

**INTERNATIONAL COMMITTEE
FOR RADIONUCLIDE METROLOGY**

Proceedings part II of the

**21st International Conference on
Radionuclide Metrology and its
Applications (ICRM 2017)**

15-19 May 2017, Buenos Aires, Argentina

Editors

Pablo Arenillas, Dirk Arnold and
Uwe Wätjen



The International Committee for Radionuclide Metrology (ICRM) is an association of radionuclide metrology laboratories whose membership is composed of delegates of these laboratories together with other scientists (associate members) actively engaged in the study and applications of radioactivity.

It explicitly aims at being an international forum for the dissemination of information on techniques, applications and data in the field of radionuclide metrology. This discipline provides a range of tools for tackling a wide variety of problems in numerous other fields, for both basic research and industrial applications. Radionuclide metrology continues to play an important role in the nuclear industry, supporting activities such as radionuclide production, nuclear medicine, measurement of environmental radioactivity and of radionuclides in food and drinking water, decommissioning of nuclear facilities, nuclear security and emergency preparedness, nuclear physics research, etc.

International Committee for Radionuclide Metrology (ICRM)

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PREFACE

This first issue of the ICRM Technical Series on Radionuclide Metrology is devoted to the 21st International Conference on Radionuclide Metrology and its Applications (ICRM 2017), which was held from 15 - 19 May 2017 at the Panamericano Hotel, Buenos Aires, Argentina. The conference was hosted by the Argentinian *Comisión Nacional de Energía Atómica* (CNEA), the local organisation was undertaken by a team comprising staff of the CNEA Ezeiza Atomic Center and the CNEA headquarters. The *Instituto Nacional de Tecnología Industrial* (INTI) of Argentina cooperated in hosting the conference. The city of Buenos Aires with its rich culture, often called the “Paris of South America” for its architecture and European heritage offered a pleasant conference setting with uncountable possibilities for relaxation in the evening, stimulating discussions about present and future collaborations and giving plenty of opportunities to meet old and make new friends.

Plenary meetings of the International Committee for Radionuclide Metrology (ICRM) are held biennially and have developed into scientific ICRM conferences, a successful instrument of communication among various specialists, truly encouraging international cooperation, also across borders of their disciplines. This series of meetings dates back to the 1st International Summer School on Radionuclide Metrology which was held in 1972 at Herceg Novi, Yugoslavia. After two previous meetings on radionuclide metrology, organised by the International Atomic Energy Agency (IAEA) in 1959 and 1966, contacts among radionuclide metrologists from several countries had grown stronger at the Herceg Novi summer school bringing forth the idea of a permanent committee to promote applications of radionuclide metrology. ICRM was founded at Paris in 1974, subsequently inviting more laboratories to join, and by 1976 ICRM counted 15 members. More than 40 years later, ICRM has grown to be a truly worldwide association with, at present, 47 member institutions.

The International Committee for Radionuclide Metrology (ICRM) is an association of radionuclide metrology laboratories whose membership is composed of appointed delegates of these laboratories together with other scientists (associate members) actively engaged in the study and applications of radioactivity. It explicitly aims at being an international forum for the dissemination of information on techniques, applications and data in the field of radionuclide metrology. This discipline provides a range of tools for tackling a wide variety of problems in numerous other fields, for both basic research and industrial applications. Radionuclide metrology continues to play an important role in the nuclear industry, supporting activities such as radionuclide production, nuclear medicine, measurement of environmental radioactivity and of radionuclides in food and drinking water, decommissioning of nuclear facilities, nuclear security and emergency preparedness, and nuclear physics research. Papers presented at the ICRM 2017 conference covered the metrological aspects pertaining to many of these fields.

The 21st International Conference on Radionuclide Metrology and its Applications (ICRM 2017) was attended by 158 participants from 31 countries worldwide and 4 international organisations plus 15 registered company exhibitors. Of the 151 submitted abstracts, the scientific programme committee had selected 134 contributions for presentation, either as oral (38) or as poster (96). Finally, during the four very busy

symposium days from 15 to 18 May, 2 invited talks, 34 oral contributions and 86 posters were presented.



Fig. 1. Conference opening ceremony: (a) Dirk Arnold, ICRM President, Pablo Arenillas, Scientific Secretary of ICRM 2017, and Osvaldo Calzetta Larrieu, CNEA President; (b) Héctor Laiz, invited speaker of INTI; (c) the audience.

Participants were welcomed during the conference opening by Lic. Osvaldo Calzetta Larrieu, President of the CNEA, and by the ICRM President Dr Dirk Arnold. The scientific programme of the conference opened with two invited talks. Dr Héctor Laiz of the *Instituto Nacional de Tecnología Industrial* of Argentina (one of the 17 founding countries of the Metre Convention) presented an overview of international metrology, in particular the Inter-American Metrology System, SIM. He also reviewed the impact of the revision of the International System of Units (SI), envisaged to match the increasing demands on measurement by updating the definitions of four of the SI base units solely based on physical constants and expected to come into force in 2019. Dr Lisa Karam (NIST) summarized the efforts of radionuclide metrology laboratories within NMIs and Designated Institutes (DIs) of the SIM, often supported by the IAEA, to improve realization and subsequent distribution of radioactivity standards in their countries or regions. The scientific programme continued with a session on Aspects of International Metrology, followed by all other sessions covering the fields in which ICRM is active during the four days of the scientific conference.

Whereas much of the presented work will be published in a special issue of Applied Radiation and Isotopes⁺, three papers are published in this first issue of the ICRM Technical Series on Radionuclide Metrology. They were only accepted after a very rigorous reviewing process to standards comparable to the review of papers for Applied Radiation and Isotopes. We expect that, with future ICRM conferences, more papers will be published in this Technical Series, due to the increasing constraints on the number of papers to be selected for a proceedings special issue of any journal, based on their novelty and completeness of the presented work. Thus, in the future, the conference proceedings will become more evenly distributed between a journal special issue and the ICRM Technical Series with this first issue turning out to be the pilot issue.

