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THE COLLABORATION OF THE RADIONUCLIDE METROLOGY LABORATORY OF IFIN-HH, OWNER OF THE PRIMARY ACTIVITY STANDARD, WITH UNITS INVOLVED IN NUCLEAR ENERGY FIELD*

M. SAHAGIA**, A.C. RAZDOLESCU, E.L. GRIGORESCU, A. LUCA, C. IVAN

"Horia Hulubei" National Institute of R&D for Physics and Nuclear Engineering, IFIN-HH, POB MG-6, code 077125

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***Corresponding author < msahagia@ifin.nipne.ro>*

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Abstract. The paper presents the calibrations and measurements performed in the following fields: direct measurement of the activity for samples containing radionuclides such as ^{3}H, ^{63}Ni, ^{14}C, by using the Liquid Scintillation Counting, in the classical, and in the variant of Triple to Double Coincidence Ratio (LSC-TDCR); preparation of special types of radioactive solution recipients, with various radionuclide compositions, for the calibration; direct services provided to the Nuclear Power Plant, such as the calibration of standard sources, or delivery of some types of standards; propositions of future collaboration, regarding the offer of new radioactive standards, or new calibration services are presented.

Key words: radioactivity standards, calibration, Liquid Scintillation Counting, LSC-TDCR.

1. INTRODUCTION

The Radionuclide Metrology Laboratory (RML) from IFIN-HH has a long collaboration tradition with the units involved in the nuclear energy field, both in direct relation with the Nuclear Power Plant and in relation with other units involved in nuclear energetics. In a previous paper, presented at SIEN 95 [1], our contribution in development of standards to be used for the calibration of such special kind of equipment was underlined. A review of destination of all kind of radioactive standards, with a special emphasize of the nuclear field was also presented [2, 3]. On the other side, a tight collaboration with the Nuclear Research Institute (NRI), Pitesti in the field of obtaining radioisotopes was reported [4]. In the present paper, the most recent developments and the offer of future metrology services for the nuclear field are underlined.

2. RECENT COLLABORATIONS REGARDING RADIOACTIVE STANDARDS, MEASUREMENTS AND CALIBRATIONS

2.1. RADIOACTIVE STANDARDS

During the decade 1980–1990, our laboratory was involved in the Institute's program for development of dosimetry equipment, and implemented the AECL-Canada Ltd. quality standards, what offered to it the opportunity of being selected as a provider of radioactive standards for the Nuclear Power Plant (NPP). In these circumstances, the NPP ordered, and received from IFIN-HH, point and large area beta ^{90}(Sr + Y) sources, having certified the activity and the particle emission in the geometry 2p*sr*, in full agreement with the requirement of the Standard ISO 8769 [5]. It also ordered ^{152}Eu point and volume gamma-ray spectrometry sources, from our normal production.

Special requirements, regarding the physical shape of standard sources and solutions came from the NRI-Pitesti. Standard solutions from the multi-energetic gamma-ray emitters, ^{133}Ba and ^{152}Eu were used to prepare special stainless steel capsules, taking care to fill the whole volume; a very tight neck was accessible for filling, special tools being necessary; standard samples, with activities lying within the interval: 9 kBq … 3500 kBq were prepared for the survey equipment produced by the NRI for both, first and second Unit of the NPP.

2.2. MEASUREMENTS

In the frame of the own research program, we offered ourselves to expertize, free of charge, a number of oil, emulsion and water NPP waste samples, suspected to be contaminated with ^{3}H, ^{134}Cs and ^{137}Cs. A number of 55 oil, 3 emulsion and 3 water samples were extracted from the waste barrels on the NPP site, on November 21, 2001 as 15 mL vials, and transported to the laboratory. The emulsion samples were first analyzed by gamma spectrometry, according to the Analyze Procedure IFIN-HH, Code AC-PL-05-32; the radionuclides: ^{134}Cs, in proportions varying from 120 Bq/kg to 487 Bq/kg and ^{137}Cs from 736 Bq/kg to 5893 Bq/kg with an uncertainty of 16% (k=3, coverage) were found. The ^{3}H content was measured by the Liquid Scintillation Counting, in the Department for Life and Environment Physics, by Dr. Niculina Paunescu, according the Procedure Code AC-PL-08-05. The uncertainty was 15% (k=3) for all the samples. The emulsion samples contained ^{3}H from $1.2{\cdot}10^{6}$ Bq/kg to $8.5{\cdot}10^{8}$ Bq/kg, the water samples, from $1.9{\cdot}10^{6}$ Bq/kg to $2.5{\cdot}10^{9}$ Bq/kg, while oil samples contained from $1.8{\cdot}10^{3}$ Bq/kg to $3.9{\cdot}10^{7}$ Bq/kg; the majority of them contained about $3.0{\cdot}10^{5}$ Bq/kg. Following the measurements, a number of 53 Analysis Reports, Form Code AC-PC-CPR-02-01, were issued, with no.19/2.04.2002…71/2.04.2002. It is important to underline the practical use of the reports, as they refer to the limits imposed by the Fundamental Norm for Radiation Protection, NFSR 01, CNCAN Ord.14/2000 and point the samples situation regarding the exemption or exclusion levels.

