NEEDS OF RADIOACTIVITY STANDARDS AND MEASUREMENTS IN THE APPLICATIONS OF NUCLEAR TECHNOLOGIES

MARIA SAHAGIA

“Horia Hulubei” National Institute of R&D for Physics and Nuclear Engineering
IFIN-HH RO 76900 POB MG-6 Bucharest

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Abstract: Some aspects regarding the use of radioactivity standards and measurements in various applications are emphasized. The definitions of the specific notions, as they are used in the field of metrology, a short history of the development of radioactivity standards and of the first traceability assurance achievements are presented. The main radioactivity measurement fields, requiring standards are underlined and some relevant application examples are exposed. Our Laboratory achievements in the obtaining and distribution of standards and in carrying out special measurements are shortly presented, too.

Key words: radioactivity, standard, measurement, traceability, equivalence.

1. INTRODUCTION

Metrology involves three distinct branches [1]:
(i) scientific metrology, dealing with the development of systems of standards, aimed to guarantee the materialisation of the units of measurement;
(ii) industrial metrology, dealing with the accomplishment of adequate technical conditions for the measuring equipment and instruction of the staff, in order to assure the continuity of the traceability chain;
(iii) legal metrology, promoting the control activities, on behalf of the government. Legal metrology entered first into operation in connection with the trade development as a sum of rules governing changes. That is why, nowadays the Metrology Institutes belong to the Trade and Industry Departments. Metrology operates with some specific notions, as follow [2]:
– Standard in English derives from the old French “estandard” designing a flag raised on a pole to indicate the rallying point of an army. It tends to be used as a fixed reference point.
Calibration or “the art of measurement”. It derives from the French “calibre” the measurement of “the bare of a gun”. It consists in the determination of the response or detection efficiency of an instrument, by using standards.

Uncertainty [3] is “a parameter, associated with the result of a measurement, that characterizes the dispersion of the values that could reasonably be attributed to the measurand" or the accepted limit of variation of the stated result of the measurement.

Traceability [4], “the property of a result of a measurement whereby it can be related to appropriate standards, generally international or national standards, through an unbroken chain of comparisons”. Traceability must be understood as [5] “a vertical, hierarchical concept adopted by individual countries for their national measurement systems”.

Equivalence is [6] “the condition of being equivalent, i.e. equal for practical purposes, in significance or worth. Note: equivalence does not imply identity”. This concept applies to the national laboratories in their international relations; it is a horizontal concept.

Key comparison is [6] “one of the sets of comparisons selected by a Consultative Committee to test the principal techniques and methods in the field”.

2. SHORT HISTORY AND PRESENT TRENDS IN THE FIELD

In our country Al. Ioan Cuza promulgated the law for the adoption of the metric system in 1864 [7]. In 1875, 18 countries signed the convention of the Metre; the Bureau International des Poids et Mesures (BIPM) was established at Sèvres, as custodian of the international standards of mass, length and time. Romania adhered to the convention of the Metre in 1883.

Ionizing radiation metrology appeared after the discovery of ionizing radiations (particularly X-rays in 1895); radionuclide metrology, dealing with the precise measurement of the activity of the radioactive samples, developed after the discovery of radioactivity in 1896.

The first radionuclide activity measurements were made by Marie Curie and the first unit of measurement was a $10^{-11}$ A ionization current measured between the parallel plates of a condenser with a piece of pechblende inside.

The first radioactivity standards were prepared by Marie Curie and Otto Hönigschmidt in 1911 by weighing very precisely small quantities of radium chloride and sealing them in Thuringian glass ampoules; a platinum wire was fused at one end of the ampoule. Two pieces of the series were chosen as primary standards at the BIPM in Sèvres and the Institut für Radiumforschung in Vienna. All the others were compared with these standards. In 1934, Professor Hönigschmidt prepared other 21 samples, which constituted the first national radioactivity standards.
They were approximately immuable ($^{226}$Ra has $T_{1/2} = 1600$ years) and universal as at that time no artificial radioisotope was discovered.

The activity unit “Curie” was established as the number of $\alpha$ particles emitted during one second by the radon in equilibrium with 1 g of radium, namely $3.7 \times 10^{10}$ s$^{-1}$.

In 1950, this activity unit was attributed to all the radioactive isotopes. Recently, the SI unit Becquerel, 1Bq=1 s$^{-1}$ replaced it.