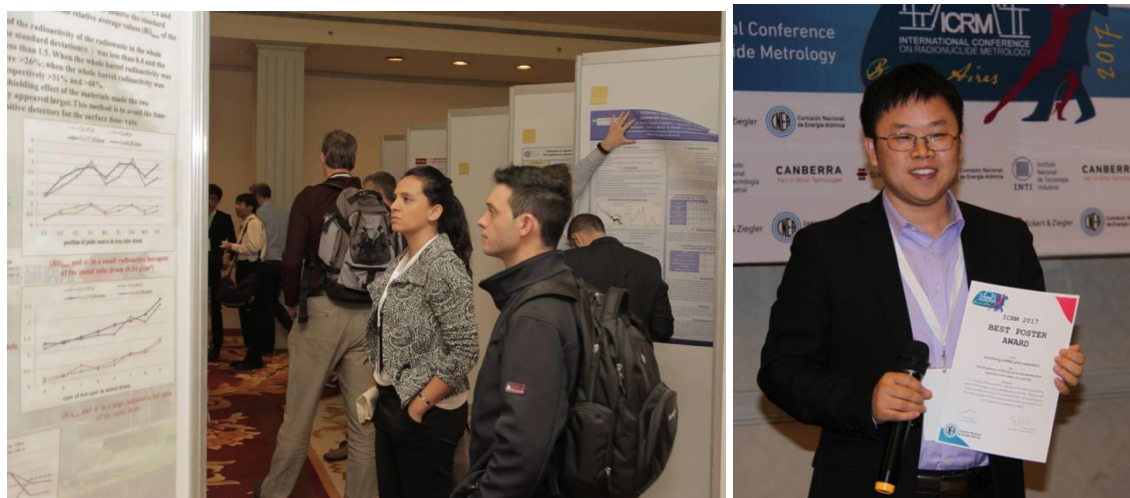


Fig. 2. Poster presentations: (a) exhibiton hall; (b) Juncheng Liang, NIM, China, award winner for the best poster.

Business meetings of all ICRM Working Groups were integrated into the conference programme. An exhibition of scientific equipment, literature and services, exposed by 10 companies, was held during all four symposium days, combined with the scientific poster presentations. The scientific conference was followed by an ICRM Executive Board meeting. The final day was devoted to the General Meeting of the ICRM.

The social programme encompassed an informal get-together on Sunday, a welcome reception on Monday, and an introduction to the Argentinian Tango (Tango class), followed by a Tango show conference dinner.

Our appreciation and thanks go to all who contributed to this very successful and busy meeting. In particular we recognize the contributions made by Pablo Arenillas and his Local Organising Committee, comprising members of the Radionuclide Metrology Laboratory and Public Relations group. Many thanks are also addressed to the Scientific Programme Committee, the ICRM Executive Board, the ICRM Working Group coordinators, the referees and session chairs, and to the authors of papers. We are particularly thankful to the coordinating referees for the efficient organisation of the review process for their conference sessions.

⁺ Applied Radiation and Isotopes vol. 133 (2018)



Fig. 3. ICRM 2017 conference dinner and Tango show.

We are indebted for the support received from the hosting institutions - Comisión Nacional de Energía Atómica (CNEA) and Instituto Nacional de Tecnología Industrial (INTI). We are also thankful for the financial support received from the exhibitors and all other sponsors named below.

There are 47 institutions now represented by delegates in the ICRM. The ICRM has no membership fee and no paid secretariat or other staff. Its overall direction is determined by the delegates at General Meetings, which convene usually every two years, during which organisational guidelines and directions for the work programmes are agreed upon. The following officers of ICRM were elected by the delegate members at the General Meeting of 19 May 2017.

President	Eduardo García-Toraño ¹
Vice-Presidents	John Keightley ² Mikael Hult ³ Akira Yunoki ⁴
Past President	Dirk Arnold ⁵
Secretary	Franz Josef Maringer ⁶

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We express our thanks to Pierino De Felice⁷, Uwe Wätjen⁸ and Tae Soon Park⁹ for their contributions as Past President, Secretary and Vice-President, respectively, during their terms.

ICRM activities are largely the responsibility of its working groups. Each group is guided by a coordinator who acts as a centre for ideas and communications and may organise conferences and workshops. There are now eight working groups with the following fields of interest and assigned coordinators:

- (1) Radionuclide Metrology Techniques: John Keightley², Mike Unterweger¹⁰, Christophe Bobin¹¹, Pierino De Felice⁷
- (2) Alpha-Particle Spectrometry: Stefaan Pommé³
- (3) Gamma-Ray Spectrometry: Marie-Christine Lépy¹¹
- (4) Life Sciences: Jeffrey T. Cessna¹⁰
- (5) Liquid Scintillation Counting: Karsten Kossert⁵
- (6) Low-Level Measurement Techniques: Mikael Hult³
- (7) Beta-Particle Spectrometry: Xavier Mougeot¹¹
- (8) Nuclear Decay Data: Mark Kellett¹¹

At this point, we want to thank Octavian Sima¹² for his contributions to the activities of the gamma-ray spectrometry working group as their coordinator from 2011 until now.

Anyone wishing to participate in ICRM's activities or to receive further information is encouraged to contact one of the officers above. Details of ICRM and its Working Groups are available from the ICRM web site at <http://physics.nist.gov/icrm>. An ICRM Newsletter reporting activities in the member laboratories is issued biennially. The most recent, Issue 30 and several past annual Newsletters may be viewed at <http://www.lnhb.fr/seminaires-et-publications/icrm-newsletter/>.

The next biennial ICRM conference (ICRM 2019) will be held from 27 to 31 May 2019 in Salamanca, Spain, organised by the University of Salamanca. The contact person of the local organising committee and Scientific Secretary of the conference is Dr Begoña Quintana Arnés (quintana@usal.es). The meetings will be open to all interested persons. They will follow guidelines similar to those of ICRM 2017 comprising a general conference on radionuclide metrology and its applications that will cover the fields of aspects of international metrology, intercomparisons, measurement standards and reference materials, radionuclide metrology techniques, alpha-particle and beta-particle spectrometry, gamma-ray spectrometry, liquid scintillation counting techniques, nuclear decay data, low-level radioactivity measurement techniques, radionuclide metrology in

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life sciences, source preparation techniques, and quality assurance and uncertainty evaluation in radioactivity measurements, together with working group meetings that will offer a less formal frame for scientific discussions. We are wishing the organisers and participants of the 22nd edition of the ICRM conference in Salamanca as much success, joy and excitement as we had with the ICRM 2017 conference.



Fig. 4. ICRM 2017 conference photo.

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Low-level direct measurement of ^{238}U in environmental water using state-of-the-art gamma-ray spectrometry

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Abstract

Detection and measurement of uranium in environmental water is of primary importance for site monitoring in normal and incidental situation. Current recommended analytical methods for measurement of this parameter involve fluorimetry, mass spectrometry or possibly alpha spectrometry. As a proof of concept, taking benefit of the Gamma³ spectrometer large detection efficiency and very low background, detection limits for ^{238}U in environmental water samples have been investigated. Values as low as, 44 mBq/L (*i.e.* $\sim 3.6 \mu\text{g/L}$ of ^{238}U) have been obtained.

Keywords: ^{238}U , Gamma-ray spectrometry, Environment, Water

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1. Introduction

Detection and measurement of uranium in environmental water is of primary importance for site monitoring in normal and incidental situation. Current recommended analytical methods for measurement of this parameter involve fluorimetry, mass spectrometry or possibly alpha spectrometry. Detection limits ranging from $\sim 1 \text{ ng/L}$ to $\sim 10 \mu\text{g/L}$ are achieved with these techniques at the cost of time-consuming radiochemical procedures and/or large financial investments.

