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TECHNICAL NOTE

The National Standard Unit of Radionuclide Activity and the related standards in Poland*

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Abstract The idea of functioning and technical realization of the National Standard Unit of Radionuclide Activity established in Poland by President of the Central Office of Measures is presented in this paper. The Radioisotope Centre POLATOM has been appointed as depositary of that National Standard. The detection circuits as well as the absolute methods of measurements used to realize the standard unit are described. Application of a liquid scintillation technique was emphasized, and the determination accuracy of measurements has been presented. All kinds of the secondary standards, prepared in the RC POLATOM for application in isotope laboratories of nuclear medicine and in industrial and scientific centres, are related to the National Standard by a chain of presented comparisons. Relationship between the National Standard Unit of Radionuclide Activity and the international system of standards has been realized in the RC POLATOM by taking part in international comparisons of the measurement results.

Key words standards • radionuclide • radioactivity • coincidence • liquid scintillator

Introduction

The National Standard Unit of Radionuclide Activity has been established in Poland by order No. 3 of President of the Central Office of Measures dated 4th February 1999, basing on the "Law of Measurement". The Metrological Laboratory of Radioactive Materials (MLRM) in RC POLATOM was appointed as depositary of that National Standard. It is obligatory for the MLRM to make available the National Standard to any user and to compare it to all secondary standards. The National Standard is made up of three coincidence measuring systems and two types of measuring sources with which the unit of measure of radionuclide activity is effected by using seven absolute measuring methods. The liquid scintillation technique (LSC) is the basis of applied counting methods where reproduction of the activity unit of measure can be obtained for any radionuclide with expanded uncertainty not higher than ±2% with the coverage factor $k = 2$ (which corresponds to the confidence level of 95%). The obtained results are verified by international comparisons of measurements.

The measuring systems of the National Standard Unit of Radionuclide Activity

Three coincidence measuring systems of the National Standard have been worked out in the MLRM, RC POLATOM, Świerk. The electronic circuits were realized in CAMAC system.

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The 4π(LS)-γ coincidence and anticoincidence measuring system

There are two detection systems: a liquid scintillation detector with two photomultipliers type EMI 9899 B working in the coincidence system and two crystal scintillators NaI(Tl) with other two photomultipliers working in the sum system (Fig. 1). The counted source in liquid form is dissolved in the liquid scintillator.

The radioactive decays are recorded in the following detection paths:
a) path "β" with liquid scintillator – recording of α, β, x, e_A,
b) path "γ" with NaI(Tl) crystals – recording of γ-photons,
c) coincidence path – recording of coincidence between α, β, x, e_A and γ-photons,
d) anticoincidence path – recording of anticoincidence between α, β, x, e_A and γ-photons.

A set of counting points in any path is obtained by changing the high voltage supplying photomultipliers path "β" and causing a change of counting efficiency.

The triple-double coincidence measuring system (TDCR system)

The TDCR system consist of a detection head with three photomultipliers type RCA 8850 with a symmetric setup at a plane level around the chamber with a radioactive source in the liquid scintillator. A general scheme of the TDCR system is presented in Fig. 2.