The obtaining of the artificial radionuclides in 1934 at a cyclotron ($^{32}$P) produced a challenge in matter of radioactivity standards, as many of them had much shorter half-lives, and consequently the mass of substance was no more a measure, and the measurement of the activity had to be carried out by measuring the effects of the emitted radiations. These are very different from a radionuclide to another, which are usually accepted as having different disintegration schemes. Particular methods for the standardization of every radionuclide were developed. The immediate consequence was the lost of meaning for the primary $^{226}$Ra BIPM standard. The new international system of radioactivity standards consisted in international comparisons of the measurements made by the national laboratories. NBS from USA and NRC from Canada organized the first comparison in late 1940s for $^{131}$I and $^{60}$Co. During the period 1955-1958 several comparisons regarded the radionuclides $^{24}$Na, $^{32}$P, $^{131}$I, $^{198}$Au, etc. They were organized by the International Commission for Radiation Units (ICRU). Since 1963, this responsibility has been transferred to the BIPM. Our laboratory has participated constantly in these comparisons since 1962. At present, it is recognized that such large-scale comparisons must be organized only for special situations (new radionuclides, new methods to be validated, etc.) and they become key comparisons. For gamma-ray emitting radionuclides, the new equivalent of a “primary standard” consists in two ionisation chambers situated at the BIPM and their response values, $A_e$, known as the “Système International de Référence” (SIR). These $A_e$ results, compared with the key ratios, $A_{ek}$, obtained during the key comparisons, establish the degree of equivalence between the participating labs. A liquid scintillation counter, situated also at the BIPM, is intended to be the “primary standard” for alpha and pure beta-ray emitters.

At the national scale, the national metrology labs, which are equivalent with the international counterparts must assure the continuity of the traceability chain. What is then traceability? Besides the rigorous definition presented above, it can be [2] “the competence that can be periodically demonstrated”.

They deal with the development of the most precise and adequate methods and equipment for standardization of radionuclides, as described by E.L.Grigorescu [8]. The first step in ensuring the local traceability is the elaboration of the adequate types of radioactivity standards – solid, liquid, gas standards whose activity is certified with uncertainties of 0.1%-5% for a precised coverage factor k. Adequate methods for using the standards in special measurements are put into operation too, to be offered to the laboratories. These are the scientific aspects of radionuclide metrology.
Technical aspects regard the traceability exercises, national comparisons and technical checks, metrological verifications.

The following chapters present the specific needs of standards and measurements in some defined fields, such as: nuclear power plant operation, personal radioprotection, environmental survey, medical applications of the radioisotopes. Our response to the real situations that we were asked to solve, and our results are presented, too.

3. USE OF RADIOACTIVITY STANDARDS AND MEASUREMENTS IN DIFFERENT APPLICATIONS

3.1. NUCLEAR POWER PLANT (NPP) OPERATION SURVEY

A large extent of applications of the radioactivity standards in these complex installations can be found, covering the following areas:

3.1.1. Calibration and metrological check of the equipment used in the dosimetric survey of the personnel and environment

In this case, the main measurement systems can be classified as [9]:

- Equipment for area and personal external contamination measurements, which contains large area, thin window alpha and beta detectors (PC, GM, scintillation) and associate electronics. Alpha ($^{241}$Am, $^{238}$Pu, $^{239}$Pu) and beta ($^{63}$Ni, $^{14}$C, $^{35}$S, $^{147}$Pm, $^{90}$Sr, $^{204}$Tl) large area standard sources, having dimensions from 1-2 mm$^2$ to maximum $250 \times 250$ mm, activities from 10 Bq to 400 kBq, uniformity of the emission better than \(\pm 10\%\) and uncertainty of activity and $2\pi$sr particle emission of the order \(\pm(3-6)\%\)\,(3\(\sigma\)) are required.

- Equipment for global alpha and beta contamination measurement of the environmental samples, provided with low-level counters (PC or NaI (Tl)) in difference, anticoincidence circuits and Planchet systems. They use the same alpha and beta radionuclides as above and the gamma-emitters $^{60}$Co and $^{137}$Cs. The standards must have activities from 2-3 Bq to 500 Bq, active diameter 50 mm and maximum uncertainty \(\pm 5\%\) (3\(\sigma\)).

- Usual gamma spectrometry systems provided with PGe or GeLi detectors which are calibrated in energy and efficiency by using standard spectrometry sets consisting of point sources, prepared from gamma monoenergetic emitters covering the energy interval 60-2000 keV, as: $^{241}$Am, $^{57}$Co, $^{135}$Ba, $^{22}$Na, $^{137}$Cs, $^{54}$Mn, $^{60}$Co, $^{88}$Y with activities of 4.0, 40, 400 kBq and uncertainty \(\pm(1-4)\%\) (3\(\sigma\)).

