbonate and the ambient concentration. In addition, the efficiency of the particular track-counting procedure should also be known.

The model is applied for detectors with thickness $L = 570\ \mu\text{m}$ exposed in water at temperature of $21\ ^\circ\text{C}$ (the same detectors and conditions as in the experiments described in Section 3). The value of D for this temperature is obtained using data from Pressyanov, 2009.

2.2. Contribution of the water source to the signal

Water can be ignored as a direct source of alpha-particles (provided that there are no other alpha-emitters present in the water with activities comparable to these of radon and its progenies). This is because the contribution of radon and its progenies in the water to the surface track density is negligible compared to the contribution of radon and its progenies inside the polycarbonate. To illustrate this, the model described in the Appendix is used to estimate the track density formed at the detector's surface for exposure in water to integrated activity concentration of radon $I_{\text{out}} = 1\ \text{kBq m}^{-3}\ \text{h}$. The following estimates are obtained:

- The surface track density formed by alpha-particles emitted in the water is $1.6 \times 10^{-3}\ \text{cm}^{-2}$. The estimate is made in the case of equilibrium between radon and its progenies in the water and possible deviations from the equilibrium will lead to smaller surface track density.

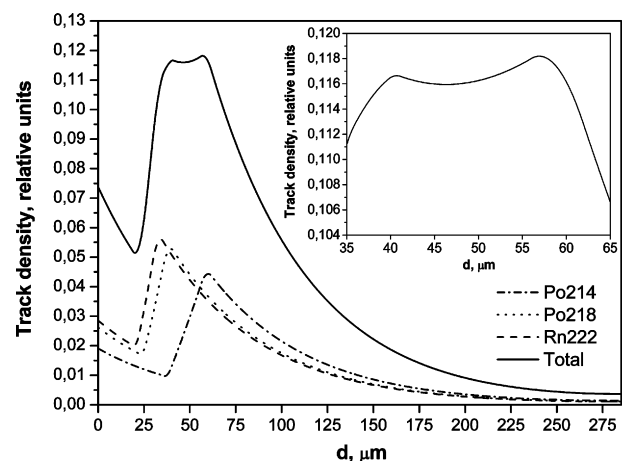


Fig. 2. In-depth track density distribution inside polycarbonate detectors with thickness $L = 570\ \mu\text{m}$ exposed in water at temperature $21\ ^\circ\text{C}$, obtained by the model given in the Appendix. The distribution is symmetrical with respect to the middle of the detector and half of it is shown. The solid line represents the total track density, obtained by summing the three components due to radon and its progenies (the dashed lines). The region where the track density is highest is shown magnified.

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