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Determination of the gross activity of uranium, plutonium, americium and strontium in environmental samples using solid-state scintillation

Jiří Janda*, Jitka Tichá

University of Defence, NBC Defence Institute, Vita Nejedleho, Vyskov, CZ, Czech Republic

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ABSTRACT

Rapid determination of selected gross alpha and beta emitters in environmental matrices by solid-state scintillation technique is discussed. This method is based on sample treatment using microwave reactor and direct measurement of digested products using powder scintillator and alkaline solution as a substitute for traditional liquid scintillation cocktail. The selected group of radionuclides was chosen with respect to their use in nuclear industry, high radiotoxicity, and the possibility of potential misuse. The work aimed at verifying the connection of microwave decomposition using alkaline solution with solid-state scintillation using a powder scintillator YAP:Ce together with an alkaline medium.

1. Introduction

Monitoring artificial radioactivity in the environment is of utmost importance in order to verify that there is compliance with the regulatory standards, such as Basic Safety Standards (BSS) (Euratom/96/29 currently being revised) and to monitor tendencies over time.

The monitoring system generally consist of monitoring of air pollution using high-volume air samplers, such as JL-150 Hunter, or JL-900 Senya (National Radiation Protection Institute, Czechia), ambient dose/dose rate using autonomic stations placed according to the scheme over the territory, survey (on foot, by car, using different types of aircrafts), personal dosimetry, water monitoring and sampling. The last task is very broad and involves many techniques and type of the samples, which are based on the advanced level of the country and implemented procedures. Usually, the layout for the variety of the samples originates from Safety Guide No. RS-G-1.8 (IAEA Safety Standards, Environmental and Source Monitoring for Purposes of Radiation Protection) and it should reflect current conditions, such as landscape, weather, climate and food habits. All these conditions result in properly chosen samples, which often use biomarkers for their specific accumulation of artificial radionuclides in the ambient environment. In central and northern part of Europe, but also in other parts of the world, the mosses and lichens are stated as the biomarker etalon for determination of pollution (Sujetovienė and Galinytė, 2016; Gerdol et al., 2014; Kim et al., 2018; Oishi, 2018), especially heavy elements including actinides, such as uranium, plutonium and americium (Wei et al., 2009; Dudev and Lim, 2014; Paatero et al., 1998; Loppi et al., 2003; Di Lella et al., 2003; Mietelski et al., 2000; Rosamilia et al., 2004; Jia et al., 1997). As a traditional aquatic sample, algae, fish and shrimps (Desideri et al., 2003; Lusa et al., 2009; Hashimoto et al., 2002; Ikäheimonen et al., 1997; Baxter et al., 1995; Sanchez-Cabeza and Molero, 2000; Ryan et al., 1999) are often used as a representative marker of pollution in water bodies. Finally yet importantly, due to the enormous consumption of dairy products throughout the world, the presence of radionuclides in milk would result in huge contamination of population, therefore, the milk sampling is off great importance (Green, 1993; Oliveira and Carvalho, 2006; Taddei and Silva, 2006).

In peacetime, or in the situation, where time is not an essence, there are numerous procedures and techniques based on drying, leaching, ashing, robust and demanding radiochemistry subsequently followed by measurement techniques. They allow obtaining very precise results in the activity ranging from mBq per kilogram of the sample, such as Inductively coupled plasma mass spectrometry (ICP-MS), Inductively coupled plasma atomic emission spectroscopy (ICP-OES), alpha spectrometry, or Photon-Electron Rejecting Alpha Liquid Scintillation (PERALS) (Ayranov et al., 2009). These techniques have one in common and that is they are very expensive and time/consuming, which, in the case of emergency situation, is against all requirements for emergency monitoring. There are employed rapid methods, cheap and available instruments, sample throughput, easy to operate approaches and robust field methods. The downside is the inaccuracy in determination of mainly the gross activity.

As was stated in (Janda et al., 2016; Janda, 2017) it is possible to use fine yttrium aluminum perovskite activated by cerium (YAP:Ce) powder instead of traditional liquid scintillation cocktail in liquid scintillation counting. This paper deals with the rapid determination of

* Corresponding author.
E-mail address: jiri.janda@unob.cz (J. Janda).

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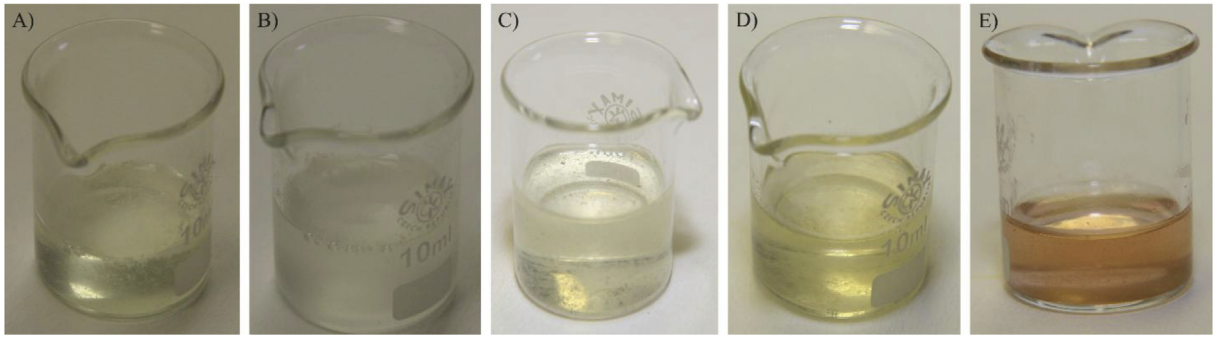