- Pipeline spectrometric systems used for monitoring of the fisil elements. The standards consist of multigamma emitters solutions $^{133}$Ba, $^{155}$Eu.
Equipment for the effluent activity measurement. The liquid effluent monitoring is accomplished both by using beta ray detectors, or gamma-ray spectrometers. For their calibration standard solutions of $^{90}\text{Sr}+^{90}\text{Y}$, $^{204}\text{Tl}$, $^{147}\text{Pm}$, $^{60}\text{Co}$, $^{137}\text{Cs}$ and mixture of monoenergetic gamma emitters are used. The activity range is from 3 kBq to 60 MBq; the uncertainties must be from $\pm 1.5\%$ to $\pm 5\%$.

A special problem is the NPP operation is the occurrence of gas effluents, consisting mainly in: $^{131}\text{I}$ and noble gases as $^{85}\text{Kr}$, $^{88}\text{Kr}$, $^{133}\text{Xe}$, $^{135}\text{Xe}$, and in the CANDU NPP, tritium as tritiated water vapours.

In the case of $^{131}\text{I}$ one measures cylindrical charcoal filters with different dimensions. The needed standards are $^{131}\text{I}$ or mock iodine ($^{133}\text{Ba}+^{137}\text{Cs}$ in definite ratios) uniformly spiked in charcoal matrices prepared from standard solutions, with activities from 50 Bq to 500 MBq and uncertainty less than $\pm 5\%$.

In the case of noble gases, the measurement equipment must be provided with recipients where the gas flows in a controlled manner and detectors for beta rays (GM or CP). Many studies were performed to decide whether a volumic noble gas source is needed, or a point source and Monte Carlo calculation of the detection efficiency is enough. In the process of the calibration of the equipment for NPP Pécs, Hungary, it was established that the most accurate method is the use of a noble gas standard [10].

Tritium as tritiated water vapours is measured by using different calibrators, provided with large volume ionization chambers[11]. Calibration methods are based on the use of saturated vapours of tritiated water [12] and tritiated water standard solutions with uncertainties less than $\pm 5\%$.

**3.1.2. Special measurements requiring radioactivity standards**

The most known method for the monitoring of the neutron flux values is the use of activation foils as: $^{198}\text{Au}$ for thermal neutrons, $^{32}\text{S}(n, p)^{32}\text{P}$ for fast neutrons, $^{58}\text{Co}$ threshold indicator and so on.

The measurement of $^{198}\text{Au}$ activity implies difficulties as the heavy gold metal produces significant self absorption, and supplementary corrections must be made [13].

Other measurements refer to the monitoring of the activation nuclides $^{63}\text{Ni}$, $^{60}\text{Co}$ and fission products as $^{134}\text{Cs}$, $^{137}\text{Cs}$, $^{90}(\text{Sr}+\text{Y})$, $^{106}(\text{Ru}+\text{Rh})$, etc. which can occur in the moderator as a result of the corrosion of the fuel elements and other metallic components. Standard solutions containing the above radionuclides are used to calibrate the equipment used in measurements (2$\pi$PC or $\gamma$ spectrometers).

Some radioactivity measurements are useful in the determination of other “non nuclear” parameters, as presented by Dryak [14] who monitored the leakage of water from the primary into secondary circuits in the NPPs Temelin and Dukovany.
The fast neutrons (threshold 10 MeV) nuclear reaction $^{16}\text{O}(\text{n},\text{p})^{16}\text{N}$ and measurement of $^{16}\text{N}$ activity were used. $^{16}\text{N}$ is a radionuclide with $T_{1/2}=7.1$ s which emits gamma quanta with the energy $E_{\gamma}=6.13$ MeV, beyond the usual energies used for the calibration of gamma spectrometers. This problem was solved by using a Pu$(\alpha,\text{n})^{13}\text{C}$ emitting gamma rays of 6.13 MeV, standardized with an uncertainty of ±8%. In order to verify the efficiency determination and validate the efficiency curve, a gamma spectrometry set, containing additionally $^{208}\text{Tl}$ (2614 keV), $^{24}\text{Na}$ (2754 keV) standards, was used.

### 3.2. PERSONAL RADIOPROTECTION

Three categories of personal exposure to the action of radioactivity can be distinguished; professionally exposed persons, patients investigated or treated in nuclear medicine procedures and the population as a whole. The main damages are produced by the radionuclides ingested or inhaled from the environment or injected in nuclear medicine procedures, as well as by the contamination of the skin. The main effort is made to determine the content of radioactive elements on the skin and in the body, by using contaminometers; whole body counters, or collimated detection systems in the case of the measurement of a special organ like thyroid, head or lungs contaminated by U, Th, Pu inhalation.

