



# Preparation of water samples for proficiency testing on radionuclides



H. Polkowska-Motrenko\*, L. Fuks, P. Kalbarczyk, J. Dudek, K. Kulisa, A. Oszczak, M. Zuba

Institute of Nuclear Chemistry and Technology, Dorodna 16 street, 03-195 Warsaw, Poland

## HIGHLIGHTS

- By participating in PTs the radiochemical laboratories demonstrate their competence.
- Test materials should be homogeneous, stable, provide proper values.
- Water used as test material should contain low self-activity and proper salt content.
- The procedure allows to obtain homogeneous, stable materials with a desired activity.

## ARTICLE INFO

### Article history:

Received 24 November 2014  
 Received in revised form  
 27 April 2015  
 Accepted 28 May 2015  
 Available online 29 May 2015

### Keywords:

Proficiency tests  
 Radionuclides  
 Water

## ABSTRACT

The Institute of Nuclear Chemistry and Technology (INCT) procedure for preparation of water samples for proficiency testing on determination of  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$ ,  $^{239}\text{Pu}$ ,  $^{226}\text{Ra}$  and  $^{90}\text{Sr}$  in water is presented. Natural waters were applied as the raw materials and spiked with the aforementioned radionuclides. The procedure of spiking water allows to prevent losses of the radionuclides and to assign property values from formulation. The main advantages of this procedure are (i) similarity of the test materials to the water samples routinely analysed by radioanalytical laboratories, and (ii) traceable assigned values with low uncertainties.

© 2015 Elsevier Ltd. All rights reserved.

## 1. Introduction

Participation of laboratories in proficiency tests (PT) provides a unique opportunity of external and objective demonstration of their performance. It is also a way to prove the laboratories' ability to conduct specific measurements or tests and generate reliable results. From this point of view, proficiency test materials should be similar as much as possible in type of matrix and content of analytes to the real materials analysed by the laboratories. A determination of radionuclide contents in different types of water is required in the Regulation of Polish Government ([Decree of Police Cabinet, 2002](#)) by radiochemical laboratories forming radiation monitoring network in Poland. On the request of the National Atomic Energy Agency (PAA), Institute of Nuclear Chemistry and Technology (INCT) has conducted PTs on determination of  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$ ,  $^{239}\text{Pu}$ ,  $^{226}\text{Ra}$  and  $^{90}\text{Sr}$  in different materials since 2004. One of them was water. The PTs have been provided annually. They met requirements of the international standards ([ISO/IEC Standard 17043, 2010](#); [Thompson et al. 2006](#); [ISO 13528, 2005](#)). The INCT uses as a rule natural matrix materials as the PT materials. Water

samples were prepared from the natural water (tap or well) by spiking them with selected radionuclides. To this aim, certified standard solutions were used in this study. This paper describes a developed procedure for the preparation of spiked water samples.

## 2. Experimental

### 2.1. Materials and methods

Three types of water have been used to prepare the test materials: drinking water, ground water and laboratory water. Drinking water was taken from the public water supply in Warsaw. Ground water was taken from artesian wells with Oligocene water in Warsaw. Laboratory water  $18\text{ M}\Omega\text{ cm}$  was obtained from Milli-ORG Millipore Co. purification system.

Certified standard solutions of  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$ ,  $^{239}\text{Pu}$ ,  $^{226}\text{Ra}$  and  $^{90}\text{Sr}$  were supplied by Amersham. Certified solutions of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  contained nonradioactive carriers i.e. Cs and Sr+Y, for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , respectively. Certified solutions of  $^{241}\text{Am}$ ,  $^{239}\text{Pu}$  and  $^{226}\text{Ra}$  were carrier free. The test materials were prepared by spiking the raw water with a certified standard solution of only one radionuclide from those mentioned above. The raw water was analysed radiometrically. Activity of  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$  was measured by gamma-

\* Corresponding author.

E-mail address: [h.polkowska@ichtj.waw.pl](mailto:h.polkowska@ichtj.waw.pl) (H. Polkowska-Motrenko).

ray-spectrometry and those of  $^{239}\text{Pu}$ ,  $^{226}\text{Ra}$  and  $^{90}\text{Sr}$  by liquid scintillation counting after radiochemical separation of radionuclide of interest (Fuks et al., 2006; Polkowska-Motrenko et al., 2011). Only water with appropriate low initial activity concentration of radionuclides in question was used as the raw materials. Certified standard solutions were added in the amounts covering a concentration range of radionuclides prevailing in drinking and ground waters. All preparation steps were done gravimetrically. The analytical balances (Sartorius, Radwag) calibrated with a calibrated weight set traceable to national standards were used to prepare the spiked waters. Before spiking, raw natural water was analysed for content of alkali and alkaline earth ions as well as transition metals and common anions ( $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ) by ion chromatography. When ions ( $\text{Fe}^{+3}$ ,  $\text{SO}_4^{2-}$ ) concentrations were too high, laboratory water was added to avoid precipitation and the pH was adjusted to 1 with concentrated  $\text{HNO}_3$ . To avoid radionuclide losses nonradioactive carriers were added to the water before spiking. Homogeneity was obtained by mixing the spiked water by bubbling with gaseous nitrogen and then the water was distributed into bottles made of polyethylene terephthalate (PET).

### 3. Results and discussion

A participation in PTs plays an important role in quality assurance/quality control (QA/QC) system of a laboratory and an increasing demand for such activities is observed (Albano et al., 2014). A participation in PTs allows the laboratory to demonstrate the laboratory's competence for the specific measurements. It also allows to provide confidence to the results supplied by the laboratory to the users of its services (ISO/IEC Standard 17043, 2010). To achieve the above goals, test materials must meet high demands. First of all test materials should be homogeneous, stable and provide property values traceable to the SI units (Albano et al., 2014). False in homogeneity or/and stability may affect the confidence in a PT. However, homogeneity and stability tests are very rarely reported by the PT providers (Albano et al., 2014).