Even though implementation of direct gamma-ray spectrometry would be more efficient for these latest aspects, until now this technique has not demonstrated sufficient sensitivity. Hampered by the low γ -ray emission intensities of ^{238}U , minimum detectable concentrations as large as $0.6\text{--}1.2 \text{ Bq/L}$ (*i.e.* $\sim 48\text{--}96 \mu\text{g/L}$ for ^{238}U) are typically encountered in routine operation of standard gamma-ray spectrometers.

A state-of-art gamma-ray spectrometry facility has been designed at our surface-level laboratory. Taking benefit of the combined large detection efficiency and optimized background - as a proof of concept - performances of this setup for detection of ^{238}U in environmental water have been investigated. Performances are compared to the ones obtained using a “standard” coaxial High Purity Germanium (HPGe) spectrometer.

2. Method and material

2.1 Minimum detectable concentration

From Currie (1968), the Minimal Detectable Concentration (*MDC*) of a radionuclide using γ -ray spectrometry relies on the following equation:

$$MDC [\text{Bq.l}^{-1}] = \frac{2.71 + 4.65\sqrt{\mu_B}}{t \times V \times I_g \times \varepsilon} \times K$$

Where, t denotes the acquisition time [s], V stands for the sample volume [L], I_g is the emission intensity of the considered transition for the radionuclide of interest, ε denotes the detection efficiency at the considered energy. μ_B corresponds to the variance of the blank-subtracted signal at the energy of interest (the integral is performed over $\pm 1.25\text{FWHM}$ (Full-Width at Half-Maximum) on either side of the hypothetical peak centroid). Finally, various corrections are summarized in the factor K . This equation shows that four ways are available to optimize *MDC*: i)

increase of acquisition time (for long-lived nuclides), ii) increase of sample volume, iii) increase of detection efficiency, iv) background reduction (and possibly resolution optimization). However, most of these parameters are not independent. For instance, detection efficiency and background are to a great extent positively correlated, whereas detection efficiency and sample volume exhibit a negative correlation.

2.2 The conventional γ -spectrometer

For comparison of performances, a conventional low background coaxial n-type HPGe spectrometer (Ortec GMX40-76-LB) has been used. According to ANSI/Std325 (1996) its efficiency is 40%. It is using a passive lead shield (15 cm thick) and analog electronics. The water sample is measured using a standardized SG500 (500 mL) polyethylene cylindrical bottle.

2.3 The Gamma³ system

The Gamma³ system has been designed with the objective of *MDC* optimization. The setup consists in a triple crystal arrangement (Canberra BEGe5030P) enclosed in an optimized passive and active shielding. This instrument has been extensively described in Cagniant *et al.* (2015, 2017); the measurement configuration system is shown on Fig. 1.



Fig. 1: Gamma³ measurement setup (1 L vial).

Multi-channel digital electronics are used for data acquisition. Three of the four channels are used to acquire individual spectrum from each spectrometer while the last one is used for the cosmic veto (plastic scintillator). List-mode spectra are treated off-line by dedicated software producing a single muon-vetoed gamma-ray spectrum. Thus the whole system is operating as a virtual single high-efficiency γ -ray spectrometer.

It is obvious that the cylindrical geometry is not the most efficient one for the triple crystal configuration of the Gamma³ setup. Indeed the two horizontal spectrometers would not be in close contact of a cylindrical container. Taking benefit of versatility of the Gamma³ setup, a 1 L polyethylene rectangular shaped plastic bottle has been preferred (Fig. 1). This configuration is the best compromise between detection efficiency (ϵ) and sample volume (V) (Jutier and Douysset, 2012).

2.4 Calibration

Due to their respective half-lives, ^{238}U (4.5×10^9 y) and its first daughter ^{234}Th (24.1 d) are considered in secular equilibrium in environmental soil samples. In ground-water, because of differences in solubility of uranium and thorium, this equilibrium might be perturbed for fresh samples (Korun and Kovačič, 2011). For the present study, sample age is large enough to assume that the two radionuclides are in equilibrium. Therefore, ^{238}U is quantified from the 63.3 keV γ -line of ^{234}Th . Nuclear data are taken from the DDEP on-line database (www.nucleide.org).

The two γ -ray spectrometry systems have been calibrated in terms of full-energy peak efficiency (simplified as “detection efficiency” in text).

A resin matrix (density = 1.13) containing a certified mixture of gamma-ray emitters has been used. The geometry and material of the calibration vial is identical to the one of the measurement container. An empirical efficiency curve has been obtained for each system covering the range [22-1836 keV]. A model of the Gamma³ setup has also been created using the GEANT4 Monte-Carlo code (Agostinelli *et al.*, 2003) in order to provide a simulated efficiency curve. A good agreement (discrepancy <5%) is obtained with the experimental data (Fig. 2).

True-Coincidence Summing (TCS) corrections have been implemented when needed (^{57}Co , ^{60}Co , ^{65}Zn , ^{85}Sr , ^{88}Y , ^{113}Sn ,

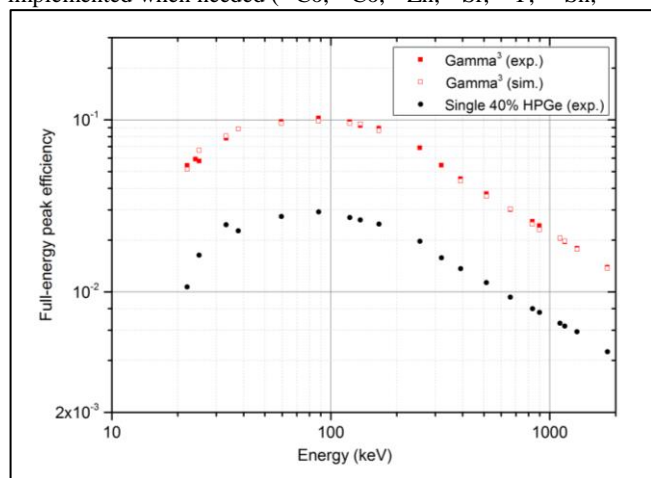


Fig. 2: Empirical and simulated full-energy peak efficiency curves of the two γ -spectrometers used in this study. The Gamma³ system is operating a 1 L rectangular bottle whereas the 40% single coaxial spectrometer is using a 500 mL cylindrical vial.

^{139}Ce) in order to establish the experimental efficiency curves. It is worth noticing that even though the detection efficiency of the Gamma³ system is very large, the net TCS corrections are rather limited. Since the three crystals are using separated acquisition channels, the net TCS correction of the whole system is similar to the one of a single crystal system and maximal correction (15%) is obtained for ^{139}Ce .

The efficiency curves of the two γ -ray spectrometry systems used for this study are shown in Fig. 2.