Subsequent efforts were made to establish the optimum models for the calculation of the effective doses due to these radionuclides [15], based on the ICRP 60 publication principles [16] which establish the relations existing between the absorbed dose and effective dose.

The most usual systems regard the detection of gamma rays (as in the case of $^{131}\text{I}$, $^{134}\text{Cs}$, $^{137}\text{Cs}$ from the Chernobyl accident) and standard phantoms, containing standard solutions from these radionuclides are prepared. During the measurements of lung contamination due to the low energy X-rays which must be detected, the phantom and thorax skeleton absorption must be accurately reconstituted and uranium, thorium and plutonium standards must be introduced in the “lung phantom”[17].

A special method and standard phantom construction was reported in the case of the measurement of $^{90}\text{Sr}$ fixed in the bones, for the population living the Southern Urals Region (Russia) near the Mayak plutonium breeding plants as a result of the use of the Techa river contaminated water. Several $\beta\gamma$ and bremsstrahlung measurements were made, but the most precise was the measurement of the $^{90}\text{Y}$, the $^{90}\text{Sr}$ daughter, with $E_{\text{max}}=2281$ keV [18]. The phantom contained standard $^{90}\text{Sr+Y}$ solution pipeted on small pieces of paper placed inside, outside and into holes drilled in a human skull bone. The scalp was made of skin-equivalent wax. A large area proportional counter, working in anticoincidence, with another beta particle detector was used; a detection limit of 60 Bq was reached.
The maximum activity found for Tscheljabinsk inhabitants was 22.5 kBq of $^{90}$Sr which resulted in a supplementary irradiation of these persons with an absorbed dose of 500 mGy, over the background value of 260 mGy and external irradiation of 240 mGy [19]; a 20 mGy/kBq absorbed dose constant was considered.

### 3.3. ENVIRONMENTAL SURVEY

Everywhere the environment contains radionuclides. The natural component consists of the long life terrestrial $^{238}$U, $^{232}$Th and their radioactive series, $^{40}$K, as well as the cosmogenic radionuclides as $^{14}$C, $^{7}$Be, $^{3}$H and others. The artificial contamination is produced in normal operation of NPP as consequence of nuclear weapons production, test and uses and mainly of the accidents in NPP operation (Three-Mile Islands, Windscale, Cernobyl), the inadequate treatment and storage of radioactive wastes. The main contaminants are $^{131}$I (short life) and $^{134}$Cs, $^{137}$Cs, $^{90}$Sr as well as $^{235}$U, $^{238}$U and transuranian isotopes Np, Pu, Cm, Am. Minor contaminations can be due to accidents in other nuclear facilities or hospitals. The consequence of high levels of contamination is the exposure of the whole population to internal and external irradiation. At the same time, some special measurements of very low activities, in conditions of low level background, are necessary for the testing of reference materials and for some experiments of fundamental physics.

#### 3.3.1. Natural radioactivity measurements

As is already known, two noble alpha radioactive gases, $^{222}$Rn (radon) and $^{220}$Rn (thoron) are descendents of $^{238}$U and $^{232}$Th series. They are exhaled from the solid matrices (soil, building materials) into the air. Their descendents are: $^{218}$Po, $^{214}$Pb, $^{214}$Po, $^{212}$Pb, heavy metals, attached to the aerosols in the air, which are inhaled by the subjects. The mean natural irradiation of the population is 2400μSv y$^{-1}$, about 1/2 of it being attributed to $^{222}$Rn, $^{220}$Rn, and their descendants, according to UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) publications. There are regions with an enhanced natural radioactivity, mainly due to human activities, and consequently the risk of a supplementary irradiation must be evaluated. All the measurements and dose evaluations are based on the accurate measurement of $^{222}$Rn activity. Different systems, consisting of alpha detectors as ZnS(Ag), solid track, ionization chambers, or gamma detectors like NaI(Tl), GeLi or thermoluminescent detectors, need a calibration, using $^{222}$Rn standards.