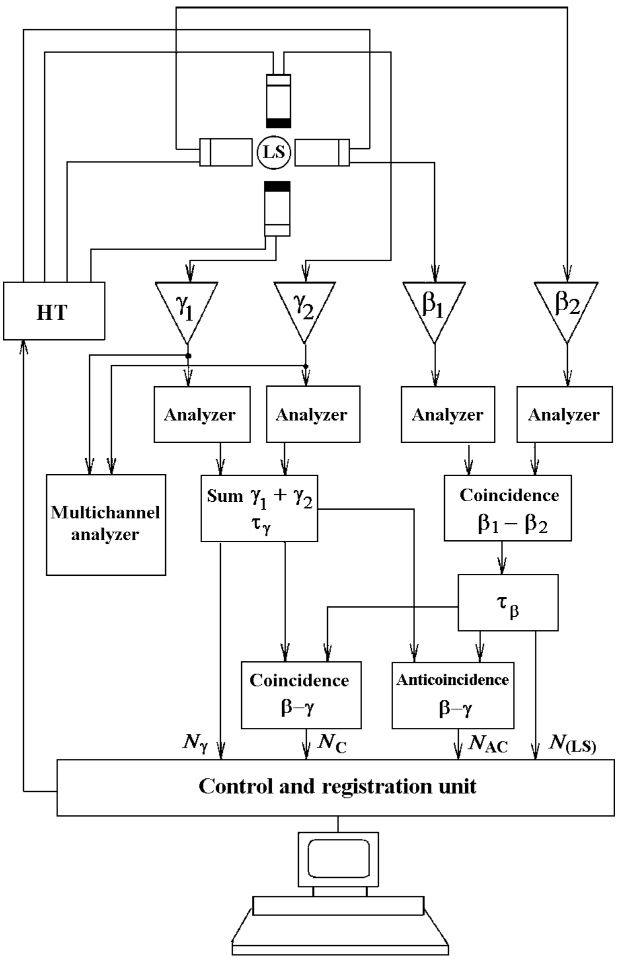

Fig. 1. A scheme of the 4π(LS)-γ coincidence and anticoincidence measuring system (LS – liquid scintillator; HT – high tension supplier; τ – dead time unit; triangle indicates amplifier).

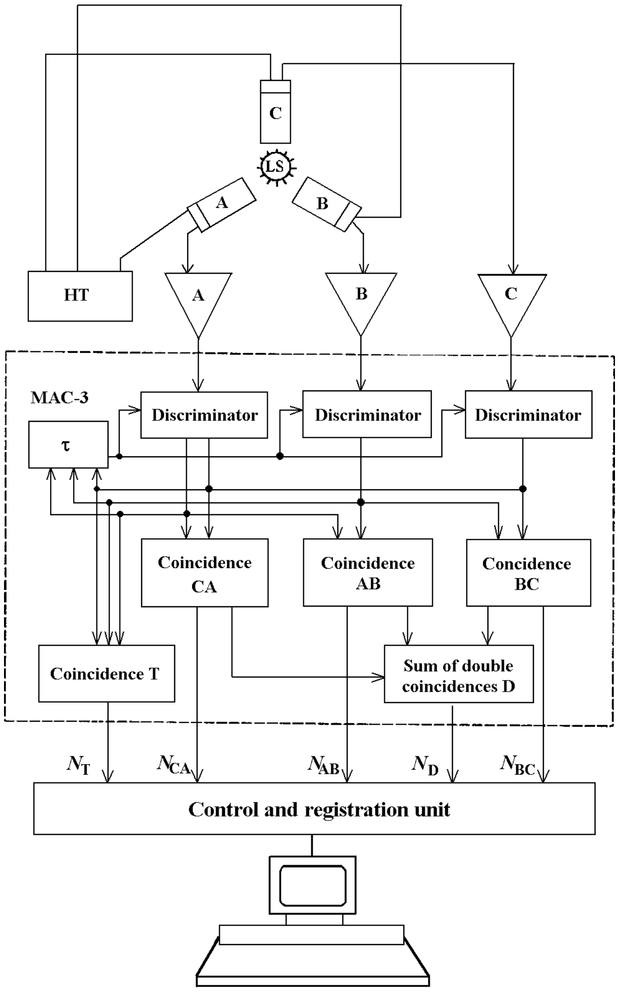

Fig. 2. A simplified scheme of the TDCR measuring system with the MAC-3 coincidence module (LS – liquid scintillator; HT – high tension supplier; τ – dead time unit; triangle indicates amplifier).

The coincidence module MAC-3 with a discrimination level below one-electron peak pulse accepts the signals from the three detection paths (A, B, C). There are three double coincidence pulses (AB, BC, CA), triple coincidence pulses (marked T) and a logical sum of three double coincidence pulses AB+BC+CA (marked D). The counting efficiency is changed by high voltage supplying the focusing electrodes of photomultipliers and a set of counting points is obtained.

The x-γ coincidence measuring system

The x-γ coincidence measuring system is made up of two NaI(Tl) scintillation detectors with photomultipliers EMI type 6097 B. The measuring point source on a Mylard foil, prepared as a dry residue after evaporation of the radioactive solution, is placed between the crystals. The pulses arising in the detectors are registered in path γ_1, γ_2, and in coincidence. A general scheme of the system is presented in Fig. 3.

The measuring methods of the National Standard Unit of Radionuclide Activity

Absolute measuring methods used at the National Standard are the following:
1. 4π(LS)β-γ coincidence and anticoincidence method,
2. 4π(LS)α-γ coincidence and anticoincidence method,
3. 4π(LS)x,e_A-γ coincidence and anticoincidence method,
4. multiparametric 4π(LS)α,β,(x,e_A)-γ coincidence and anticoincidence method,
5. 4π(LS)α,β,(x,e_A)-γ coincidence and anticoincidence tracer method,
6. triple-to-double coincidence ratio (TDCR) method,
7. x-γ coincidence method.