It appears that in the range of interest [60-100 keV], the detection efficiency of the Gamma³ setup is larger by a factor of ~3.5 when compared to the conventional system. As shown before, *MDC* is decreasing as the product of sample volume and detection efficiency. Thus, the net sensitivity gain for detection of ^{238}U (via ^{234}Th) from a single coaxial 40% spectrometer to the Gamma³ system is close to a factor of about 7.0.

3. Results

3.1 Background comparison

Thanks to the combination of optimized passive shield, efficient muon veto and Rn flushing system, the integral background signal, when a volume of ultra-pure water is placed in the measurement container, is as low as 123 counts/h in the [20-100 keV] region and 649 counts/h in the [20-2000 keV] region for the Gamma³ spectrometer. Comparison of some characteristics of the background of Gamma³ and of the conventional system are recalled in Table 1 and Fig. 3.

It should be noticed that the total mass of Ge is 2.58 kg for Gamma³ and 0.89 kg for the single 40% efficiency system.

Basically the background signal in the Region of Interest (ROI) for detection of ^{238}U via ^{234}Th is reduced by a factor of about 4.2 from the conventional spectrometry system to the Gamma³ setup even though its detection efficiency is significantly larger. It should be recalled that when measuring an actual water sample, background signal in the ROI might increase due to Compton scattering of high energy photons related to other nuclides (*e.g.* ^{40}K , ^{214}Pb , ^{214}Bi , ...).

Table 1: Background characteristics of the Gamma³ setup (3 crystals in additive mode) and a single 40% efficiency conventional low background γ -spectrometer (unit: counts/h). The two systems are operated in the same surface-level laboratory.

Energy	Gamma ³	single 40% HPGe
20-100 keV	123	516
20-2000 keV	649	2941
46 keV (Pb-210)	0.8	-
63 keV (Th-234)	0.3	-
66 keV (Ge-73m)	1.4	5.8
75 keV (Pb-Xrays)	2.9	21.8
186 keV (U-235+Ra-226)	1.1	-
352 keV (Pb-214)	2.5	4.9
511 keV (e^+ annih.)	6.4	61.7
609 keV (Bi-214)	2.3	4.1

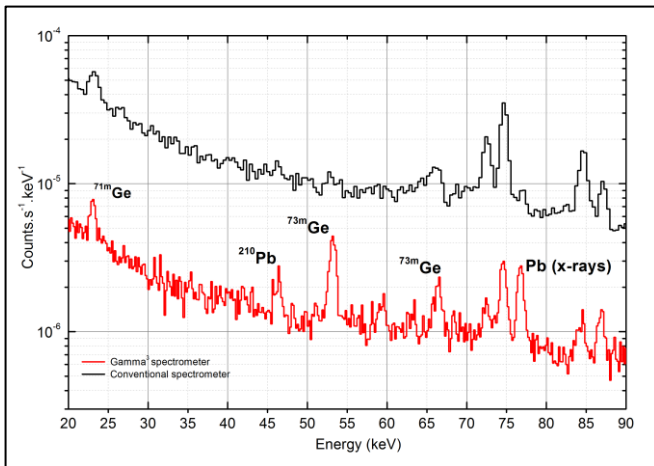


Fig. 3: Compared background spectra in the range of interest for Gamma³ (lower curve) and for a conventional 40% coaxial spectrometer (upper curve).

3.2 Detection and measurement of ²³⁸U in environmental water

Filtered (0.45 μm) environmental water samples have been measured for 7 days on both systems. No prior pre-concentration was implemented.

It appears that no ²³⁴Th (or ²³⁸U) is detected within this time when using the conventional system. *MDC* for this nuclide is estimated to 710 mBq/L (or ~57 μg/L). This value lies in the lower range of typical γ-ray spectrometer sensitivities.

Thanks to its large detection efficiency, a large sample volume and a very low background, ²³⁴Th is clearly detectable when operating the Gamma³ setup (Fig. 4). A 10.9% relative efficiency correction for 63 keV self-absorption difference between sample and calibration matrix has been estimated by Monte-Carlo simulation and implemented. Finally, the ²³⁸U activity of the sample is estimated to 26±13 mBq/L (or ~2.0±1.0 μg/L).

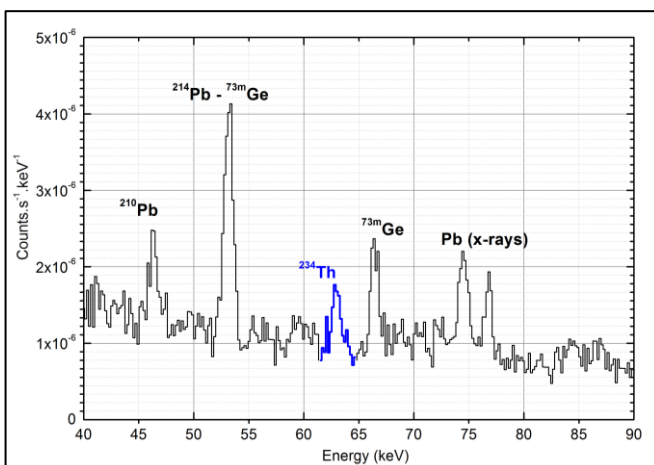


Fig. 4: Detection of ²³⁸U (via Th-234) in an environmental water sample using the Gamma³ system. Acquisition time is 7 days. ²³⁸U concentration is estimated to 26 mBq/L.

Based on the analysis the overall *MDC* for ²³⁸U is as low as 44 mBq/L (*i.e.* about 3.6 μg/L) for a 1 L sample and an acquisition time of 7 d. Compared to a conventional “off-the-shelf” system the gain in terms of sensitivity is a large as a factor of ~16.

4. Conclusions and perspectives

Taking benefit of the enhanced sensitivity and the optimized background of the Gamma³ system, detection limit for ²³⁸U as low as 44 mBq/L (*i.e.* 3.6 μg/L for ²³⁸U) is obtained. This value is larger than the required detection limit (20 mBq/L) stated in Euratom Directive (2013) and close to one-tenth of the latest WHO’s (World Health Organization, 2011) provisional guideline value for maximum uranium concentration in drinking water (30 μg/L).

Therefore γ-ray spectrometry, even if not as sensitive as some other techniques, might be competitive for simple assessment of uranium content of water samples when using an advanced detection system. Any sample of seawater, groundwater or drinking water might be analyzed simply as received without any preparation. The technique also offers the advantage to monitor in the same run some other relevant nuclides such as ²²⁶Ra/²²⁸Ra, ²²²Rn or ²¹⁰Pb.

Future developments will concentrate on mitigation of ²³⁴Th signal in the Gamma³ background aiming at pushing forward the detection sensitivity of the setup for detection of uranium.