They are generally obtained from a $^{226}$Ra standard solution which emits $^{222}$Rn, circulated in different glassware systems; the activity is measured both absolutely [20] or using a calibrated GeLi for the measurement of the descendants’ activity. Glass recipients, sealed or provided with adaptors for $^{222}$Rn circulation through the installations to be calibrated, are prepared as $^{222}$Rn standards.
3.3.2. Artificial radioactivity measurements

Regarding the artificial contamination measurement, combinations of alpha, beta, gamma spectrometry with radiochemistry methods are used. In order to make measurements and to evaluate the yield of radiochemical processing, suitable standards must be prepared. Alpha emitters are generally measured after the chemical separation and electrodeposition [21]. Alpha electroplated or vacuum evaporation standard sources, with spectrometrical qualities and the $2\pi\text{sr}$ emission of particles accurately known are necessary for the calibration of the passivated, ion implanted planar silicon (PIPS) detectors used in spectrometry.

Beta emitters are chemically separated and measured both as solid sources and then beta standard sources, from the radionuclide to be measured, mainly $^{90}\text{Sr}$, $^{90}\text{Sr}$ or $^{99}\text{Tc}$, $^{63}\text{Ni}$ are used or by using low level liquid scintillator spectrometers, which are calibrated by using standard solutions from the above mentioned radionuclides.

Both alpha and beta measurements have a common characteristic: their radiations have short ranges and the detectors have high efficiencies, besides the low attainable background levels, so small volume, low activities are measured.

In the case of the gamma emitters, due to the long ranges of the gamma rays, low efficiencies and generally high background values, the gamma spectrometry analyses are accomplished using nondistructive methods, and high quantities of materials are measured. Due to the large number of gamma emitters existing, the spectrometrists calibrate their detectors (GeLi, HPGe or NaI(Tl), CdTe, etc.) for some recommended energies and draw calibration curves by analytical methods. For these operations, they need standard sources from monoenergetical emitters in the region 60 keV-2000 keV (see point 3.1.1.) as separate standard sources and solutions, or mixtures from these radionuclides. The multigamma emitters $^{133}\text{Ba}$ and $^{152}\text{Eu}$ are used with precaution, due to the coincidence summation effect. In some cases standards from a special radionuclide as $^{134}\text{Cs}$, $^{24}\text{Na}$, $^{131}\text{I}$ (mock) are needed. The main problem is to obtain a large volume (Marinelli or cylindrical) of solid sources, having the same matrix or a similar one with the samples to be measured: gases ($^{85}\text{Kr}$), liquids (solutions) and mainly solid: water equivalent, soil, vegetation, ash, etc.

The most known standards are prepared by spiking and uniformizing the respective matrices with standard solutions [22]. A new tendency is the preparation of standards from soil, vegetation, and even animal residues (shellfish, bone ash), collected from areas near nuclear facilities, as the Cumbrian Coast (northwest side of the UK) containing all the natural and artificial contaminants of the environment. International teams carry out individual measurements in low level laboratories and the final certified activities of the individual radionuclides are the mean values [23,24]. The respective spectrometric systems were previously also calibrated using standard sources.
3.3.3. Accuracy of environment measurements

The need of adequate standards and methods of coincidence summation evaluation was underlined in two national comparisons that we organized during 1992 and 1993 regarding the measurement of $^{133}$Cs, $^{137}$Cs, $^{152}$Eu activities of water equivalent volume standard sources [25,26]. While for $^{137}$Cs (monogamma emitter) the results showed a gaussian distribution, the mean $R = A_{measured}/A_{certified} = 1.0$, in the case of $^{134}$Cs the mean $R = 0.93$; $A_{measured}$ and $A_{certified}$ are activity values measured by the participant laboratories and respectively certified by our laboratory. The result was similar with a national comparison organized in the UK. In the case of $^{152}$Eu, some participants applied summation corrections, and others used $^{134}$Cs standards; both corrections were positive and compensated; consequently mean $R = 1.0$.

For this reason, large volume, similar matrix, monoenergetic sources and special nuclides are needed. Calculation models for coincidence summations, validated through the adequate standards use are issued [27].

3.4. MEDICAL APPLICATIONS OF RADIOISOTOPES

Medical applications are the most extended field of radioisotope applications; they are used for diagnosis as well as for therapy purposes. The diagnosis methods are “in vivo” and “in vitro”; the radiotherapy is based on: teletherapy, brachytherapy and radiopharmaceuticals [28,29].

In all these applications, the correct measurement of activity is of maximum importance, as the basis of the calculations of the effective doses for patients, which must be very near the optimum recommended values. Too high values expose them to unnecessary damage, while the too lower ones have not the expected results. As a general rule, the reglementations in the field as National or European Pharmacopoeias [30] impose in the medical practice limits of uncertainty of $\pm 10\%$ which implies use of radioactivity standards with uncertainties less $\pm 5\%$.

