Ionizing Radiation Metrology Laboratory from IFIN-HH, Romania Presentation

Bercea, S., Sahagia, M., Celarel A. Razdolescu, C.

"Horia Hulubei" National Institute of Physics and Nuclear Engineering
407 Atomistilor Str. Magurele, Ilfov County, POB MG-6, 077125, Rumania.
bercea@nipne.ro

ABSTRACT

The Ionizing Radiation Metrology Laboratory from IFIN-HH has as main research area the development of standardization methods in the field of radiation dosimetry and activity measurement and assurance of traceability of measurements. In the field of dosimetry, the research is focused on the development of the method and equipment of absolute calibration for beta absorbed dose, based on the cavity chambers principle (extrapolation method). In the field of activity measurements, the research is basically in the development of absolute methods for standardization, such as: 4π Proportional Counter-Gamma coincidence method, and Liquid Scintillation Counting–Triple and Double Coincidence Ratio. The paper presents the most significant results obtained by the laboratory concerning traceability of dose equivalent rate and activity.

1. INTRODUCTION

The Institute for Atomic Physics (IAP) of Romanian Academy started its activity in 1949. It was designated as a Romanian leader in basic and applied research in Nuclear Physics, Nuclear Energy, Applications of Nuclear Technologies. Nowadays, after some restructurations, it became the "Horia Hulubei" National Institute of R&D for Physics and Nuclear Engineering, IFIN-HH.

From the very beginning, the Ionizing Radiation Metrology problems to be solved were a challenge in the Institute's activity, in direct connection with the following measurement necessities:

- Dosimetrical survey of the Basic Radiation Installations: VVRS Nuclear Reactor, Cyclotron, Betatron, Linear accelerators, Van der Graaf Tandem.
- Applications of the nuclear technologies.
- The IFIN-HH Nuclear Reactor was the main producer of radioisotopes, for practical applications. The measurement of activity of the obtained batches of radioisotopes, of the final products activity measurement and gestion of resulted radioactive wastes, were another metrological priorities. On the other side, for the development of uses of nuclear technologies, a Section for Applications of Nuclear Technologies, especially Radioactive Sources (ANT) was created during fifties. In this case, both types of measurements, in dosimetry and activity became soon necessary .
- The environment impact of radioactive substances.
- During sixties period, before the interdiction of nuclear explosion tests in atmosphere, a significant environmental radioactivity was registerred, needing precise activity measurements. This interrest was gradually diminished untill the occurence of the Cernobil accident, when the activity measurements became again a first priority.
- Personal and area dosimetry program for Cernavoda NPP deployed at IFIN-HH, during the eighties period, consisted from the design and production of radioprotection mean. Their calibration required also ionizing radiation standards.

All these practical requirements imposed the early creation of the Ionizing Radiations Metrology Laboratory (IRML), and permanent involvement to enlarge its competencies and refine the methods, in order to improve the measurement precision. The IRML response was the development of absolute standardization methods both in dosimetry and in activity measurement and their transfer to the field measurements by the development of secondary standards. The IFIN-HH, IRML role in Romanian metrology network was established through a Collaboration Protocol signed in 2005, with the National Metrology Institute, under the authority of the Romanian Bureau of Legal Metrology (BRML). It establishes the responsibility of IFIN-HH, as the owner of the Primary Standard of Activity Unit (Becquerel), and of the Reference Standard for Dose Equivalent, on national as well as international level. BRML designated IFIN-HH as a participant in the activities of the International Committee for Weights and Measures - Mutual Recognition Arrangement (CIPM-MRA) in the field of ionizing radiations, such as presented on the CIPM-MRA, Annex A. BRML appointed IFIN-HH as a representative in the EURAMET Technical Committee for Ionizing Radiations (IR-TC).The laboratory is authorized for legal metrology activities by the BRML, which also attests IFIN-HH periodically, as a calibration laboratory for the field of ionizing radiations.

2. STANDARDIZATION METHODS

2.1. Dosimetry Methods

The traceability chain was assured by the development of absolute and relative dosimetry methods.

