OPTIMIZATION OF ^{90}Sr SCREENING METHOD IN WATERS VIA CHERENKOV RADIATION

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INTRODUCTION

Radiostrontium (^{90}Sr), having half-life of 28.8 years, is a product of nuclear fission and is present in spent nuclear fuel and in radioactive waste from nuclear reactors, as well as in fallout from nuclear weapon tests and nuclear accidents. Conventional methods of ^{90}Sr measurement in water are time-consuming and require sophisticated radiochemical separation techniques followed by gas proportional or liquid scintillation counting [e.g. 1,2,3]. Here we present optimization and validation of a simple method for ^{90}Sr screening in water via Cherenkov radiation detected by a liquid scintillation counter [3-5].

The ^{90}Sr beta spectrum has a maximum energy of 546 keV, i.e. well above the Cherenkov threshold in pure water (263 keV). More realistic is the Cherenkov counting of ^{90}Sr daughter, the ^{90}Y beta spectrum, which has a maximum energy of 2283 keV. ^{90}Y has a half-life of 64.1 h, thus, this pair is in secular equilibrium in environmental samples [3,4].

EXPERIMENTAL

Two laboratories participated in this study: Laboratory for low radioactivity at the Department of Physics, University of Novi Sad (UNS), Serbia, and Laboratory for measurement of low-level radioactivities of the Ruđer Bošković Institute (RBI) in Zagreb, Croatia. Each laboratory prepared independently calibration solutions and determined measuring conditions.

A standard radioactive source (aqueous ^{90}Sr/^{90}Y solution) of certified activity $A(^{90}\mathrm{Sr})$ = 38.18 Bq mL^{-1} with combined standard uncertainty 0.5%, reference date 1/10/2013, produced by the Czech Metrology Institute, Inspectorate for Ionizing Radiation, was used for instrument's calibration.

Several solutions were prepared by adding between 10 μL and 500 μL of the certified activity to 20 mL of distilled (at RBI: ultra pure) water in different types of vials: plastic (Super PE vial Cat.No. 6008117), glass (High Performance Glass Vial, Cat.No. 6001009) and low-potassium glass vial (High Performance Glass Vial, Cat.No. 6000134) at UNS and plastic vials at RBI (Super low diffusion PE vial, Cat.No. 1200-422).

Measurements were performed using the same type of the counter, Ultra Low Level Liquid Scintillation Spectrometer Quantulus 1220™ (Wallac Oy, PerkinElmer, Finland). The spectra were acquired and evaluated by WinQ and EASYView software in both laboratories.

Strontium activity concentration A (in Bq L^{-1}) was computed as:

$$A = \frac{R_s - R_b}{\varepsilon\, V} \qquad (1)$$

where R_s and R_b are sample and background count rates, respectively, V is the original sample volume analyzed, and ε is the counting efficiency.

Detection limit DL was determined by using the uncertainty of the background count rate $\sigma(R_b)$ [3]:

$$DL = 3.29\, \frac{\sigma(R_b)\,\sqrt{2}}{\varepsilon\, V} \qquad (2)$$

RESULTS

The obtained spectra of several calibration solutions are shown in Figure 1. Background spectrum did not exhibit any structure in the region of $^{90}Sr/^{90}Y$ spectra. Consequently, the channels comprising the whole spectra were chosen for numerical data acquisition: channels in the regions 130 – 430 and 100 – 500 were chosen at UNS and RBI, respectively (Table 1).

Calibration curves were obtained by comparison of measured count rates of prepared solutions of the certified standard and the activity added to water (Figure 2). The measurement efficiency ε was determined as the slope of the fitted line (linear fit with intercept set to 0). Determined measured efficiencies, background count rates and detection limit DL for different types of vials are presented in Table 1. Plastic vials give higher efficiency and lower background count rate than glass vials. It should be noted that different plastic vials used at UNS and RBI give slightly different values, indicating that one should carefully check the performances of vials used.