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Use of the TDCR method for standardization of radionuclides in the FTMC

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Abstract

The TDCR method was implemented in the FTMC, the National Metrology Institute in Lithuania. Seven beta-emitters have been standardized with the TDCR apparatus: ^3H , ^{14}C , ^{36}Cl , ^{63}Ni , ^{90}Sr , ^{99}Tc and ^{129}I . Codes TDCRB-02, TDCR07c and TDCR2014 were used for experimental data assessment. The performance was tested within two comparisons on measurement of ^3H in tritiated-water. FTMC results deviated, in the comparison between FTMC, LNE-LNHB and VNIIM in 2013, by 0.38 % from the CCRI(II) 2009 reference value.

Keywords: TDCR; beta-emitters; standardization; double-label counting; low-level activity; quenching

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1. Introduction

In the Lithuanian National metrology institute, the Center for Physical Sciences and Technology (FTMC), the equipment that uses the Triple-to-Double Coincidence Ratio (TDCR) method for electron capture and beta-emitters standardization is based on the MAC3 electronic module (Bouchard and Cassette, 2000) and a three photomultiplier liquid scintillation counting apparatus. The radionuclide standardization equipment of FTMC was constructed in the frame of the project “Development of National Ionizing Radiation Measurement Infrastructure according to the Practice of the European Union Member states”, in 2007. More details about the Ionizing Radiation Metrology Laboratory can be found in Karam et al. (2012).

Radionuclides standardized with the TDCR method at FTMC are used in various programs and projects such as research into radionuclides transfer from a near-surface radioactive waste repository to the environment (Gudelis and Gorina, 2015) as well as applications and development of liquid scintillation counting (LSC) methods for standardization of radionuclides. Moreover, applications include decommissioning studies at the Ignalina nuclear power plant (Druteikienė et al., 2014; Druteikienė et al., 2015; Jermolajev et al., 2014; Mažeika et al., 2015; Remeikis et al., 2012) with an emphasis on high or moderate activity determination, and environmental research (Ežerinskis et al., 2016) where low-level activity measurements are predominant.

2. Standardization of radionuclides at FTMC

The first radionuclides standardized with the TDCR method were the pure beta-emitters ^{36}Cl , ^{63}Ni and ^{90}Sr . Experimental data were processed with the TDCRB-02 software (LSCWG, 2007). It was verified that the TDCR primary standardization method had a potential to measure activity of beta-emitters with high accuracy even at relatively low activity levels (Gudelis et al., 2012). Later on, radionuclides ^3H , ^{14}C , ^{99}Tc and ^{129}I were measured, this time using the TDCR07c code (LSCWG, 2007) for data analysis.

The maximum beta energies and transition probabilities of the standardized radionuclides according to the Decay Data Evaluation Project database (DDEP, 2013) and relevant to this set of studies are summarized in Table 1.

In Tables 2 and 3, the uncertainty components of the standardized radionuclides are presented.

Table 1: Nuclear data of the radionuclides standardized in this study.

Radionuclide	E_{max} (keV)	Transition probability (%)
^3H	18.591 (1)	100
^{14}C	156.476 (4)	100
^{36}Cl	709.53 (5)	98.1 (1)
^{63}Ni	66.980 (15)	100
^{90}Sr	545.9 (14)	100
^{90}Y	2278.7 (16)	99.983 (7)
^{99}Tc	293.8 (14)	99.99855 (30)
^{129}I	151.2 (11)	99.5 (5)

Table 2: Uncertainty components for standardization of ^3H , ^{14}C and ^{99}Tc ($k=1$).

Uncertainty component (in %)	^3H	^{14}C	^{99}Tc
Counting statistics	0.19	0.20	0.21
Weighing	0.05	0.05	0.05
Background	0.10	0.10	0.05
Dead-time	0.10	0.10	0.10
Decay correction	0.10	0.05	0.05
TDCR model	0.01	0.01	0.01
Sample stability	0.01	0.01	0.01
kB parameter	0.52	0.09	0.05
Ionization quenching	0.20	0.12	0.10
Quadratically combined standard uncertainty	0.62	0.30	0.27

The standardization performance was tested during two comparisons in 2013 and 2015 when tritium activity was determined.

3. Participation in comparisons

3.1 Comparisons on ^3H activity

The Ionizing Radiation Metrology Laboratory participated in a ^3H activity comparison between FTMC, LNE-LNHB and VNIIM in 2013. This allowed the FTMC values to be compared with the CCRI(II)-K2.H-3 2009 ^3H international comparison results through the established traceability chain (Cassette et al., 2016). In this comparison, experimental data were processed with the

Table 3: Uncertainty components for standardization of ^{36}Cl , ^{63}Ni , ^{90}Sr and ^{129}I ($k=1$).

Uncertainty component (%)	^{36}Cl	^{63}Ni	^{90}Sr	^{129}I
Counting statistics	0.05	0.24	0.20	0.13
Weighing	0.05	0.05	0.05	0.05
Background	0.05	0.10	0.05	0.10
Dead-time	0.10	0.10	0.10	0.10
Decay correction	0.05	0.10	0.10	0.05
TDCR model	0.01	0.01	0.01	0.20
Sample stability	0.01	0.01	0.01	0.01
kB parameter	0.05	0.50	0.10	0.12
Ionization quenching	0.08	0.15	0.10	0.12
Quadratically combined standard uncertainty	0.17	0.60	0.29	0.33

aid of the TDCR07c software. The FTMC result 29.21 kBq/g deviated by 0.38 % from the reference CCRI(II) 2009 value. This difference was meaningfully less as compared to the uncertainty of the FTMC result that was 0.68 % at a coverage factor $k=1$ (which was, on the other hand, comparable to the uncertainty of the reference CCRI(II) 2009 value 0.52 % at $k=1$).

In 2015, FTMC participated in the CCRI(II)-S12.H-3 Supplementary comparison: Comparison of methods for the calculation of the activity and standard uncertainty of a tritiated-water source measured using the LSC-TDCR method and used the code TDCR2014 (Cassette, 2014). The purpose of the latter exercise was to compare the calculation methods used at the National Metrology Institutes and estimate the dispersions of TDCR measurement results when measurement conditions are well defined by evaluating the same experimental data sets (Cassette et al., 2017). A total of 17 laboratories calculated the activity and standard uncertainty of the source, and all results were compatible.