2.3. CALIBRATION SERVICES

Our laboratory has a very good and continuous relation with the laboratories of the NPP for the standardization and certification of all kinds or Radioactive Standard Sources, made in IFIN-HH, or imported: alpha sources, beta point and large area sources, and all kind of gamma point or volume sources.

At the end of the year 1998, in the frame of the IFIN-HH - NPP collaboration arrangement, a team from LMR, provided with standard solutions, from the radionuclides ^{57}Co, ^{60}Co, ^{131}I, ^{137}Cs, ^{241}Am prepared in our laboratory, went to the NPP and carried out the calibration of the Liquid Effluent Monitor. In order to do that, they prepared on the site, special recipients with standard solutions, similar with those used for liquid effluent monitoring. The whole calibration process, consisting in the determination of the response values for every radionuclide, $[(\text{imp} \times s^{-1})/\text{Bq}]$, on the measurement interval, was carried out and calibration certificates were issued by our laboratory.

3. NEW POSSIBILITIES OF COLLABORATION

3.1. DEVELOPMENT OF A NEW ABSOLUTE STANDARDIZATION METHOD AND EQUIPMENT; NEWRADIOACTIVE STANDARDS

Our most important concern in the specific nuclear energetics measurement problems in Romania is the development of the method and equipment for absolute standardization of low energy pure beta radionuclides, such as ^{3}H, ^{63}Ni, ^{14}C. On this purpose, with the contribution of the French colleagues, from the Laboratoire National Henri Becquerel (LNHB), a Liquid Scintillation Counter, based on the principle of the Triple to Double Coincidence Ratio (LSC-TDCR) was constructed in LMR.

The method consists in the followings: a well determined quantity of the solution to be standardized is dissolved in a Liquid Scintillator solution. The emitted radiations interact with the scintillator, resulting in the emission of luminous photons, by the fluorescence process. They produce photoelectrons as consequence of interaction with the photocathodes of three photomultipliers, disposed in a 120^0 geometry. These photomultipliers are connected to three circuits of double coincidences, and a circuit of triple coincidence, through a special module, MAC 3, a French product, allowing the calculation of the detection efficiency, which is less than 100%. The system and the standardization methods were validated by participation at international comparisons [6-9].

The determinations are made both experimentally and theoretically, by registering the double and triple counting rates, and a mathematical model for the efficiencies calculations. The detailed description of the method is presented in the above cited papers. A short description is presented in the followings.

The counting rates, R_D, R_T, are expressed as function of the activity, N_o, by the relations:

$$R_D = N_0 e_d\,;\ R_T = N_0 e_t\,, \tag{1}$$

where $\boldsymbol{e}_d$ and $\boldsymbol{e}_t$ are detection efficiencies.

The system is equilibrated, and the efficiencies $\boldsymbol{e}_d$ and $\boldsymbol{e}_t$ are correctly calculated, when:

$$\frac{e_d}{e_t} = \frac{R_D}{R_T}. \tag{1'}$$

For the calculation of e_d and e_t, mathematical relations, containing spectra shapes $S(E)$, expressed by Fermi function for beta emitters, emission probabilities and energies for discrete spectra (alpha, Auger and conversion electrons, influence of X and gamma radiations) are used.The activity is calculated as

$$N_0 = \frac{R_D}{e_d} \ \text{ or } \ N_0 = \frac{R_T}{e_t}. \tag{2}$$

The DETECSZ and TDRCB-1, or other new programs, written at the LNHB, France, are used for calculations.