PT samples should match the matrix of samples routinely analysed by the laboratory. So, when one determines radionuclides in waters, the composition of analysed test water should be similar to routine samples. Also activity concentration should cover ranges prevailing in such samples. However, several PT providers use synthetic water prepared by dilution of radionuclide standard solutions with distilled water (Brookman, 2008; Merssova et al., 2010; Pentelic et al., 2010; Shakhshiro et al., 2008) as the test materials. Such water can differ from the natural one and the laboratory has no evidence regarding reliability of the results of the routinely analysed samples. The INCT has conducted PTs on the determination of radionuclides in drinking and ground waters containing  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$ ,  $^{239}\text{Pu}$ ,  $^{226}\text{Ra}$  and  $^{90}\text{Sr}$  (Fuks et al., 2006; Polkowska-Motrenko and Fuks, 2010). The concentration of radionuclides covered ranges prevailing in such waters (NAEA (PAA) President's Reports). To establish a preparation method of the test samples, a design and feasibility study was undertaken. The quality of spiked natural water depends to a large extent on the proper characterisation of the raw water used for spiking. The activity concentration of radionuclide of interest in raw water (blank) should be much lower than that in spiked test (NAEA (PAA) President's Reports). The first step of a PT sample preparation was collection of natural waters. Then the determination of radionuclides was conducted and the appropriate waters were chosen. The activity concentrations of radionuclides in raw and spiked waters are shown in Tables 1 and 2 respectively. As it can be seen from Tables 1 and 2, the blank values are negligible comparing to the activity concentration values in the test samples.

**Table 1**  
Activity concentration of  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$ ,  $^{239}\text{Pu}$ ,  $^{226}\text{Ra}$  and  $^{90}\text{Sr}$  in raw and spiked drinking water.

Radionuclide	Activity concentration, $\text{Bq dm}^{-3}$	
	Raw water	Spiked water
$^{241}\text{Am}$	$\leq 0.1$	0.3–2
$^{137}\text{Cs}$	$\leq 0.015$	0.05–1.5
$^{239}\text{Pu}$	$\leq 0.05$	0.15–2
$^{226}\text{Ra}$	$\leq 0.0025$	0.05–0.5
$^{90}\text{Sr}$	$\leq 0.010$	0.03–1

The concentration of the radionuclides in the test waters (Tables 1 and 2) covers the whole range found in the routine analysis of the samples of drinking and ground water (NAEA (PAA) President's Reports).

It is important that the test samples contain dissolved solid in the amounts prevailing in real water matrix. However, the content of salts should be sufficient to avoid inhomogeneity and losses of analytes due to precipitation. This is especially important in the case of such salts as  $\text{RaSO}_4$  for which the solubility product is very low ( $\text{pK}=10.43$ ). Prior to spiking, the water was analysed to determine the level of ions and when necessary diluted with the laboratory water in order to decrease the ions content and acidified to the  $\text{pH}=1$  with  $\text{HNO}_3$ . The typical ion concentrations in test waters are shown in Table 3.

Then the water was spiked with radionuclides. Certified standard solutions of radionuclides were used. Each test sample was prepared in one batch avoiding precipitation of solids and mixed by bubbling with gaseous nitrogen. The obtained waters were expected to be homogeneous as they were the real solutions (Shakhshiro et al., 2007, 2008). Except precipitation, also adsorption on container walls can effect concentration of radionuclides in spiked water. To avoid losses of radionuclides by adsorption, nonradioactive carriers (appropriate salts, mainly chlorides or nitrates (Table 4)) were added. Final concentration of  $10^{-4}$ – $10^{-5}$  M was kept (Benes and Majer, 1980). It was checked that the applied carriers did not contain the measurable amounts of radionuclides determined. Generally, Ba is used as a carrier for  $^{226}\text{Ra}$  (Benes and Majer, 1980). However, in the case of natural waters containing  $\text{SO}_4^{2-}$ ,  $\text{BaSO}_4$  ( $\text{pK}_{\text{BaSO}_4} = 9.96$ ) can precipitate and Ra would be lost by co-precipitation. As it can be seen from Fig. 1, an addition of barium carrier in concentration of  $5 \times 10^{-5}$  M causes significant losses of  $^{226}\text{Ra}$ . Losses were not observed when Ca or Na were used for stabilisation of  $^{226}\text{Ra}$  solution.

The stability of spiked waters was examined by a comparison of the initial radionuclide concentration with that determined after four months (Table 5). The concentration of sulphuric ion in water was below  $60 \text{ mg dm}^{-3}$ .

As it can be seen from Table 5, the results obtained for samples of the same PT material after four months are consistent with the initial values. The two means for each radionuclide agree within

**Table 2**  
Activity concentration of  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$ ,  $^{239}\text{Pu}$ ,  $^{226}\text{Ra}$  and  $^{90}\text{Sr}$  in raw and spiked ground water.

Radionuclide	Activity concentration, $\text{Bq dm}^{-3}$	
	Raw water	Spiked water
$^{241}\text{Am}$	$\leq 0.1$	2–10
$^{137}\text{Cs}$	$\leq 0.015$	0.1–15
$^{239}\text{Pu}$	$\leq 0.05$	1.5–8.5
$^{226}\text{Ra}$	0.004	0.5–2
$^{90}\text{Sr}$	$\leq 0.010$	0.1–1

Download English Version:

<https://daneshyari.com/en/article/1878925>

Download Persian Version:

<https://daneshyari.com/article/1878925>

[Daneshyari.com](https://daneshyari.com)