The LSC technique is applied in the first six methods. Application of several measuring methods gives mutual assurance of the method verification and increases the confidence and exactness of radioactive source measurements. The measuring methods from 1 to 5 are realized in the 4π(LS)-γ coincidence and anticoincidence measuring system, while method 6 – in the TDCR system and method 7 – in the x-γ coincidence measuring system. The methods described below, except for the methods 1, 2 and 3, which are very similar to the methods 4 and 5.

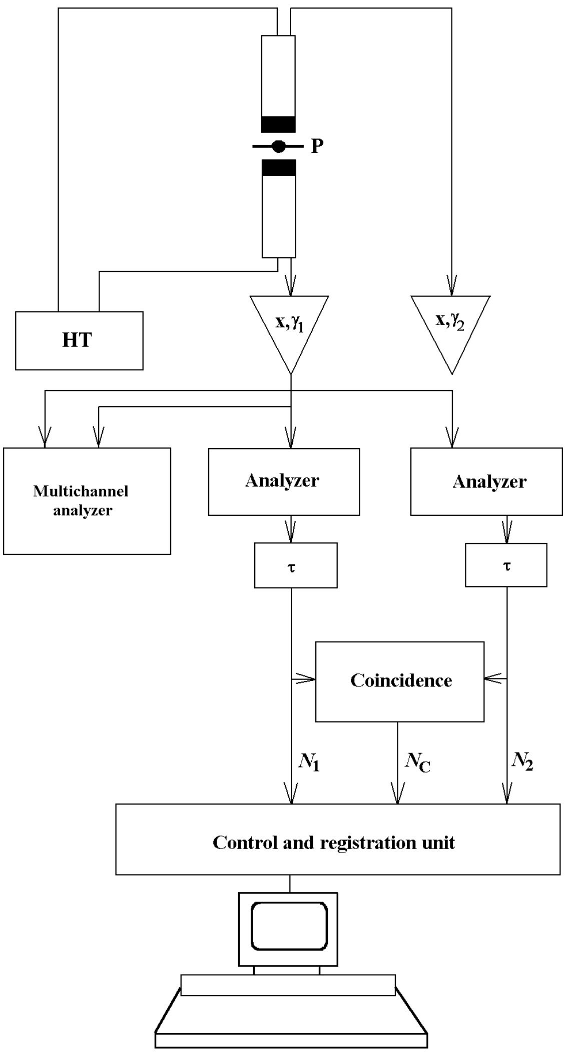

Fig. 3. A scheme of the x-γ coincidence measuring system (P – point source on the Mylard foil; HT – high tension supplier; τ – dead time unit; triangle indicates amplifier).

The multiparametric 4π(LS)α,β,(x,e_A)-γ coincidence and anticoincidence method

This is the method applied to describe the activity of β-γ, α-γ and (x,e_A)-γ emitters and also radionuclides having complex decays. Registration of disintegration rate in detection paths is realized for several chosen parameters of gamma path [4–6].

The chosen energy range of γ spectrum serves as parameter. The gamma photon counting rates $N_{\gamma 1}$, $N_{\gamma 2}$ for two-parametric methods, or $N_{\gamma 1}$, $N_{\gamma 2}$, $N_{\gamma 3}$ for three-parametric methods are registered in that chosen gamma path. The counting rate of α, β, β^+, (x,e_A) in a "beta" path $N_{(LS)}$, counting rate N_C of α, β, β^+, (x,e_A)-γ in the coincidence path, and counting rate N_{AC} of anticoincidence path (see Fig. 1) are registered for any gamma path parameter in any decay event. The "beta" counting efficiency $\varepsilon_{(LS)}$ is calculated for a given set of counting points. The linear extrapolation of $N_{(LS)}N_\gamma/N_C$ or $N_{(LS)}N_\gamma/(N_\gamma - N_{AC})$ as a function of $(1 - \varepsilon_{(LS)})/\varepsilon_{(LS)}$ to $\varepsilon_{(LS)} = 1$ is carried out.

The obtained extrapolated values N_0 determine the mean value of the counting rate for the measured radioactive source. These kinds of measurements performed for several gamma path parameters give several straight extrapolation lines. The slope of the extrapolation line determines the counting efficiency of gamma quantum in the "beta" path, depending on the complex decay and the value of internal conversion ratio. The counting rate N_0 obtained from these extrapolations for various parameters of gamma path is mutually verified.