2.1.1. Absolute Methods

The absolue method for measurement of absorbed dose in the field of medium gamma rays energy by the use of a cavity ionization chamber was elaborated during sixties-seventies [1-3]. The experience gained in the field was used for the design and construction of an extrapolation ionization chamber for the dosimetry of Beta and X-ray dosimetry [4]. For this purpose a variable volume cavity chamber was constructed, according the Bohm paper [5] (Fig. 1).

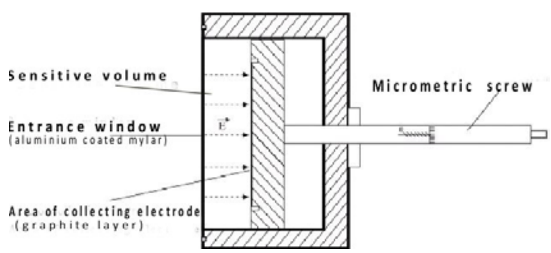

Fig. 1. Scheme of the extrapolation ionization chamber.

This chamber meets the requirements of the cavity theory, with the specific features imposed to make it compatible to the beta rays. In the case of such a chamber, dedicated to absorbed dose (D) or absorbed dose rate (dD/dt) measurement, the output quantity, accessible to direct measurement, is the electrical charge produced by the rdiation ionization in the gas from the sensitive volume, or the ionization current, respectively. In order to ensure a precise measurement of the ionization charge or ionization current, a complete and rigorous electrical characterization of the detector is necessary.

In this part of our work, we shall present only the result of the long time measurement of electrical capacity, for different values of the distance between the electrodes. For some of these distances the electrical capacity was also calculated, as follows:

Theoretical calculation of the electrical capacity of the ionization chamber.

The geometry parameters of the cylindrical chamber are: r = 30 mm and electrode distance, d, a variable parameter.

Accordingly, the electrical capacity is calculated as follows:

$$C = \varepsilon_0 \, \varepsilon_{air} \, (S/d) \quad , \qquad (1)$$

where ε_0 = 8,854·10^{-12} F/m, ε_{air} =1.00059, and S = πr^2 = 28.2743 cm^2 .

The corresponding values of the capacity obtained for different values of d –parameter are given in in Table 1.

Table 1. Capacity values for various distances.

d (mm)	C(10^{-12} F)
0.5	50.098
5	5.0098
10	2.5049
15	1.6699

For the theoretical calculated values of capacity, the maximum estimated uncertainty is ±5 %.

The method for charge measurement was developed, by taking into ccount the published literature [6-8]. The charge values recording was started, and after 5 seconds, the polarizing voltage is applied on the chamber (A sudden variation of the voltage, from 0 V to 100 V is applied, in 5 s). In the first 5 seconds of the measurement, the charge on the chamber's electrodes was Q_1. After 5 s, by charge collection in the inner electric field of the chamber, when applying the polarizing voltage of 100 V, the charge on the chamber quth is Q_2.

$$Q = Q_2 - Q_1. \qquad (2)$$

The chamber's capacity is calculated with the relation:

$$C=Q/V=(Q_2-Q_1)/V \, . \qquad (3)$$

2.2. Activity Measurement Methods

2.2.1. Development of Methods and Construction of Equipment for Absolute Standardization

The efforts were focused on two main directions:

4πPC-γ Coincidence Method
The 4πPC-γ coincidence method is used for the standardization of radionuclides emitting several coincident radiations and of pure beta emitters, by the tracing method. The first system, manually operated, was constructed during sixties; it contains a home made Proportional Counter (PC), flow type, working with pure methane and a NaI(Tl) detector. The electronics is NIM of types NE and Canberra. An X, gamma - X, gamma coincidence detection block, provided with two thin NaI(Tl) detectors for the standardization of radionuclides, such as ^{125}I, was built. Recently, a new 4πPC-γ coincidence system, based on the use of a PC, two NaI(Tl) detectors and new electronic modules, ORTEC type or home - IFIN-HH made, operated in a semi automatic regime, was constructed and is under testing [9].

