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PSA discriminator influence on ^{222}Rn efficiency detection in waters by liquid scintillation counting

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HIGHLIGHTS

- Modification of calibration procedure (EPA method 913.0) for precise radon determination in waters by LSC is presented.
- ^{222}Rn efficiency dependence on PSA variation and activity concentration of ^{226}Ra referential standard used was examined.
- Quench effects on reduction of efficiency detection were also studied and parameterized.
- Modified radon determination procedure according to activity of ^{226}Ra standard used, dependent on PSA setup, was evaluated.
- Verification analysis included drinking water/^{226}Ra solution samples with assessment of measurement uncertainty variation.

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ABSTRACT

A procedure for the ^{222}Rn determination in aqueous samples using liquid scintillation counting (LSC) was evaluated and optimized. Measurements were performed by ultra-low background spectrometer Quantulus 1220™ equipped with PSA (Pulse Shape Analysis) circuit which discriminates alpha/beta spectra. Since calibration procedure is carried out with ^{226}Ra standard, which has both alpha and beta progenies, it is clear that PSA discriminator has vital importance in order to provide precise spectra separation. Improvement of calibration procedure was done through investigation of PSA discriminator level and, consequentially, the activity of ^{226}Ra calibration standard influence on ^{222}Rn efficiency detection. Quench effects on generated spectra i.e. determination of radon efficiency detection were also investigated with quench calibration curve obtained. Radon determination in waters based on modified procedure according to the activity of ^{226}Ra standard used, dependent on PSA setup, was evaluated with prepared ^{226}Ra solution samples and drinking water samples with assessment of measurement uncertainty variation included.

1. Introduction

The ^{222}Rn concentration in water originates from decay of ^{226}Ra present in the rock and soil. The concentration of radon in groundwater or well water depends strongly on the character of the host rock. Because of the solubility of radon in water, very high levels of radon can occur even at the hot water (Lee et al., 2012). Many uses of water release radon into the indoor air, which contributes to the total indoor airborne radon concentration. Ingestion of radon in water is also thought to pose a direct health risk through irradiation of sensitive cells in the gastrointestinal tract and in other organs once it is absorbed into the bloodstream. Thus, radon in drinking water could potentially produce adverse health effects in addition to lung cancer (National Research Council, 1999). Taking into account that ^{222}Rn is the most substantial natural source of population radiation exposure, it is important to provide accurate and reliable measurement methods of radon activity monitoring in air, water and soil.

EPA method 913.0 [EPA 913.0] had been successfully applied in Laboratory for low level radioactivity measurements in Novi Sad (Faculty of Sciences, Department of Physics) (Todorović et al., 2014, Stojković et al., 2015) for radon activity measurements in aqueous samples.

Ultra-low level LS counter Quantulus 1220 coupled to alpha-beta discrimination – PSA (Pulse Shape Analysis) circuit – allows rapid and simple determination of gross alpha and beta activities that are simultaneously measured (Todorović et al., 2012), therefore it can be applied for ^{222}Rn measurements. The physical basis of α/β discrimination procedure lies in the fact that alpha particles

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cause signals with an important time delayed component due to the de-excitation of triplet states of solute molecules. At the same time beta particles give signals with a shorter time decay period due to faster de-excitation of singlet states of solute molecules (Villa et al., 2003).

This paper presents investigation of calibration procedure described in method (EPA method 913.0), i.e. ^{222}Rn efficiency detection dependence on PSA variation and activity concentration of ^{226}Ra referential standard used.

2. Experimental

Results of experiments were obtained using Ultra Low Level Liquid Scintillation Spectrometer Wallac 1220 Quantulus manufactured by PerkinElmer. The spectra were acquired by WinQ and analyzed by EasyView software.

A standard radioactive source activity (aqueous ^{226}Ra solution) produced from Czech Metrology Institute, Inspectorate for Ionizing Radiation, was used for instrument's calibration, certified activity A(^{226}Ra)=39.67 Bq ml^{-1} with combined standard uncertainty 0.5%, reference date 1/10/2013. For PSA setup aqueous ^{241}Am and ^{90}Sr/^{90}Y solutions produced from Czech Metrology Institute, Inspectorate for Ionizing Radiation, were also used: certified activity A(^{241}Am)=37.57 Bq ml^{-1} with combined standard uncertainty 0.2%, reference date 1/10/2013, and A(^{90}Sr/^{90}Y)=38.18 Bq ml^{-1} with combined standard uncertainty 0.5%, reference date 1/10/2013. All LSC samples were prepared in 20 ml high performance glass vials with Teflon-lined caps (Perkin Elmer) using scintillation cocktail OptiPhase HiSafe 3. EPA method [EPA 913.0] suggests usage of Mineral Oil Scintillator which is immiscible with water and induces two phases in vials. First modification of method [EPA 913.0] implemented in this paper was selection of cocktail miscible with water, OptiPhase HiSafe 3, i.e. application of so-called single phase method, which leads to a formation of homogeneous samples. Advantages and disadvantages of both two-phase and single-phase method are discussed in detail in literature (Salonen, 2010). According to (Kitto, 1994), glass vials must be capped using silicone rubber septa with an attached Teflon liner in order to avoid radon emanation.