3.4.1. Measurement of radiopharmaceuticals activity

The most extended field of nuclear technology in medicine relies on the use of radiopharmaceuticals, practiced in nuclear medicine units.

The use of in vivo techniques for diagnosis purposes is based on radionuclides emitting gamma rays in the range 100-600 keV, with low electron emission (electron capture, isomer transitions) with short half-lives like $^{99m}$Tc (80% from total), $^{111}$In, $^{67}$Ga, $^{201}$Tl, $^{125}$I, $^{81m}$Kr and positron emitters as: $^{18}$F, $^{11}$C, $^{15}$N, $^{12}$O. In our country $^{131}$I is still used, as gelatin capsules for thyroid diagnosis (scintigraphy and iodine uptake tests) although its use results in high irradiations of the patients. The “in vitro” investigations known as Radioimmuno analysis (RIA), Immuno Radiometric Analyses (IRMA) and Radioreceptor Analysis (RRA) including also Tumour Markers use the electron capture radionuclide $^{125}$I, considered to be optimum for these purposes.
The widest development is concentrated now on the use of radiopharmaceuticals for therapy and palliation of oncological diseases. The radionuclides of interest belong to the following types: alpha emitters $^{211}$At, $^{213}$Bi, high energy pure beta emitters as $^{90}$Y, $^{32}$P, $^{89}$Sr and strong beta – weak gamma radionuclides as $^{153}$Sm, $^{166}$Ho, $^{169}$Er, $^{177}$Lu, $^{186}$Re, $^{188}$Re, $^{199}$Au [31]. $^{131}$I plays the most important role in the therapy of thyroidian affections (cancer and hyperthyroidism).

New radiopharmaceuticals used in labelling of DNA and RNA, with action at subcellular level belong to the Auger electron emitters as $^{193m}$Pt, $^{195m}$Pt, $^{125}$I.

Some specific problems of measurements are implied in this field.

Measurement of the radiometrological parameters as: activity, specific activity (activity per solid mass) radioactive concentration, radionuclidic purity and radiochemical purity, both in the production unit and in the control laboratories.

Several types of equipment are used:

The activity and their derived units are measured by using radioisotope calibrators provided with well type, high pressure, ionisation chambers. Generally these devices are calibrated by their manufacturers both directly, using standard solution of the above mentioned radionuclides, or by the national metrological laboratories, referring to high precision calibrated ionization chambers [32].

Due to the rapid development of the field, the newly introduced radionuclides are under study only within the national laboratories, no calibration figure being provided to the users. Also, some manufacturers use inadequately the standards or only Monte Carlo simulations of the responses, resulting in high errors in calibration [33].

The radionuclidic purity is measured by the high resolution gamma spectrometry. As the limit of allowed impurities is generally less than 0.1%, it is necessary to know very accurately the efficiencies for the main radionuclide and the possible impurities, as well as the decay scheme parameters of the two. Standards from all these radionuclides are necessary [34]. The radiochemical purity is generally measured with a radiochromatograph, which must be tested for the radiopharmaceuticals to be verified, with solid standard sources.

In hospitals, the staff distributes the radiopharmaceuticals for injections or oral administration. Commercially available calibrators are used to measure the initial received vial and patient doses (serynges). The decalibration or misuse of these calibrators are very frequent, and traceability exercises are necessary besides the metrological checks.

An example of a complete solution in our laboratory regarding the $^{186}$Re, $^{188}$Re radiopharmaceuticals measurement was recently presented in the paper “Precise measurement of $^{186}$Re, $^{188}$Re, radiopharmaceuticals” accepted for presentation in the ICRM 2001 Conference, Braunschweig, Germany, May 2001, Life Sciences Section [34]. The two radionuclides were standardized absolutely by the $4\pi$ PC-$\gamma$ coincidence method, with combined uncertainties of 0.98 and 0.48%.
theoretical and experimental beta detection efficiencies for different branches were
determined and compared with literature data; the responses of the secondary
standard CENTRONIC IG 12/20A ionization chamber were measured and the
individual contributions due to gamma – rays and bremsstrahlung – radiations were
evaluated. The standards were used in the calibration of the ROBOTRON M-27-03
calibrator belonging to the Radioisotope Department and of a CURIEMENTOR – 2
hospital calibrator.

The decay schemes of the $^{186}$Re, $^{188}$Re isotopes are still studied, as they were
neglected in the past. In the case of $^{186}$Re we determined the emission probabilities
of the emitted quanta by using the gamma efficiency values determined with a $^{57}$Co
standard source (122.06 keV and 136.74 keV).