The LSC-TDCR method and equipment allowed the preparation and legal qualification of new standard solutions, from the above pure beta radionuclides, which will be used for the calibration/metrological check of all existing scintillation counters in the future.

3.2. CALIBRATION OF TRITIUM MONITORS

As it is already known, a sensitive tritium monitor, designed for the measurement of tritiated water vapors, provided with a 8.7 L ionization chamber, type MT-1 [10] was made in IFIN-HH. The classical method used for the calibration of tritium monitors had been developed by Osborne [11].

In our laboratory, an original method, based on the continuous circulation in a closed circuit of saturated HTO vapors, was developed [12]. The saturated vapors are obtained by bubbling from vials containing tritium standard solutions with various activity levels, standardized by using the LSC-TDCR method. The main difference from the method of Osborne, is the use of saturated vapors, which eliminates the measurement in liquid scintillator of the recovered non-saturated vapors. The measurements were carried out for three levels of the activity concentration: 0.0180, 0.701 and 1.73 $MBq\cdot g^{-1}$.The mean response was R=$2.25\cdot 10^{-13}$A/($MBq\cdot m^{-3}$) with a relative combined uncertainty lower than 4%. The value agrees with the result obtained with the Osborne method, R_{Os}=$2.30\cdot 10^{-13}$ A/($MBq\cdot m^{-3}$) and with the estimated theoretical value, $2.29\cdot 10^{-13}$A/($MBq\cdot m^{-3}$).The method may be used in the future for the calibration/metrological check of all tritium monitors designed for tritium vapor concentration measurement .

3.3. INTERNATIONAL RECOGNITION, AUTHORIZATIONS AND ACCREDITATIONS OBTAINED BY LMR

As a consequence of the continuous participation in international key comparisons and of the development of methods and equipment for preparation and standardization of radioactive sources and solutions, IFIN-HH was designed by the Romanian Bureau of Legal Metrology (BRML) to represent Romania in the Mutual Recognition Arrangement of the International Committee for Weights and Measurements (CIPM-MRA). IFIN-HH is a member of the Consultative Committee for Ionizing Radiations, Section II, Activity, CCRI(II); it is also the representative in the Ionizing Radiations Technical Committee (IR-TC), EUROMET. At the national level, IFIN-HH, via our laboratory, is recognized as the owner of the Primary Standard of Activity (Becquerel) in Romania.

The laboratory and the personnel are authorized for deploying nuclear activities, all the radioactive standards that we produce and deliver obtained Authorization for Radiological Security from the National Commission for Nuclear Activity Control (CNCAN). In agreement with the requirement of the Metrology Law, the LMR is authorized by the BRML for legal metrology activities (Authorization B-042-05/12.12.2005); the laboratory members are also authorized as metrological verifiers, by the BRML.

Regarding the Quality Assurance System, besides the old qualification by the AECL regulations, the new Standard SR EN ISO/IEC 17025:2001 [13] was applied. The Quality System of the whole Ionizing Radiation Metrology Laboratory of IFIN-HH, including the Radionuclide Metrology Laboratory, was presented and approved by the EUROMET QS Forum, for National Metrology Institutes, Bucharest, February 2005 and Q-TC EUROMET Meeting, La Valletta, Malta, February 2006. The CNCAN designated our laboratory as a Notified Standardization/Calibration Laboratory for Activity measurement in the Nuclear Field, Notification LE 02/2005. The BRML designated the laboratory as an Attested Standardization Laboratory (Attestate B-12-20-05/12.12.2005). The Laboratory is also under the process of accreditation, by the Romanian accreditation body, RENAR.

4. CONCLUSIONS

The Radionuclide Metrology Laboratory had a fruitful, continuous collaboration with the units involved in Nuclear Energy field, consisting from delivery of all kind of radioactive standards, standardization/calibration, measurement services.

The new equipment and standardization method, LSC-TDCR, will allow for an amplified involvement of the LMR in the measurement of tritium and other low energy beta emitters as well as in the calibration of tritium monitors.

The international recognition, national authorizations and accreditations are a guarantee for the recognition of measurements and certificates abroad, as well as for their legality and quality certification.

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