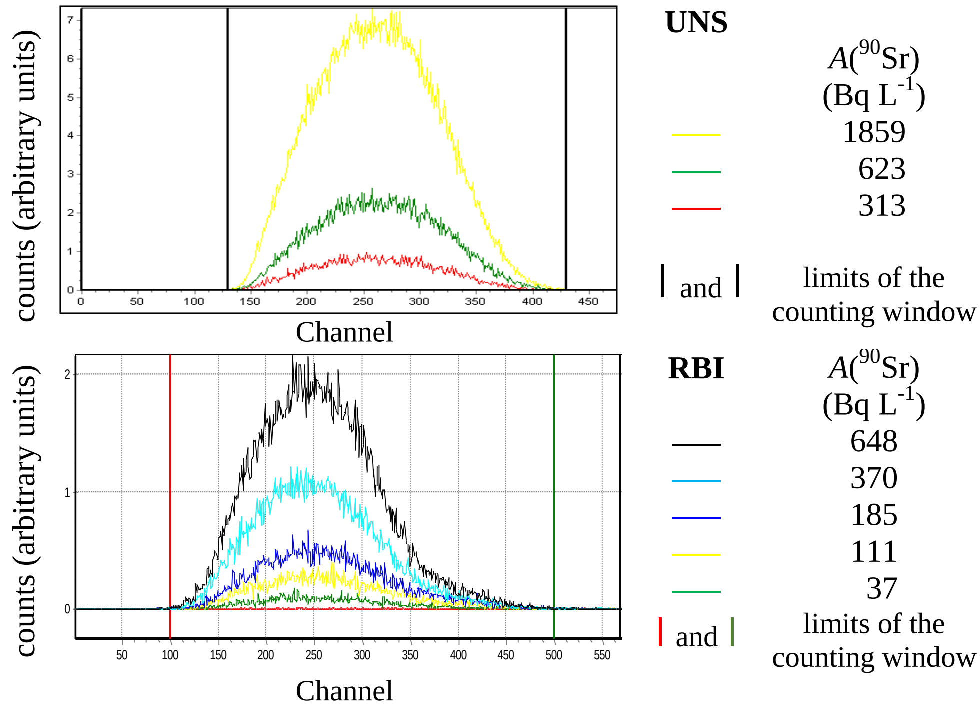

Figure 1. Spectra generated by Cherenkov radiation from ^{90}Sr.

Table 1. Measured efficiencies (ε) in the counting window, background count rates (R_b), and detection limit (DL) for different types of vials.

Type of vial	**Window** (ch)	**Efficiency** $\boldsymbol{\varepsilon}$ (%)	$\boldsymbol{R_b}$ (s^{-1})	$\boldsymbol{DL}$ (Bq L^{-1})
Plastic UNS	130 – 430	45.86 ± 0.09	0.008	0.32 (300 min)
Glass	130 – 430	32.45 ± 0.19	0.021	-
Low ^{40}K glass	130 – 430	32.24 ± 0.14	0.014	-
Plastic RBI	100 – 500	37.7 ± 0.3	0.006	0.34 (300 min)

INTERCOMPARISON

Five samples with known spiked activity of standard radioactive source ^{90}Sr of certified activity $A(^{90}Sr)$ = 38.18 Bq mL^{-1} were independently measured at UNS and RBI (Table 2). Measured activities were compared with the spiked activity values by means of the *z*-score values

