ORIGINAL PAPER

METHODS FOR THE REALIZATION OF RADIOACTIVE STANDARDS AND FOR THE ENVIRONMENTAL RADIOACTIVITY MEASUREMENT

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Abstract. *The radionuclide metrology assures: the realization, the conservation and the transmission of the "activity" standards, in various fields of measurement, environmental radioactivity characterization being included. The Radionuclide Metrology Laboratory (RML), IFIN-HH, usually prepares radioactivity standards (reference materials), consisting from: standard solutions, point and large area alpha and beta standard sources, and gamma volume standard sources used for the measurement of environmental samples and for the calibration of surface contamination monitors.*

The most important measurement methods used in the environmental radioactivity measurements, developed in IFIN-HH, are: gross alpha and gross beta measurements and gamma-ray spectrometry measurements. The choice of the adequate method depends on several factors, such as: type of emitted radiation, existence of a single or mixture of radionuclides, physical shape of the matrix.

The authors describe the methods for the realization of standard sources used for the calibration of equipment used in such monitoring and methods for the environmental radioactivity measurement.

Keywords: *radioactive standard sources, environmental radioactivity measurement.*

1. STANDARDIZATION METHODS FOR RADIOACTIVE SOURCES

Our laboratory has undertaken radionuclide metrology studies since sixties, materialized by: the designing and the construction of standardization installations, the development of methods for the absolute standardization and preparing activity standard sources.

The activity standards consist from standard radioactive sources: point and large area alpha and beta standard sources, used for the environmental radioactivity measurement samples and for the calibration of surface contamination monitors; and standard solution used for the standardization of medical calibrators. These sources are prepared in the Radionuclide Metrology Laboratory, from IFIN-HH. Also, the RML prepare volumic standard sources. The volumic activity standard sources having matrices type water equivalent, soil, zeolyte, with different radionuclides are used for energy and detection efficiency calibration of the gamma spectrometry systems and other systems which are used for measuring low activities for the water, soil and food samples.

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These systems are working in Environment Ministery, in Agriculture Ministery, in Industry Ministery and in Health Ministery.

To standardize these sources the RML use absolute and relative methods. The absolute standardization methods used are:

i) the method of standardization in defined solid angle (including 4πsr detection geometry for radionuclides emitting a single type of radiations).

ii) the coincidences methods for the radionuclides emitting coincident radiations.

The relative standardization methods are:

i) the method of standardization with large area proportional counter for the calibration of the alpha and beta sources in 2πsr particle emission.

ii) 4π gamma ionization chamber method for secondary standardization of radioactive solutions.

iii) NaI(Tl) detector for the calibration of gamma-ray sources.

iv) the method of standardization with semiconductor detector.

2. RADIOACTIVITY STANDARDS PREPARED IN OUR LABORATORY

2.1 POINT AND VOLUMIC GAMMA STANDARD SOURCES

Kits of point sources for gamma spectrometry containing: ^{241}Am, ^{57}Co, ^{133}Ba, ^{134}Cs, ^{137}Cs, ^{60}Co, ^{152}Eu standard sources [1], with activities of 4 kBq, 40 kBq, 400 kBq (uncertainty 2 – 3.5%), are directly prepared quantitatively from standard solution, or prepared from high concentration solutions and standardized by the relative method [2, 3].

After the Cernobyl accident and increase of interest for the measurement of a big number of volume samples by gamma-ray spectrometry, the RML started the preparation of reference materials for such measurements. The first stringent request was for the contamination measurement of liquids like water and milk, or approximately water equivalent solids like meat. Most of the measurements are carried out by the designed Romanian laboratories, in cylindrical samples, SARPAGAN-geometry, external dimensions: diameter = 78 mm, height = 41mm. The most important contaminants were ^{134}Cs and ^{137}Cs. Other radionuclides of interest were: ^{241}Am, ^{133}Ba, ^{152}Eu and ^{60}Co.

The preparation method of the volumic activity standard sources with water equivalent matrix is an originaly method, which allows obtaining a very good quality for sources. The acrylamide used for the matrix, containing standard radioactive solution is transformed in solid polyacrylamide by irradiation, with the atomic number Z_{eff} = 3.7 and density ρ = (1080±20) kg m^{-3}, such as reported by [4]. The preparation procedure is quantitative and the uniformity of sources is very good. The polyacrylamide matrix was then adopted in many world laboratories: chemical polymerization [5, 6], radiation polymerization [7].

The sources obtained in this way are used for measuring the food and enviromental samples with a gamma spectrometry system. The use of the volumic activity sources, which are delivered with an authorised calibration certificate, assures the measurement traceability to the national standard. The volumic activity standard with soil matrix, having the density in interval 1200-1500 kg/m^3 is used to measure the soil activity and in studies regarding the interaction of soil with water and vegetables.