Our results compared with other literature data are presented in Table 1.

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<td>137.16</td>
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<td>0.0935(10)</td>
<td>0.0939(9)</td>
<td>0.0945(16)</td>
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These results were not included in the above paper, but they are in agreement
with others; our uncertainty was of the same order.

3.4.2. Measurements in brachytherapy

Historically, one of the first applications of the $^{222}$Ra sources beginning 1901,
was the brachytherapy. $^{60}$Co, $^{137}$Cs sources followed, but due to the high gamma
energies, they are no more used. At present the brachytherapy procedures are based
on sealed sources as follows [39]. Gamma sources (beta rays filtered by shielding)
from $^{192}$Ir, $^{169}$Yb used mainly for intracavitar and interstitial therapy; both as
manual systems using needles, hairpins wires, seeds, or high activity (370 GBq)
seeds for authomatical after loading systems.

Beta or beta-gamma baloons or stents used in arterial angiography for the
prevention of restenosis occurrence contain $^{32}$P, $^{90}$Y, $^{188}$Re, $^{198}$Au, $^{125}$I. Particular
attention is now devoted to the 20-35 keV X-ray emitting radionuclides $^{125}$I, $^{103}$Pd,
which are used as seeds in the treatment of the prostate cancers. All these sources
with different physical configurations and shieldings (platinum, stainless steel,
titanum) raised difficult questions to the metrologists, with respect to two aspects:
the precise measurement of the activity, the chagement of the beta, X, gamma ray
spectra in the shieldings and the calculation of the air kerma rate (AKR) values in
the range of action for their radiations. As the most used radionuclide was $^{192}$Ir,
special efforts were made to standardize it absolutely [40] to calibrate the
radioisotope calibrators for it, and compare these results with calorimetric
measurements [41].
An interesting study of the AKR values and their distribution map was accomplished by a Romanian team [42] for $^{192}$Ir needles prepared in our Radiosotope Department and used in the Oncological Institute, Bucharest.

The beta emitting applicators are calibrated in terms of absorbed dose by using beta extrapolation chambers, which are calibrated using large area beta sources, having a good uniformity and high activities (more than 40 MBq).

### 3.4.3. Measurements in teletherapy

The irradiation units, containing large activities of $^{137}$Cs, $^{60}$Co are characterized in terms of AKR (or exposition rate) measured with calibrated debitmeters, for which calibration stands containing standard sources (with uncertainly less ± 5%) are built. Their activity is measured by relative methods, leaving from standard solution, point standard sources and so on.

### 4. RADIOACTIVITY STANDARDS AND MEASUREMENTS ACHIEVED IN OUR LABORATORY

Our laboratory has undertaken radionuclide metrology studies since the sixties, materialized by the designing and the construction of standardization installations and the development of methods for the absolute standardization, as presented in paper [8]. As a practical result, the Romanian standards are recognised as equivalent of the international ones. A number of 39 radionuclides are offered as standard solutions, more than 45 being standardized. In 1983, our Laboratory was recognized as a Producer of Radioactivity Standards by the IAEA and included in the International Directory of Certified Radioactive Sources.

These achievements allowed us to offer the majority of the necessary radioactivity standards for different applications as presented in chapter 3 and to assure the traceability of standards and measurements carried out in Romanian laboratories at the national level.

#### 4.1. RADIOACTIVITY STANDARDS OBTAINED IN OUR LABORATORY

These standards include radioactive solutions, solid standard sources and gaseous standards and were presented in several publications [9,29].

Two significant applications required us to solve rapidly the practical necessity of a large variety of standards: the Dosimetry Program for NPP (IFIN Bucuresti-Nuclear Equipment Laboratory and SCN Pitesti ) during the period 1980-1989 and the Environmental survey, after the Chernobyl accident 1986 (mainly regarding the volume standard sources). In both situations our response was prompt and adequate, which is proved by the large volume and variety of standards that we delivered to the NPP Cernavoda and the environmental, sanitary and alimentary survey networks throughout Romania.
The second unit in NPP construction and operation is expected to require us new standards in the near future.

4.1.1. 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.

Besides the single radionuclide solutions, standard mock \(^{131}\)I (a mixture of \(^{133}\)Ba and \(^{137}\)Cs) as well as a mixture of several monoenergetic radionuclides \([43]\) were obtained:

- Recently: \(^{89}\)Sr, \(^{3}\)H, \(^{169}\)Yb solutions were standardized.