The 4π(LS)α,β,(x,e_A)-γ coincidence and anticoincidence tracer method

This is the method applied to activity measurement of the so called "pure" β, α emitters, and also "pure" electron capture emitters [3]. The idea of tracer method is to trace the counting efficiency of a "pure" β-emitter (called the emitter) by a α, β, (x,e_A)-γ emitter (called the tracer). The tracer activity is determined by coincidence and anticoincidence methods. To calculate the emitter activity, the tracer is added and the activity of the total source is measured. Afterwards, the activity of the tracer is subtracted from the sum, and the "pure" emitter activity A_E is obtained.

The triple-to-double coincidence ratio (TDCR) method

The TDCR method is applied mainly to the "pure" α, and β emitters, as well as to the "pure" electron capture radionuclide activity measurement. To realize the method, the counting rate of various five coincidences: N_{AB}, N_{BC}, N_{CA}, N_T and N_D (see Fig. 2) are registered simultaneously. The characteristic parameter (K) of TDCR method is obtained as the ratio of the N_T to N_D counting rate [1, 2].

The theoretical counting efficiency of the counting arrangement is determined as a function of K parameter for each counting point. The disintegration rate N_0 in the measured radioactive source was calculated afterwards. Independence of the N_0 value of the counting efficiency of the TDCR system is taken as an accuracy criterion of the determination of radionuclide activity.

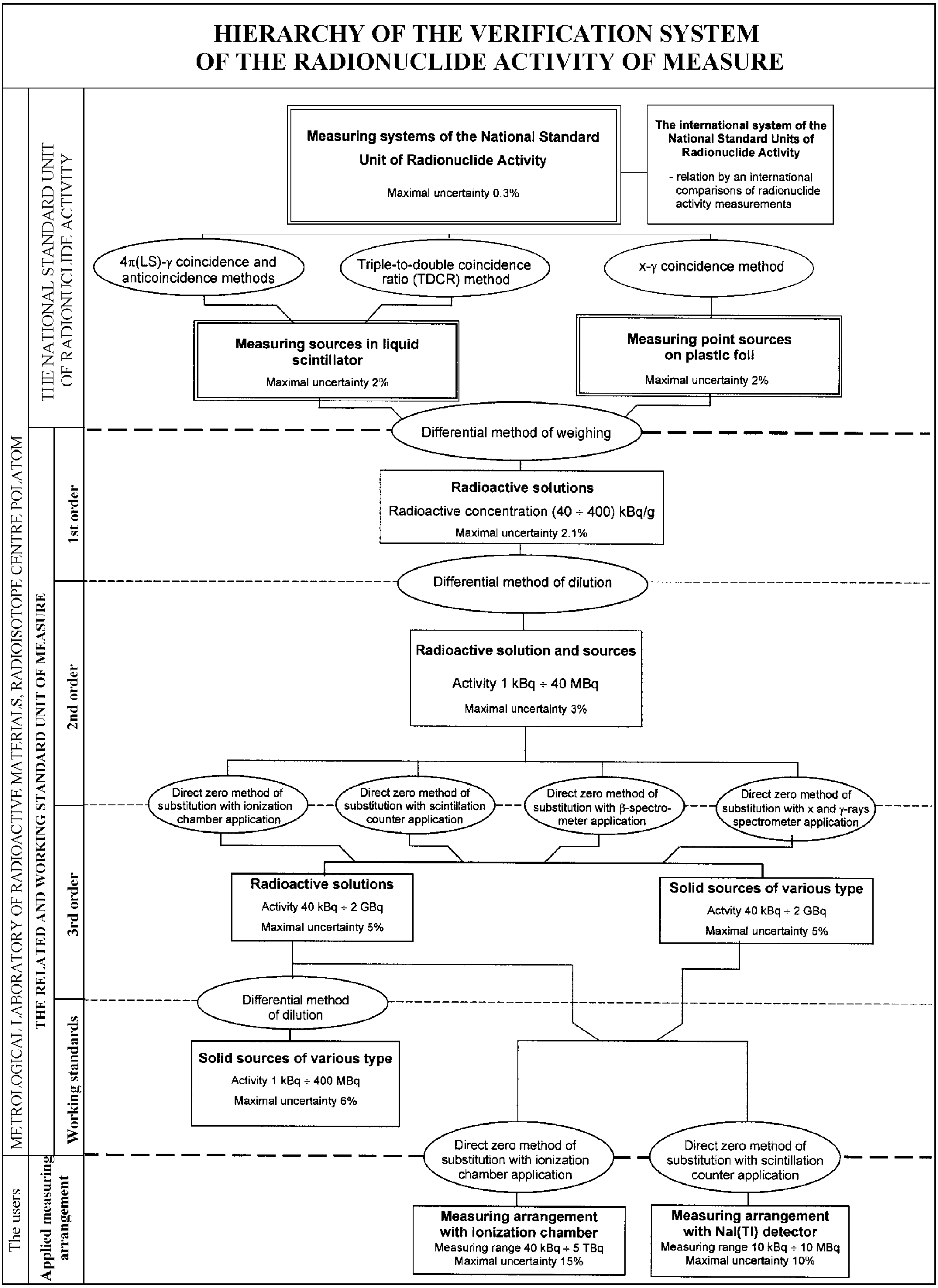

Fig. 4. Hierarchy of the verification system of the unit of measure of radionuclide activity presenting a chain of comparisons of measuring arrangements with the National Standard Unit of Radionuclide Activity.