Other volume sources are prepared from zeolyte matrix. A gravimetric, multilayer spiking with standard solution, followed by the homogenisation of samples by using a vibratory sieve shaker, is used. The sources have also a SARPAGAN geometry and a density ρ = (700 ±20) kg m^{-3}.

All three types of volume sources are directly traceable to primary Romanian activity standard, as they are prepared individually by gravimetric dispensing from standard solution.

2.2 POINT AND LARGE AREA ALPHA AND BETA STANDARD SOURCES.

Alpha ^{241}Am sources were prepared by electro-deposition. The beta standard sources, from ^{63}Ni, ^{35}S, ^{147}Pm, ^{204}Tl, ^{90}(Sr+Y), with dimensions from point sources up to 250 x 250 mm are prepared both by electrodeposition such as reported by [8] and by absorption of quantitative dispensing standard solution on thin paper foil and sealing between two thin plastic foils. The activity is calculated and emission rate in 2πsr geometry is directly measured [3].

2.3 STANDARD RADIOACTIVE SOLUTIONS

All kinds of emitters alpha-gamma, pure beta, beta-gamma, electron capture- gamma, electron capture, positron-gamma are prepared as standard solutions. These are radioactive solutions, stable physico-chemical, wich are absolute standardize using the coincidence methods, or the relative method with the ionization chamber CENTRONIC IG12/20A. Lately, the most types of the radioactive solutions prepared and standardized in the RML are used in radiopharmaceutical therapy (^{153}Sm, ^{177}Lu, ^{186,188}Re), such is presented in [9, 10, 11]. All standard sources and solutions were locally qualified and authorized by the National Commission for the Control of Nuclear Activities (CNCAN). The EURAMET Q-CT recognized the laboratory Quality System and on national level the RML is accredited by the Romanian Accreditation Association (RENAR).

3. MONITORING OF THE ENVIRONMENTAL RADIOACTIVITY

According to the main objective of environmental radiation monitoring, assessing the dose to the public from the manmade sources and from natural radiation sources is a priority.

The significant part of all the air contaminants is *alpha* or *beta – gamma emitters.*

Because of that the most important measurement methods used in the environmental radioactivity measurements, developed in IFIN-HH, are: gross alpha and gross beta measurements and gamma - ray spectrometry measurements.

Considering their origin there are two types of contaminants; *natural* - ^{222}Rn and ^{220}Rn, heavy metals ^{212,214}Pb, ^{212,214}Bi, ^{214,216,218}Po and *artificial* – long life -^{90}Sr, ^{137}Cs and short life ^{99}Mo, ^{103}Ru, ^{131}I effluents, discharged during the operation of nuclear installations.

In our Institute the environmental measurements and monitoring, radioactive measurements devices and installations have a long tradition and importance due to the fact that is a nuclear centre, involving research, production and use of radioisotopes Radioisotope Center and the Department for Radioactive Wastes Treatment, where open radioactive sources are manipulated, a nuclear reactor under decommissioning and a radioactive wastes treatment unit, the radioactive content of air is continuously monitored on the placement of the IFIN-HH area.

All the measures are taken, in order to avoid any elimination of radioactive gaseous effluents over the established limits in atmosphere; a permanent survey is undertaken, in order to adequately limit the radiological hazard in the shortest time.

In IFIN-HH, the Department of Life and Environment Physics, in charge with the environmental survey, established procedures for sampling, measurement and reporting of the

results. In this department for the air survey and sampling is based on the radiometric method the using of the air filters [12]. The authors described the methods in the cited paper, the measurements are carried out at established time intervals chosen such as to cover the measurement of the short lived natural radon daughters, as well as the monitoring of long lived natural and artificial radioisotopes.

The measurements are performed by the following methods*: gross alpha and beta activity measurements* 3 minutes after collection, after 20 h, and after 5 days, by the use of a high sensitivity gross alpha-beta activity system; *gross gamma activity* after 5 days from prelevement, by the use of a gamma measurement system.

In general the aspiration is done within pre established periods of time, minutes up to several hours (1-5), the total volume of air can vary between 19 000-21 000L.

The air filter, N0521147 type, of known porosity, 1μm, retention yield of 96-99%. Usually the aspiration point is placed at a height of 16 m from the soil for aerosols, and the aspirations are performed every two weeks.

The *radiometric method* it is used for determination of gross alpha and gross beta activity measurements of various types of samples: air and aerosols samples, drinking water, effluents, ground water, surface and rain water, sediment, different types of soils, green, vegetation, milk.