3.2 Organization of comparisons

FTMC has organized two comparisons with the purpose of measuring tritium activity in water, VMT.RA.021 in 2013 (Gudelis and Gorina, 2016) and FTMC.RA.022 in 2015 (this work). Participants in the comparisons were various Lithuanian laboratories dealing with radionuclide activity measurements in their specific fields of application: Center for Physical Sciences and Technology (FTMC), Ignalina Nuclear Power Plant, National Food and Veterinary Risk Assessment Institute, Nature Research Centre, Radiation Protection Centre, Vilnius University (students from the Physics Faculty programs “Energetic Physics” and “Environmental and Chemical Physics”). Not all participants were part of both comparisons, some participants had reported two results from their different laboratories. Among the participants, two of them, National Food and Veterinary Risk Assessment Institute and Radiation Protection Centre, were accredited laboratories according to the ISO/IEC 17025 (ISO/IEC, 2005) standard. The purpose of these comparisons was ensuring the traceability of laboratories to the National standard of radionuclide activity. The tritium activity levels varied from (1852 ± 28) Bq/kg ($k=1$) in 2013 to (2082 ± 28) Bq/kg ($k=1$) in 2015. The results of these comparisons showed that the determination of tritium in water was not a straightforward task for all participating laboratories. The comparisons revealed problems in both calibration and uncertainty evaluation. One can see from Fig. 1 that results of participants 1, 4, 6 and 7 deviated from the assigned value considerably, indicating possible errors in calibration, while participants 1 and 4 used the counting statistics only as a measure for the uncertainty.

The next comparison of measurement of tritium in water is scheduled for Lithuanian laboratories in 2018.

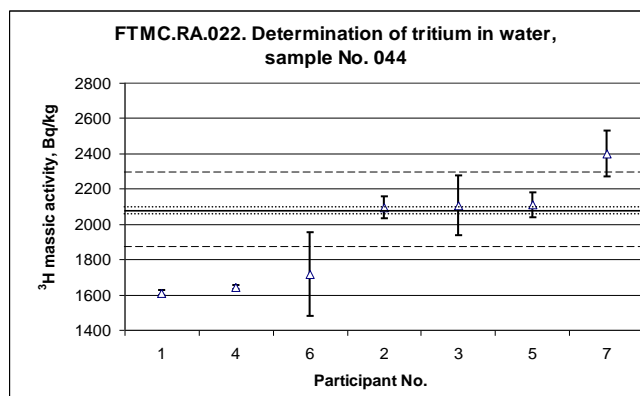


Fig. 1: Results of laboratories in the fifth comparison in 2015. The assigned value for ^3H of (2082 ± 56) Bq/kg (coverage factor $k=2$) is shown by a solid line while its uncertainty shown by dotted lines. Error bars indicate uncertainties (coverage factor $k=1$) given by participants, dashed lines show $\pm 10\%$ deviation from the assigned value.

4. Application of radionuclides standardized

4.1 Studies of radionuclide releases from radioactive waste repository

The near-surface radioactive waste repository is located in the forest about 40 km from the capital city Vilnius. The radionuclide inventory in this repository is shown in Table 4.

Table 4: Radionuclide inventory in the radioactive waste repository in February 2016.

Radionuclide	Inventory (Bq)	Half-life (a)
^3H	5.49E+13	12.312 (25)
^{137}Cs	2.99E+13	30.05 (8)
^{60}Co	2.07E+11	5.2711 (8)
^{90}Sr	3.37E+11	28.80 (7)
^{204}Tl	4.84E+10	3.788 (15)
^{239}Pu	9.14E+11	24100 (11)
^{14}C	1.76E+11	5700 (30)
^{226}Ra	1.10E+11	1600 (7)
^{152}Eu	1.34E+10	13.522 (16)
^{63}Ni	3.42E+10	98.7 (24)
^{36}Cl	1.20E+09	302 (4) E+03

Previous investigations, carried out before 2005, had confirmed an extensive leakage of tritium to groundwater. The original groundwater samples were measured with a Quantulus-1220 liquid scintillation counter. During the implementation of the Lithuanian Science Council project No. MIP-050/2011 in 2011-2012 (Nedveckaitė et al., 2013) it was found that tritium and ^{14}C can be transferred from a near-surface repository to the ground-level atmosphere, while both radionuclides were present in groundwater. This implied the need to perform calibrations using the double-label counting technique. The calibrations were carried out at different quenching levels of the LS source, as the groundwater samples taken from monitoring wells, which are installed at different distances and directions from the repository, might present various quenching levels.

The double-label counting of tritium and radiocarbon in a single sample (groundwater or air humidity) allowed omitting the

radiochemical separation procedures, shortened time necessary for ^{14}C sample preparation for analysis by LSC and therefore was effective in measuring more samples from the repository site. The results revealed seasonal variations of the massic activity of radionuclides in groundwater.

4.2 Other applications

In 2013, the laboratory participated in the intercomparison test TRIC2012 for the determination of low-level tritium activities in natural waters for age dating purposes (Hillemonds et al., 2014). In total, eight water samples were analyzed, with a massic activity of ^3H in the samples covering the range 0 Bq/kg to 56 Bq/kg. The samples were measured with the Quantulus-1220, some of them underwent an enrichment procedure. The calibration of the ultra low-level device by using solutions traceable to the national standard of activity had led to acceptable scores.

5. Conclusions

By now, the following radionuclides were successfully standardized at FTMC using the TDCR method: ^3H , ^{14}C , ^{36}Cl , ^{63}Ni , ^{90}Sr , ^{99}Tc and ^{129}I .

Tritium has been standardized with the combined relative standard uncertainty of 0.62 % ($k = 1$). The tritiated water samples, prepared after the solution has been standardized using the TDCR method, are used for calibrations of an ultra low-level liquid scintillation counter Quantulus-1220. Hence, the TDCR method, as a primary method, enables the quality assurance and quality control of the secondary radiation measuring equipment. The LSC Quantulus-1220 is used in environmental radioactivity, radiation protection and decommissioning programs.

FTMC provides support to Lithuanian laboratories via preparation of comparisons' samples traceable to the national standard of radionuclide activity.

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Coincidence summing corrections factors for volume ^{60}Co sources measured by scintillation NaI(Tl) spectrometers

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Abstract

The method for obtaining the coincidence summing correction factors for ^{60}Co measured by scintillation NaI(Tl) spectrometer in two different experimental settings using cylindrical and well-type detector is presented in this paper. For the purpose of performed experiment, the system of equations was formed, and net areas in full energy peaks as well as in entire spectrum were used as input data. The system was solved and correction factors for different measurement geometries of volume sources were determined.

Keywords: coincidence summing; correction factors; scintillation NaI(Tl) spectrometer; ^{60}Co

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1. Introduction

The gamma spectrometry often deals with the problem of true coincidence summing effects, especially in the case of well-type detectors or close source-to-detector geometries. This problem appears as consequence of simultaneous detection of two or more photons emitted in cascade from the decay of the same radionuclide within the resolving time of a detector. Therefore, if we want to determine accurate efficiencies for particular gamma-ray energies, it is necessary to determine corresponding coincidence summing correction factors, because these factors could reach values much greater than 1, sometimes even a value of 2 (Laborie et al., 2002).