Fig. 1. Digestion products of the selected environmental samples; A) Fish fillet and shrimps, B) Milk, C) Mosses, D) Algae and E) Lichen.

the selected environmental samples, such as algae, fish, shrimps, mosses, lichen and milk using microwave digestion and solid-state scintillation technique, as a possible approach in emergency monitoring.

2. Experimental

2.1. Devices and equipment

The decomposition of samples was performed by Multiwave ECO (Anton Paar GmbH) with an inner carrousel consisting of sixteen positions for pressure vessels HVT50 with 20 bar opening pressure. The measurements were carried out using Liquid Scintillation Counter Triathler (Hidex, Finland) with 20 ml polyethylene vials. The setting of the instrument was as follows: the "RN222" counting mode was modified so that the value of the pulse length index (PLI) was set to one, time of the measurement was set to 300 s, alpha and beta window was set to full width (1–1023 channel), the YGain was 167 and YPos was 48. Proper shaking was performed by shaker IKA KS 130 basic (IKA, USA) and centrifuge MPW-340 (MPW, PL) was used for accelerating of sedimentation.

2.2. Chemicals

The scintillation powder YAP:Ce (0–30 μm grain size, the Ce^{3+} dopant concentration of 0.6%) was used as a scintillation media. Radiochemical isotope tracers U_{NAT} (0.1 $Bq.\mu l^{-1}$), Pu-239 (1.1 $Bq.\mu l^{-1}$), Am-241 (1.67 $Bq.\mu l^{-1}$) and Sr-90 (0.654 $Bq.\mu l^{-1}$) were obtained from AEA Technology, UK, QSA Amersham International and CMI (the Czech Metrological Institute). U_{NAT} tracer was prepared to be self-cleaning, removing its ^{228}Th daughter using ion exchange resin BIORAD AG 1-X8 (400–800 mesh). The measuring solution contained 0.1 mol l^{-1} HNO_3.

Digestion solution consisted of sodium hydroxide and hydrogen peroxide (Lach-ner, Inc., CZ). Water was obtained from a DEMIWA 5 ROSA™ (Watek) water purification system.

The investigated samples were mosses and lichen, which were collected in the Moravian countryside, as well as algae. Milk, fish (cod fillet) and shrimps were bought from local distributors.

2.3. Experimental procedures

2.3.1. Sample preparation

The technique used for digestion of samples was as follows. A sample of known mass was placed in the digestion vessel and 50 μL of only one radionuclide solution of desired activity was added. The entire procedure was repeated for all investigated radionuclides except for U_{NAT}, where 200 μL was added resulting in four digestion vials containing the same sample but different radionuclide. They were then put into a hot air oven and slightly dried out. This step was omitted in case of milk. Afterward, all samples were treated with an appropriate mixture of sodium hydroxide and hydrogen peroxide (see "Digestion of samples" step). Vessels were kept in a fume hood until exothermal reaction occurred, and then all vessels were collocated in the Multiwave ECO. The appropriate digestion program was chosen for decomposition. All programs were adjusted to ensure the highest decomposition and measurement efficiency.

2.3.2. Digestion of cod fillets and shrimps

The 0.5 g of sample (cod fillet, shrimps or mixture) was ground into smaller pieces and put into the vessel. Afterward, 5 ml of H_2O_2 and 5 ml of 3 M NaOH were added. The digestion program was following:

T [°C]	Ramp Time [min]	T of digestion [min]	Power [W]
100	6	10	30
160	2	5	65
180	2	8	75
50	1	10	0

2.3.3. Digestion of milk

The 1 ml of milk was transferred into the vessel followed by 2 ml of H_2O_2 and 9 ml of 7 M NaOH. The digestion program was following:

T [°C]	Ramp Time [min]	T of digestion [min]	Power [W]
100	6	10	30
160	2	10	65
180	2	12	75
50	1	10	0

2.3.4. Digestion of mosses

The 0.05 g of dried mosses or 0.2 g of fresh mosses was catted into small pieces and transferred into the vessel. Then 5 ml of H_2O_2 and 5 ml of 7 M NaOH were added. The digestion was realized in the same way as milk.

2.3.5. Digestion of algae

The 0.1 g of dried algae was ground and then transferred into the vessel followed by 5 ml of H_2O_2 and 5 ml of 7 M NaOH. The digestion program was following:

T [°C]	Ramp Time [min]	T of digestion [min]	Power [W]
100	6	10	30
170	2	12	65
185	2	15	75
50	1	10	0