The radiometric method has advantages and limitations; of course the perfect method of measurement does not exist. For example this method exclude volatile constituents such as ^{3}H, ^{241}Pu, ^{35}S but assures a minimum detectable activity of 0.20Bq/L, 8 counts/min for a measuring time of 50 min, 1L of processed water and 26% measurement efficiency, 80% yield field for AB-S-28 model and 0.03 Bq/L for MPC-9300 model.

Another method used in the environmental survey, within the paper [13] the authors present a high sensitivity TLD system, designed and used for the survey of the environmental radioactivity based on the use of TL detectors type LiF:Mg, Cu, P,which are also commercially known as GR 200A. The dosimeter designed within our Institute, uses 3 detectors, and the measurement value is calculated as the arithmetic mean. The very sensitive, TL Reader, READER ANALYSER RA'94 was chosen and an optimal thermal cycle was designed, in order to enhance the measurement performances. For each of the placements, a set of 3 dosimeters is used, and survey intervals are chosen from 1 to 100 days, depending on the radioactivity level and reporting requirements.

The technical characteristics of the system were determined by exposing the dosimeters in reference X and gamma radiation fields, as required by the IEC standard 61066:2006 [14]. The main technical parameters ensure the highest quality and recommend it for use in the survey of the environmental radioactivity, at the level of ambient dose equivalent rate, due to the normal natural radioactivity, in open areas.

4. CONCLUSIONS

The RML method is very good and many laboratories adopted it.

All standard sources and solutions, obtained in RML are locally qualified and authorized.

The results obtained are in good agreement for the methods used in environmental monitoring and mentioned above, at the same time, the conclusion is that the main part of the measured radioactivity is due to the radon short life daughters: lead and bismuth, such as the measurements performed at various intervals showed.

The content of the artificial radionuclides is situated within the same interval as in other Romanian areas where no nuclear activities are deployed.

REFERENCES

[1] Farcasiu, O., Sahagia, M., *Calitatea Productiei si Metrologie*, **11,** 695, 1972.
[2] Sahagia, M., *Rom. Rep. in Physics*, **54**, 193, 2001.
[3] Sahagia, M., Razdolescu, A.C., Ivan, C., Equipment and standards for surface contamination measurements, *IRPA 9 Congress Proc.*, **3**, 34, 1996.
[4] Sahagia, M., Grigorescu, L., *Nucl. Instr. Meth. A*, **312,** 236, 1992.
[5] De Felice, P., Biagini, R., Fazio, A., *Internal report – Preparazione e caratterizzazione di fantocci antropomorfi di tipo „BOMAB" per taratura di sistemi „WBC", INMRI, ENEA*, Dip. Ambiente, Centro di Ricerche Casaccia, Roma, Italia.
[6] Morris, W. M., Simpson, B. R. S., *Appl.Radiat. Isot.*, **60**(2-4), 557, 2004.
[7] Yamazaki, I. M., Koskinas, M. F., Diaz, M. S., Andrade e Silva, L. G., Vieira, J. M., Desenvolvimento de fonts radioactivas solidas em resina acrilamida, *III Congresso Ibero Latino Americanoe do Caraibe de Fisica Medica*, Anals di Congresso, **1**, 1, Rio de Janeiro, 2004.
[8] Farcasiu, O., Sahagia, M., *St.Cerc.Fizica*, **1**(25), 135, 1973.
[9] Sahagia, M., Razdolescu, A. C., Cimpeanu, C., Grigorescu, E. L., Luca, A., Ivan, C., Preparation and standardization of a $^{153}SmCl^{3}$ radiopharmaceutical solution, *Proc. Intern. Conf.,"Applications of High precision Atomic& Nuclear Methods" -HIPAN 2002*, Ed. Academiei Romane, 101, 2005.
[10] Sahagia, M., Razdolescu, A. C., Grigorescu, E. L., Luca, A., Ivan, C., Lungu, V., The standardization of 177Lu and its use in nuclear medicine, *NEMEA-2 Conf. Proc. EUR Report EN 22136, Luxembourg, EC,* Ed. EC-JRC-IRMM, 181, 2005.
[11] Sahagia, M., Razdolescu, A. C., Grigorescu, E. L., Luca, A., Ivan, C., *Appl.Radiat.Isotopes*, **56**(1-2), 349, 2002.
[12] Stochioiu, A., Sahagia, M., Bercea, S., Ivan, C., Tudor, I., *Romanian Reports in Physics*, **61**(3), 581, 2009.
[13] Stochioiu, A., Sahagia, Tudor, I., *Romanian Journal of Physics*, **54**(7-8), 711, 2009.
[14] ***** IEC Standard 61066:2006, *Thermoluminiscence dosimetry systems for personal and environmental monitoring*.