For efficiency reduction investigation, nitromethane (100% CH_3NO_2) in increasing amount was added in a set of ^{226}Ra standards as a quenching agent in order to obtain quench calibration curve. The measurement of an external standard quench parameter [SQP(E)] on Quantulus 1220 requires the recording of two spectra during the same counting time: the first one measured when the sample is exposed to the γ-radiation of the ^{152}Eu external source (Compton recoil electron events plus the sample events), and the second one measured from the sample alone. The net external standard spectrum is obtained by subtracting the two spectra (Minne et al., 2008). The level of quench in the sample is evaluated when obtained subtracted spectra is compared with the theoretical unquenched Compton spectra. The end point SQP(E) is calculated as the channel above which 1% of all signal of the net external standard spectrum is found (Minne et al., 2008).

Radium Solution Method (EPA method 913.0) was used for calibration and standardization, where 100 ml of ^{226}Ra solution was prepared such that the final activity was 1.31 Bq ml^{-1}. According to single phase method, 10 ml of the diluted standard was transferred into the 20 ml scintillation vial, to which it has been added 10 ml of OptiPhase HiSafe 3 cocktail. The background samples were prepared using 10 ml of distillated water. The standards and background samples were set aside for a 30 days to allow radon to attain secular equilibrium with radium and to dark adapt before counting. Radon diffuses from the sample into scintillation cocktail from which it has a much greater affinity than for water. The standards were counted for 50 min in a liquid scintillation counter Quantulus 1220 equipped with PSA circuit for energy discrimination for alpha particles in order to determine calibration factor *CF*.

For radon determination in drinking water samples, 10 ml of water sample was pipetted into a glass scintillation vial to which it had been added 10 ml of scintillation cocktail. Report from (Kitto, 1994) states that a few seconds time delay prior to sample extraction from opened vials has no effect on the radon level. The samples were shaken and set aside in the dark (laboratory temperature being maintained at 20 °C) for at least three hours to reach secular equilibrium between radon and its short-lived progeny and to dark adapt (in order to eliminate chemiluminescence reactions) before counting. Otherwise, if vials stored at room temperature where they reached equilibrium are loaded into a Quantulus, the temperature of the liquids would decrease and the system would try to reach (new) equilibrium specific for this lower temperature, which could provide erroneous measurements. It had been reported (Gomez Escobar et al., 1996) that diffusion of ^{222}Rn in the organic phase is independent of shaking the vial during the preparation of samples. On the other hand, (Kitto, 1994) found that initial shaking (3–40 s) is inversely proportional to time needed to establish equilibrium though all shaken cocktails established it within five hours, while the unshaken ones took 25 h to reach equilibrium.

For optimal window selection, radon standard with activity 1.31 Bq ml^{-1} was prepared and counted for 5 min 30 days after its preparation. The region of greatest alpha activity is the region by two large peaks at the high end of the energy spectrum. The optimal window was formed by extending the region by 10% on each side of the alpha peaks, and set from channels 420–780 (Fig. 1). ^{222}Rn (5489.7 keV, $T_{1/2}$=3.825 days) has two short-lived alpha-emitting daughters: ^{218}Po (6002.55.7 keV, $T_{1/2}$=3.11 min) and ^{214}Po (7686.90 keV, $T_{1/2}$=163.69 μs) (Galan Lopez et al., 2004), as marked on Fig. 1.

Here it is important to mention one disadvantage of the operation of the guard counter of Quantulus 1220™. If the guard counter is on, the nearly simultaneous decay of radon decay product ^{214}Bi (high-energy γ radiation) in the presence of ^{214}Po alphas cause about 8% loss of ^{214}Po counts (some of the ^{214}Po alpha counts are rejected). Therefore, not all alpha emissions are counted with the same efficiency which leads to slightly wrong calibration. In (Salonen and Hukkanen, 1997) it is reported that radon measurements are performed with 8% lower efficiencies.

2.1. PSA optimization

Quantulus 1220 has two multichannel analyzers (MCA), one is used for active shield and the second one is used for spectra record. Those MCA's are divided in two halves, 1024 channels each. The optimum setting for discrimination between alpha and beta particles is the setting where there is equal and minimum interference between alpha and beta events, i.e. minimum spill of alpha pulses into β – MCA and beta pulses into α – MCA (Todorović et al., 2012), $(\tau_\alpha)_{minimal}=(\tau_\beta)_{minimal}$. This occurs at the crossover point of the alpha–to–beta spillover and beta–to–alpha spillover curves, as presented in Fig. 2, and that is the point defined as the working spillover for which PSA discriminator is adjusted. For the alpha and beta emitting radionuclide, the factors τ_α and τ_β were calculated using the formulae:

$$\tau_\alpha[\%]=\frac{\alpha-counts\ in\ \beta-window}{total\ \alpha-counts}\cdot 100$$
$$\tau_\beta[\%]=\frac{\beta-counts\ in\ \alpha-window}{total\ \beta-counts}\cdot 100 \quad (1)$$