$$z = \frac{A - A_{sp}}{\sigma} \qquad (3)$$

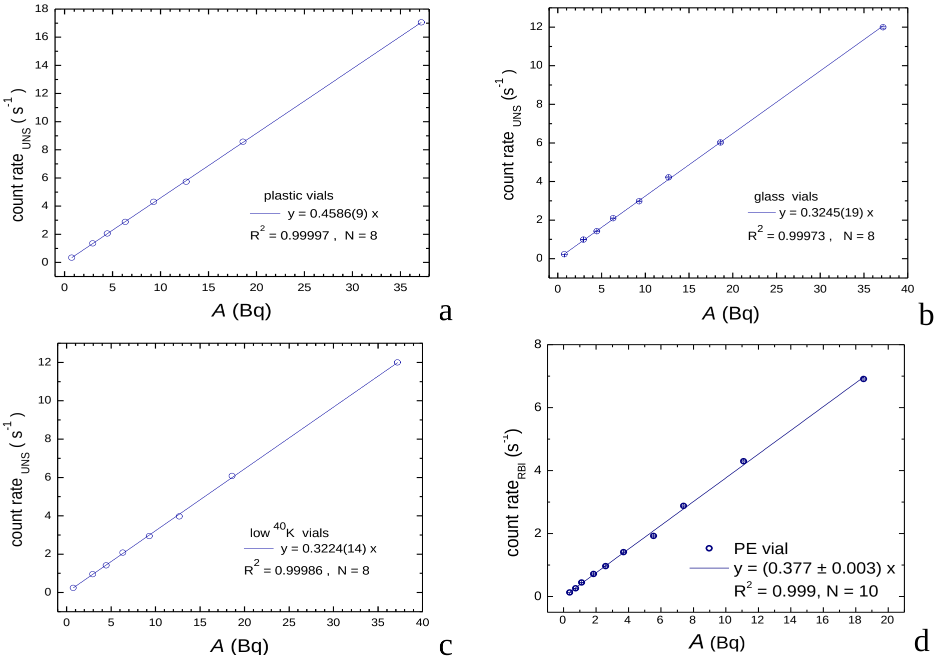

Figure 2. Calibration of ^{90}Sr measurements in water, V = 20 mL: a) plastic vials, UNS, b) high-performance glass vials, UNS, c) low-potassium glass vials, UNS, d) plastic vials (Super low diffusion PE vial), RBI.

where A_{sp} is the spiked activity, and A is the measured activity and σ the measurement uncertainty determined at either the UNS or the RBI.

Table 2. Results of the interlaboratory comparison of spiked samples.

Sample No.	A_{sp} (Bq L^{-1})	**UNS results**		**IRB results**	
		A (Bq L^{-1})	z	A (Bq L^{-1})	z
1	37.19	38.5 ± 0.6	1.96	37.8 ± 0.6	0.93
2	74.38	76.3 ± 2.4	0.80	76.1 ± 1.0	1.74
3	111.57	112.6 ± 1.3	0.77	108.7 ± 1.3	-2.21
4	148.76	149.6 ± 0.5	1.70	150.4 ± 1.7	1.00
5	185.95	187.9 ± 2.4	0.80	184.0 ± 2.0	-0.97

The A(^{90}Sr) values determined in both laboratories did not deviate significantly from the spiked activities, giving the z-scores in the interval [-2,2]. Only one sample at RBI had the z-score higher than 2 but lower than 3. All UNS results had positive z-scores.

Validation of the method at UNS was performed by determination of strontium level in a spiked sample within the IAEA world-wide proficiency test TEL-2014-03. The UNS result (A = 16.8 ± 0.4 Bq L^{-1}) was compared with the IAEA reference value of 14.93 ± 0.14 Bq L^{-1} and was acceptable in terms of accuracy, but not acceptable in terms of precision.

CONCLUSION

A simple method for screening ^{90}Sr activity in water samples has been successfully implemented in two laboratories in Novi Sad and Zagreb. Various types of vials were tested and different efficiencies were obtained indicating the importance of the choice of vials. Better performances (higher efficiency, lower background count rate) were obtained for plastic vials than for the glass ones. Detection limit for 300 min long measurements in plastic vials is 0.32 Bq L^{-1} and 0.34 Bq L^{-1} at UNS and RBI, respectively. The $^{90}Sr/^{90}Y$ Cherenkov radiation measurements of water samples can be used for rapid screening of samples without any pretreatment of the samples.

Acknowledgment

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Rapid screening of strontium activity in waters via Cherenkov radiation detection by ultra-low-level liquid scintillation counter Quantulus 1220 was established in two laboratories, Laboratory for low radioactivity of the Department of Physics, University of Novi Sad, Serbia (UNS) and Laboratory for measurements of low-level radioactivities at the Ruđer Bošković Institute, Zagreb, Croatia (RBI). Optimization of the method comprises choice of an appropriate vial type, selection of optimal spectral window, determination of the counting efficiency in various vials, evaluation of the influence of vial on efficiency and background.

The efficiency of the measurement of $A(^{90}\mathrm{Sr})$ in plastic vials is 45.6 % and 37.3 % at UNS and RBI, respectively, while in glass vials it is lower (~32 %). Background count rate is lower in plastic than in glass vials, resulting in better counting properties of the plastic vials. The achieved minimum detectable activity for a total counting time of 300 min was 0.32 $\mathrm{Bq \cdot L^{-1}}$ in plastic vials at UNS and 0.34 $\mathrm{Bq\ L^{-1}}$ at RBI.

Validation of the method was performed by determination of strontium level in spiked samples within interlaboratory comparison measurements. The $A(^{90}\mathrm{Sr})$ values determined in both laboratories did not deviate significantly from the spiked activities, giving the *z*-scores in the interval [-2,2]. Only one sample at RBI had the *z*-score higher than 2 but lower than 3. All results from UNS had positive *z*-score values, including the sample measured within the IAEA proficiency test.

The ^{90}Sr/^{90}Y Cherenkov radiation measurements of water samples can be successfully used for rapid screening of samples without any pretreatment of the samples.