They cover all the requirements of standard solutions, as presented in chapter 3, regarding applications in Nuclear Power Plant Equipment, Personal Radioprotection, Environmental Measurements and Nuclear Medicine.

4.1.2. The solid standard sources cover the usual areas of interest as follows:

- Alpha \(^{241}\)Am sources, spectrometric quality, covering all the Romanian requirements \([44]\).
- Beta standard sources, from \(^{63}\)Ni, \(^{35}\)S, \(^{147}\)Pm, \(^{204}\)Ti, \(^{90}\) (Sr+Y), with dimensions from point sources up to 250 x 250 mm prepared both by electrodeposition \([45]\) and by absorption of standard solution in thin paper foils \([46]\). The activity, as well as the particle emission in a \(2\pi\)sr geometry, is certified.
- Gamma standard sources were prepared as:
  - Kits of point sources for gamma spectrometry containing: \(^{241}\)Am, \(^{57}\)Co, \(^{133}\)Ba, \(^{22}\)Na, \(^{137}\)Cs, \(^{54}\)Mn, \(^{60}\)Co, \(^{88}\)Y, \(^{152}\)Eu standard sources \([47]\) with activities of 4 kBq, 40 kBq, 400 kBq (uncertainty 2 – 3.5\%, 3\(\sigma\))
  - Volume standard sources from \(^{241}\)Am, \(^{153}\)Ba, \(^{134}\)Cs, \(^{137}\)Cs, \(^{152}\)Eu, \(^{60}\)Co or mixtures from the above radionuclides with activities of 500-5000 Bq, uncertainty of \(\pm 5\%\), cylindrical \(\phi = 73\) mm \(h = 42\) mm and \(h = 32\) mm, with the following matrices: water equivalent \([48]\) soil \([49]\) and zeolit \([50]\).

Except Marinelli geometries, water equivalent sources, absolutely all the requirements were accomplished.

4.1.3. Gaseous standards

\(^{222}\)Rn standards, extracted from a standard \(^{226}\)Ra solution, sealed in glass capsules, were obtained; their activity was measured using a GeLi calibrated detector and measuring the progeny activity in equilibrium with \(^{222}\)Rn \([51]\)
4.2. MEASUREMENTS FOR TRACEABILITY ASSURANCE MADE IN OUR LABORATORY

Traceability can be assured at the national level by three types of actions: delivery of high quality standards to all the laboratories involved in radioactivity measurements, metrological check of the measurement equipment used in laboratories, and organization of national comparisons.

4.2.1. Delivery and certification of standards

All types of standard, required by different users were offered in conditions of legality radiological security approval (issued by CNCAN) and metrological approval issued by the BRML (Biroul Român de Metrologie Legală). Imported or Romanian standards were also certified after their standardization.

4.2.2. Equipment verification

Some equipment, mainly designed for solutions activity measurements, like radioisotope calibrators belonging to our Department, hospitals, control laboratories, NPP or liquid scintillation devices, were checked and certified [52].

4.2.3. Organization of national comparisons

The most relevant check of the traceability assurance is the organization of national comparisons. Four such comparisons, regarding the measurement of the activity in Romania, were organized.

Two comparisons, organized during 1992, 1993 regarded the measurement of volume environmental samples. Their results were largely presented in the papers [25, 26] and discussed at the point 3.3.3. Two other comparisons, regarding the measurement of radiopharmaceuticals in hospitals, were organized in 1994 for $^{131}$I [53] and in 1998 for $^{57}$Co ($^{99m}$Tc mock) [54].

The general conclusion was that less than 70% from the hospitals measure the activity administrated to patients with uncertainties less than ± 10% (according to Pharmacopoeias requirements) but there are still many others measuring with higher uncertainties.

The results of comparisons were presented as official documents to the Inspectorate for Public Health from the Ministry of Health and to the National Commission of Nuclear Activities Control. As a consequence, they imposed to the hospitals to submit their radioisotope calibrators for metrological verification in our Laboratory.

5. CONCLUSIONS

- Every radioactivity measurement must be carried out only with calibrated and certified equipment. These operations require the choice and use of the appropriate standards with respect to the radionuclide activity and its uncertainty physical construction characteristics.
Almost all the types of radioactivity standards: solid, liquid, gaseous required for the applications of nuclear technologies in Romania were obtained and offered to the implied laboratories by the Radionuclide Metrology Laboratory of IFIN-HH.

The traceability chain was assured in our country by the distribution of standards, the accomplishment of special measurements and the metrological check of the measurement equipment and organization of national comparisons.

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