The first general method proposed to solve the problem of coincidence summing is given by Andreev et al. (1972). Variety of techniques dealing with coincidence summing effects and coincidence summing correction were presented in the literature in the last few decades (McCallum and Coote, 1975; Moens et al., 1981; Debertin and Helmer, 1988; Semkow et al., 1990; Korun and Martinčić, 1993; Vidmar et al., 2003; Novković et al., 2007; Sima and Arnold, 2012).

Although the problems with the coincidence summing effects are studied more comprehensive in spectrometry with semiconductor HPGe than with scintillation NaI(Tl) spectrometers, in the last few years there are some valuable and useful papers concerning coincidence summing effects in scintillation-based spectrometry (Byun et al., 2004; Anil Kumar et al., 2009; Dhibar et al., 2013; Hansman et al., 2015; Thabet et al., 2015; El-Khatib et al., 2017). In previously mentioned papers, the methods, which significantly contribute to the improvement of understanding of coincidence summing effect, and therefore to increase the accuracy of measurements, are presented. However, a simple and reliable method for determining the coincidence summing corrections in spectrometry with scintillation NaI(Tl) spectrometers has been developed, and its application to the ^{60}Co with a simple decay scheme, is presented in this paper.

2. The case of the ^{60}Co decay

^{60}Co has a simple decay scheme with two gamma-rays in cascade. The detailed decay scheme of ^{60}Co is given by Helmer (2010). Deexcitation to the ground state of ^{60}Ni is going on, by emission of gamma-ray γ_1 ($E_{\gamma_1} = 1173.23$ keV) and γ_2 ($E_{\gamma_2} = 1332.49$ keV) in the cascade or by direct emission of

gamma-ray γ_3 ($E_{\gamma_3} = 2505.72$ keV). If the gamma-rays γ_1 and γ_2 are detected in true coincidence, coincidence losses of counts under the γ_1 and γ_2 full energy peaks will take place, whereas increases of counts under the γ_3 full energy peak will occur. Besides, the total energy deposited by one of the gamma-ray and part of the energy deposited by other gamma-ray could be detected, so the true coincidence summing effects are also dependent on the total detection efficiency.

In the case of the ^{60}Co point source, for count rates in the full energy peaks at E_{γ_1} and E_{γ_2} (N_1 and N_2), in sum peak at E_{γ_3} (N_3), and a total count rate (T), following equation were used:

$$N_1 = A \cdot p_1 \cdot \varepsilon_1 (1 - p_2 \cdot \varepsilon_{t2}) \quad (1)$$

$$N_2 = A \cdot p_2 \cdot \varepsilon_2 (1 - p_1 \cdot \varepsilon_{t1}) \quad (2)$$

$$N_3 = A(p_3 \cdot \varepsilon_3 + p_1 \cdot p_2 \cdot \varepsilon_1 \cdot \varepsilon_2) \quad (3)$$

$$T = A(p_1 \cdot \varepsilon_{t1} + p_2 \cdot \varepsilon_{t2} - p_1 \cdot \varepsilon_{t1} \cdot p_2 \cdot \varepsilon_{t2} + p_3 \cdot \varepsilon_{t3}) \quad (4)$$

where:

A – activity of the source,

p_i – photon emission intensities at energies E_i ,

ε_i – full-energy peak efficiencies at energies E_i ,

ε_{ti} – total detection efficiencies at energies E_i .

For volume source, it is necessary to integrate these equations. Thus, in the following, we approximate the volume as point source.

Equation (3) is proved to be unstable, thus the system is formed by using equations (1), (2) and (4), in which the following approximations are introduced: $p_1 \approx p_2 \approx 1$ and $p_3 \approx 0$. However, a system of three equations and four unknowns is obtained, thus it is necessary to introduce another approximation. It is assumed that the ratio of total efficiency and full energy peak efficiency for given energy is constant ($\varepsilon_{t1}/\varepsilon_1 \approx \varepsilon_{t2}/\varepsilon_2 = k$), what is reasonable as hypothesis in the cases of relatively close energies. To solve this system, as input data, area under the peaks at energies E_{γ_1} and E_{γ_2} , as well as areas in the whole spectrum (up to approximately 2700 keV) were used, whereby the data are corrected for the background, dead time and the energy range was extrapolated to 0 keV.

To solve this system of equations the program package Mathematica 5.0 (Wolfram Research) was used, whereby corrected values for efficiency were obtained. The correction factors for coincidence summing effects were determined by comparing corrected values for efficiencies with uncorrected values obtained from experimental spectrum.

3. Experimental setup

The experiment was conducted using two scintillation spectrometer systems: one consists of a NaI(Tl) well-type detector Canberra 802-4W (resolution 9.7 % at 662 keV), and the other of a NaI(Tl) cylindrical detector Canberra 802-4 (resolution 8.0 % at 662 keV), both with same auxiliary electronics: preamplifier Canberra 2005, high voltage power supply Canberra 3002 (operating voltage 1050 V), amplifier Ortec 672, and Ortec MCA Maestro.

In the measurements performed by means of NaI(Tl) well-type detector Canberra 802-4W, solution of ^{60}Co conditioned in test tube of appropriate dimensions ($\Phi = 13$ mm, $H = 98$ mm) was used, and its specific activity on the reference date was (311 ± 9) Bq/g. This solution was used for preparing so called quasi-point source (50 μl) as well as three volume sources of 3 ml, 6 ml, and 9 ml. To obtain 3 ml volume source, the ^{60}Co solution was volumetrically added into the test tube, and the weight was checked gravimetrically. Standards of 6 ml, and 9 ml volume were obtained by diluting the 3 ml solution with a suitable carrier (CoCl_2 in 1 M HNO_3), thus the total activity of volume sources still the same in all three volumes (970 ± 30 Bq). In the measurements, performed by means of NaI(Tl) detector Canberra 802-4, point source of ^{60}Co purchased from the Czech Metrology Institute (CMI) was used. The activity of this source on the reference date was (11.87 ± 0.12) kBq.

4. Measurements and results

Measurements by means of NaI(Tl) well-type detector Canberra 802-4W were performed in four different measurement geometries, i.e. with four different volumes of ^{60}Co solution, including quasi-point source. Determination of correction factors was quite simple, because all measurements of ^{60}Co solution were performed in precisely defined measurement geometries. The values obtained for correction factors (c_f) were in the range 1.313 to 1.650 for $E_{\gamma 1}$ and 1.373 to 1.807 for $E_{\gamma 2}$. These values were analyzed by using software OriginPro 6.1, and it was concluded that best fitting curve (with a correlation coefficient near unity) for their volume (v) depended values is second degree polynomial:

$$c_f = A + B_1 v + B_2 v^2.$$

Parameters A , B_1 , and B_2 , as well as correlation coefficient, r , are given in Table 1.