Liquid Scintillation Counting
A Liquid Scintillation Counter, based on the principle of the Triple to Double Coincidence Ratio (LSC-TDCR), was built with the contribution of the LNHB-France and RC-Poland, and consists from a detection block with three photomultipliers and associated electronics [10]. At present time, some improvements are under way: new photomultipliers, a new detection block with 6 Channel Photomultipliers (CPM), a NaI(Tl) probe for the application of the 4πLS-γ coincidence method. The LabVIEW operation software is under installation.

2.2.2. Relative Measurement Methods and Equipment

Spectrometric System
A spectrometry system, ORTEC product, was recently purchased and is in final testing. It contains three detection blocks:
(i) The Gamma-ray detector: Coaxial HPGe type, ORTEC product, energy interval, 30 keV - 3 MeV. An improved shield consisting from old lead, copper and a 1 mm thick tin foil is under construction.
(ii) X-ray detector: Si(Li), CANBERRA product, energy interval 1 keV-50 keV.
(iii) Alpha spectrometry chamber, provided with a surface barier silicium detector and vacuum system.

All the three detection blocks are compatible (or adapted) to the analyse system, ORTEC product, consisting from a digital spectrometer, connected to a personal computer.The following software facilities are implemented: ORTEC GammaVision-32; ORTEC MAESTRO-32;GESPECOR, used for the calculation of coincidence summing corrections and transfer of efficiency calibration between various geometries and matrices.

Ionization Chamber CENTRONIC IG12/20A
The well-type ionization chamber CENTRONIC IG12/20A is used as a transfer instrument, for secondary standardization of radioactive sources and solutions.

Chamber Calibration
Calibration of the chamber was accomplished for various gamma emitters and for various geometries (point sources, vials with solution). The calibration figures are assigned as follows:
(i) For sealed radioactive sources, high activity sources were used;
(ii) For radioactive solutions, the calibration figures were determined for: 2 ml vials, PTB, Germany; 3.6 ml vials, BIPM-CIPM comparisons; 5 ml vials for distribution of radiopharmaceuticals, as the response values are strongly dependent of the volume of solution, mainly for low energy gamma-rays emitters.

The calibration figures were determined using solutions absolutely standardized in our laboratory by the 4πPC-γ coincidence method. The list includes 17 radionuclides. A number of 10 radionuclides were verified in Key Comparisons.

An independent calibration was performed during the year 2001 at PTB, Germany, using PTB – 2 ml vials with standard radioactive solutions for 19 radionuclides;11 radionuclides were used in both laboratories, so a comparison was possible. In all cases, the results agreed within the reported uncertainties.

Chamber Electrometric System Improvement
Traditionally, the electrometric system was operated with a set of capacitors, to be used for various activity intervals. An electrometer type Keithley 6517A is now operational. By performing parallel registrations, with both electrometers, a precise determination of the capacitos' values was done and the calibration figures were transferred to units, pA MBq^{-1}.

Large Area Multiwire, Sealed Proportional Counter
It is used for large surface alpha and beta sources used for measurement of surface contamination.

The calibration is performed in terms of activity, and particle emission rate in 2πsr geometry. The calibration in activity was performed with standard sources, gravimetrically prepared from standard solutions, while the emission rate calibration is made by using sets of sources measured directly in the upper half of the open proportional counter. The response values were determined for various source dimensions, up to 250 mm x 250 mm.

3.RESULTS

3.1. Dosimetry Measurement

According to the procedure presented at point 2.1.1., an experimental determination of the capacity values, for various electrodes distances, were determined. The experimental results are presented in Table 2.

Table 2. Experimental capacity values and comparison with theoretical calculation.