The x-γ coincidence method

This is the method for radioactivity measurement of electron capture radionuclides that emit x and γ-photons of very close energy and, therefore are hard to distinguish. The method has been elaborated in MLRM and intended for the measurement of ^{125}I activity.

The frequency of x- and γ-photons registration in "gamma 1" (N_1), "gamma 2" (N_2), and in coincidence (N_C) paths (see Fig. 3) is taken as a basis to calculate the disintegration rate N_0 in the measured source of ^{125}I.

Hierarchy of the verification system of the unit of measure of radionuclide activity

Hierarchy of the verification system of the unit of measure of radionuclide activity (Fig. 4) presents a chain of comparisons of measuring arrangements with the National Standard Unit of Radionuclide Activity by means of the related standards. Rectangles symbolized standards of the unit of measure of radionuclide activity or measuring arrangements, and ellipsis – methods of comparisons.

In the upper part of Fig. 4, over the dashed line, the double-lines rectangles symbolize the National Standard and ellipsis – the measuring methods of the National Standard. International comparisons of radionuclide activity measurement secure the relation of the National Standard to the international system of standards, as it is shown.

The related standards of the 1st, 2nd and 3rd order as well as the working standards are obtained in the process of weighing and dilution of the standard solution. The uncertainty of the activity of each kind of standard is given as the maximal expanded uncertainty with the coverage factor $k = 2$ (which corresponds to the confidence level of 95%). The expanded uncertainty of most radionuclide solutions, being the 1st order related standards, is on the level ±1%. Dilution and other methods of comparisons cause an increase in the uncertainty of standards of succeeding orders. For preparation of the 3rd order related standards, any of four methods of substitution with the application of ionization chamber, scintillation counter, β-spectrometer, or x- and γ-ray spectrometer is applied. The last standards, in case of radioactive solution, can be used for the preparation of the working standards or, in case of solid sources, can be used directly for calibration of the applied measuring arrangements. The RC POLATOM produces the above mentioned standards of various kinds of application in isotope laboratories in nuclear medicine and in industrial and scientific centres.

Before using any radioactive solution for the standardization process and preparation of a standard source, the radionuclide purity of the solution is controlled by means of gamma spectrometry with a germanium HPGe detector.

International comparisons of radionuclide activity measurements

Reliability level and accuracy of the measuring equipment as well as measuring methods of the National Standard are verified by international comparisons of the results of radionuclide activity measurements of chosen radionuclides. These intercomparisons are organized yearly by the Bureau International des Poids et Mesures (BIPM) and other international metrological organizations. The MLRM participated in nearly 30 intercomparisons organized, among others, by BIPM ($^{90}Sr + ^{90}Y$, ^{60}Co, ^{139}Ce, ^{134}Cs, ^{55}Fe, ^{109}Cd, ^{125}I, ^{75}Se, ^{204}Tl, ^{152}Eu, ^{89}Sr), EUROMET (^{63}Ni, ^{55}Fe, ^{169}Yb), or by the International Committee for Radionuclide Metrology (^{3}H twice), and obtained satisfactory results. All information concerning the measuring methods and the results obtained in the comparison process are sent by the participants of measurements to the organizer of comparison, who is obliged to prepare a final report. Reports are presented during international conferences and published. Results of comparison collected in BIPM give a picture of the actual competence of the world metrology.

Summary

Over 30 years of experience of the Metrological Laboratory of Radioactive Materials RC POLATOM in standardization of radionuclides by absolute methods of measurements and participation in many intercomparisons has brought the establishing of the National Standard Unit of Radionuclide Activity. The LSC technique is the basis of functioning and technical realization of the National Standard. All kinds of radionuclides can be standardized by applying various measuring methods. The secondary standards, prepared in the RC POLATOM, are related to the National Standard. The expanded uncertainty of the activity of any radionuclide solutions, being the 1st order related standards, is not higher than ±2% with the coverage factor $k = 2$.

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