On the other hand, for scintillation NaI(Tl) spectrometer Canberra 802-4, determination of correction factors was faced with some difficulties. Namely, it was necessary to determine the correction factors for different measurement geometries. In order to avoid making a large number of standards with different geometries and matrices, numerous measurements of ^{60}Co point source, in different positions in relation to the detector itself, were carried out. Point sources were placed, for the measurement purposes, at positions defined by combination of 3 different distances from central axis (0 cm, 1.35 cm, and 2.7 cm) and 8 different distances from the detector end-cap (0.1 cm, 1.34 cm, 4.0 cm, 7.4 cm, 9.67 cm, 11.3 cm, 14.25 cm, and 18.25 cm). Several measurements were repeated with same combination of mentioned but with different position regarding to central axis.

Analysis of obtained values for correction factors showed that their values determined in the same plane and at the same distance from the central axis of the detector, but for different positions regarding to detector itself, have deviation of maximum ± 0.3 %. Thus, it was concluded that the NaI(Tl) crystal is quite correctly centered inside the detector housing. The values

obtained for correction factors were in the range 1.007 to 1.259 for $E_{\gamma 1}$ and 1.011 to 1.290 for $E_{\gamma 2}$. Further analysis using software OriginPro 6.1 indicates the spatial dependence of the correction factors: linear radial and exponential axial dependence. The linear radial dependence (c_{fl}) of the corrective factor versus the distance from the central axis (d) is given by:

$$c_{fl} = A + Bd,$$

whereas the corrective factors show an exponential axial dependence (c_{fa}) versus the distance detector end-cap distance (h) given by:

$$c_{fa} = y_0 + A_1 e^{-h/t_1} + A_2 e^{-h/t_2}.$$

Figure 1 presents, as example, the exponential axial dependence of the correction factors as a function of the distance from detector end-cap at the central axis.

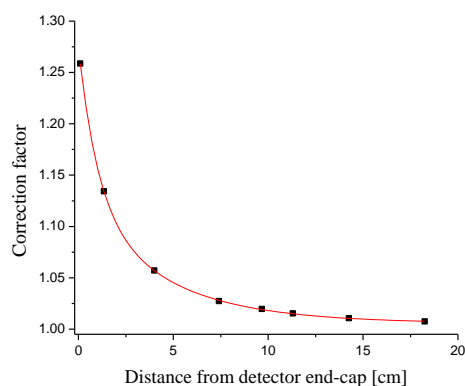


Fig. 1. Exponential axial dependence of the correction factors regarding the distance from detector end-cap at the central axis.

On the basis of performed measurements and detailed analysis, fitting curves which determine correction factors for coincidence summing effects for different measurement geometries of volume sources, were obtained. The variables in these fitting curves are the distances from the detector to the lower (h_1) and to the upper surface (h_2) of volume sources and their internal radii (x). These fitting curves are given by:

$$\begin{aligned} c_{fac l} &= y_{0c} + A_{1c} e^{-h_1/t_{1c}} + A_{2c} e^{-h_1/t_{2c}}, \\ c_{fas l} &= y_{0s} + A_{1s} e^{-h_1/t_{1s}} + A_{2c} e^{-h_1/t_{2s}}, \\ c_{fac u} &= y_{0c} + A_{1c} e^{-h_2/t_{1c}} + A_{2c} e^{-h_2/t_{2c}}, \\ c_{fas u} &= y_{0s} + A_{1s} e^{-h_2/t_{1s}} + A_{2c} e^{-h_1/t_{2s}}, \\ c_{flu} &= c_{fac l} + (c_{fas l} - c_{fac l})/2.7x, \\ c_{flu} &= c_{fac u} + (c_{fas u} - c_{fac u})/2.7x, \end{aligned}$$

where additional indices c and s denote position on the central axis and position 2.7 cm from the central axis of the detector, while l and u denote positions of lower and upper surface of volume sources from the detector end-cap.

Parameters y_0 , A_1 , A_2 , t_1 , and t_2 , as well as correlation coefficient, r , are given in Table 2.

Further analysis showed that a correction factor for a particular geometry of the sample can be determined as the arithmetic mean of coincidence summing correction factors for the four characteristic positions ($c_{fac l}$, $c_{fac u}$, c_{flu} , and c_{flu}) with an estimated uncertainty due to this approximation less than 1 %.

It should be mentioned that some preliminary measurements were performed to determine attenuation effect in the volume samples. For that purpose, three sets (each of five 125 ml PVC

Table 1. Parameters of second degree polynomial fit and correlation coefficients

	A	B_1	B_2	r
1173.23 keV	1.655 ± 0.017	-0.0243 ± 0.0094	-0.0016 ± 0.0010	0.99539
1332.49 keV	1.815 ± 0.029	-0.0206 ± 0.0153	-0.0032 ± 0.0016	0.99279

Table 2. Parameters of exponential fit and correlation coefficients

	y_0	A_1	A_2	t_1	t_2	r
1173.23 keV (c)	1.006 ± 0.001	0.149 ± 0.008	0.120 ± 0.008	1.019 ± 0.053	4.384 ± 0.265	0.99998
1173.23 keV (s)	1.005 ± 0.001	0.093 ± 0.001	0.106 ± 0.001	1.006 ± 0.001	4.427 ± 0.001	0.99999
1332.49 keV (c)	1.009 ± 0.001	0.165 ± 0.013	0.134 ± 0.012	0.991 ± 0.073	4.201 ± 0.343	0.99996
1332.49 keV (s)	1.008 ± 0.001	0.116 ± 0.001	0.105 ± 0.001	1.076 ± 0.001	4.957 ± 0.001	0.99999

boxes) were prepared. One set was used as blank (empty boxes), and other two were filled with epoxy resin and sand as matrices. Measurements with point sources were performed. On the basis of these preliminary measurements we determined that maximal discrepancies for correction factors between empty boxes and boxes with matrices for same measurement conditions are less than 0.5 %. Also, estimated uncertainty due to different chemical composition of the used matrices is less than 0.2 %.

5. Conclusions

The gamma spectrometry often deals with the problem of coincidence summing effects and therefore with determining the corresponding correction factors. This paper, as a contribution to this topic, presents the method for obtaining the coincidence summing correction factors for ^{60}Co measured by scintillation NaI(Tl) spectrometer. In experimental part there were two different measurement setups: with well-type and with cylindrical spectrometer.

On the basis of the measurements by means of scintillation NaI(Tl) well-type spectrometer and for test tube measurement geometry it was concluded that coincidence summing correction factors could be determined by using second degree polynomial dependent on the solution volume.

On the other hand, on the basis of the measurements by means of cylindrical scintillation NaI(Tl) spectrometer and for volume source geometry it was concluded that coincidence summing correction factors could be determined as the arithmetic mean of correction factors for the four characteristic positions obtained by combining exponential fit dependent on the distance from detector end-cap and linear fit dependent on the distance from central axis. Nevertheless, implementation of the integral calculus can contribute to better accuracy.

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