D, mm	V, V	Q_1, 10^{-10}C	Q_2, 10^{-10}C	Experimental C, 10^{-12} F	Difference vs theory, %
0.5	100	0.06	50.39	50.33	
0.5	100	0.08	50.52	50.44	
Avg				50.39	-0.2
5	100	0.004 1	4.775	4.771	
5	100	0.001 3	4.819	4.832	
Avg				4.802	-4.1
10	100	0.012	2.097	2.085	
10	100	0.013	2.086	2.073	
Avg				2.079	-17
15	100	0.010 3	1.276 1	1.265 8	
15	100	0.010 2	1.228 5	1.218 3	
Avg				1.242	-26

The uncertainity in capacity measurements was calculated as the quadratic combination of the uncertainties due to: Q_1 , Q_2 , V, measurements and had amaximum value of 4%. The comparison with theoretical values, emphasized that a parasite capacity, whose influence becomes more and more significant as the capacity lowers.

3.2 Activity Measurement

The results were summarized in the methods developed and in results obtained in international comparisons

3.2.1. Development of Standardization Methods

In the 4πPC-γ coincidence method, the main Romanian contribution was:

(i) Corrections for the dead time and coincidence resolution time [11].

(ii) Measurement of beta, electron capture-gamma emitters, by the efficiency extrapolation, accomplishment of linearity conditions [12,13] and subsequent papers.

(iii) Efficiency tracer for the pure beta emitters and efficiency extrapolation for emitters with transition to the ground level (triangular decay scheme) [14] and followings.

(iv) Standardization for special radionuclides, ^{99m}Tc, [15].

The LSC-TDCR method was implemented, and the system was used for the measurement of 9 nuclides. The method and system were verified by bilateral or key comparisons [16].

In the spectrometry methods development, the contributions are:

(i) Use of the coincidence and Compton suppression anticoincidence for the low level activity measurements [17].

3.2.2. Validation of Methods by Participation in International Comparisons

The participation at BIPM comparisons started in 1962, with ^{60}Co sources. After the adoption of the CIPM-MRA (Mutual Recognition Arrangement) and establishment of the Annex B, Key Comparison Data Base (KCDB), the RML became part of the program. It is registered with the following types of comparisons.

Key comparisons

(i) BIPM.RI(II)-K1, 6 registrations for 5 radionuclides.

(ii) CCRI(II)-K2, 15 radionuclides.

(iii) EUROMET RI(II)-K2, 2 radionuclides.

IAEA Supplementary comparisons

(i) CCRI(II)-S4 “Radionuclides activity measurement in environmental samples of water, soil and grass” (2006).

(ii) CCRI(II)-S5 “TENORM” (2007).

(iii) CCRI(II)-S6 – 2 radionuclides, as solutions.

This program allowed us to validate our methods for absolute and relative standardization, and to establish the equivalence of the primary Romanian Activity standards at the level of the CIPM-MRA. A number of 37 Calibration and Measurement Capability (CMC) files, for radioactivity, were recently introduced in CIPM-MRA, Annex C.

4. DISCUSSIONS

4.1. Dosimetry

The experimental results obtained during the present work, concerned with electrical characterization of the extrapolation ionization chamber, namely the determination of the capacity values corresponding to various electrode distances, lead to the following conclusions:

a. for small values of d, electrical capacity of the ionization chamber C, is high, lowering as the distance grows; this fact affects the variation rate of the ionization current produced by the chamber, which depends on the time constant RC; the agreement with calculated values is better for high capacity values.
b. for a dosimetric measuring system which uses as a detector, such a chamber and integrates the ionization current (for absorbed dose measurement, for instance), limitations are imposed on the possibility of discerning the fast variation of the dose rate in a fast variable radiation field.
c. high values of the electrical capacity, C, mean a low sensitivity and a low rate of variation of the ionization current, while low values of the capacity mean a high sensibility and a high rate of variation of the ionization current.

These conclusions could be very important when an experiment involving dose or dose rate measurements in variable radiation fields (as in an medical betatron, for instance) have to be performed .

4.2. Activity Measurement

The development of absolute and relative methods and equipment for staandardization allowed the assurance of the whole traceability chain, from the international until the end users level.

5. CONCLUSIONS

The Ionizing Radiations Metrology Laboratory, as a member of the IFIN-HH, accomplished its scientific duties in development of methods and equipment for absolute standardization.

The laboratory assures the whole traceability chain in the field and has long tradition of participation in international programs of comparisons.

The laboratory validated the standardization methods.

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