Report to the CCRI Section II on the activity carried out at the ENEA-INMRI on radionuclide measurements in the period 2005-2007

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1. INTRODUCTION

The present report summarizes the 2005-2007 activities carried out at the National Institute for Ionising Radiation Metrology (ENEA-INMRI) in the field of interest of CCRI Section II, i.e. radionuclide measurements. The main characteristics of the national standards maintained in Italy at the ENEA-INMRI in the field of radionuclide measurements are reported in the following table.

National standards maintained at the ENEA-INMRI (Italy) in the field of radionuclide measurements

Quantity	Standard	Radionuclide	Uncertainty Range (^) (%)	Measurement Range
Activity (+)	-n. 2 4πβ-γ coincidence counting systems	β and β-γ emitters	0.1 - 3	(1 - 20) kBq
	-n. 1 NaI(Tl) well-type sum-peak coincidence counting system	γ–γ emitters	0.5 - 3	(1 - 20) kBq
	-n. 1 NaI(Tl) well-type 4πγ counting system	γ emitters	0.5 - 3	(1 - 20) kBq
	-n. 1 LS (CIEMAT/NIST) counting system	β and x-ray emitters	0.6 - 3	(1 - 20) kBq
	-n. 1 Rn-in-water generator	^{222}Rn-in-water	2	(200 – 10^4) Bq/dm^3
	-n. 1 Electrostatic cell	^{222}Rn-in-air	1	(1 - 15) kBq
	-n. 1 Well-type ionisation chamber*	γ emitters	0.2 - 3	(10 - 2 10^4) kBq
	-n. 3 HPGe γ-ray spectrometers*	x and γ emitters	1 - 5	(1 - 10^5) Bq
Activity concentration	-n. 1 0.1 m^3 radon chamber*	^{222}Rn-in-air	2 - 10	(10^2 - 10^4) Bq/m^3
Surface emission rate	-n. 1 2π windowless gas flow proportional counter	α and β emitters	0.5 - 3	(1 - 20) s^{-1}

(^) Rounded values for standard combined uncertainties (1σ).
(*) High precision secondary standards.
(+) Issue of radioactivity standards: Standard radioactive sources are supplied in different geometries in the activity concentration range from 10^{-2} Bq g^{-1} to 2 10^6 Bq g^{-1} (aqueous solutions in standard ampoule or in flask of different size) and in the activity range from 1 Bq to 2 10^7 Bq (sources in Marinelli beaker, in ampoule, on paper filter and point sources).

The ENEA-INMRI programmes in the field of radionuclide metrology in the last two years (2005-2007) were focused, as in the past, on maintaining and developing the national standards for activity measurements and on the more general activities in the field of standardisation and quality-assurance in radioactivity measurements.

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Extensive restructure works in the radioactivity measurements rooms, as requested by the control authority for radiological protection, lasted about all the year. In this period, the research and calibration activity in this field was strongly reduced.

The main specific activities carried out at ENEA-INMRI in field of radionuclide metrology are summarised below.

2. DEVELOPMENT OF NATIONAL STANDARDS AND COMPARISONS

2.a Standardisation of ^{125}I by sum-peak coincidence method

A new measurement standard, based on the sum-peak coincidence counting method, was developed in 2005 for absolute standardisation of a solution of ^{125}I in the frame of an international comparison organised by the BIPM. The mother solution was initially checked for radionuclide purity. A number of sources for absolute measurements were then prepared in flame sealed glass ampoules containing 1 cm^3 of inactive diluent and increasing masses of ^{125}I mother solution (from 3.5 to 38 mg). These sources were counted in two experimental systems based on different high efficiency NaI(Tl) well-type detector (respectively 550 cm^3 and 1600 cm^3). Usual corrections for background and decay were applied. Single- (full-energy) and sum-peak superimposition was corrected by a linear extrapolation of the peak tails. Random summing and dead time effects were corrected by linear extrapolation to zero count rate (zero mass). This correction was drastically reduced thanks to the use of high performance digital multiparametric acquisition system (Fast Comtec PMA3), recently installed at ENEA-INMRI. The relative combined standard uncertainty of the ENEA-INMRI results was 0.3%. The result obtained by the ENEA-INMRI is in very good agreement with the arithmetic average of the comparison.

2.b Standardisation of ^{64}Cu by 4πβ liquid scintillation efficiency-tracing method

The 4πβ Liquid Scintillation Spectrometry Method with ^{3}H-Standard Efficiency Tracing, in literature known as CIEMAT/NIST method, has been applied to measure the activity concentration of a ^{64}Cu solution. The particular ^{64}Cu decay scheme (both presence of β^- and β^+ emissions) and the relatively long half-life (compared with the ^{18}F, a nuclide largely used in Nuclear Medicine) make this isotope suitable for both imaging and positron emission tomography (PET) and cancer therapy. The measurements of the ^{64}Cu activity has been performed at the EC Joint Research Centre (EC-JRC) of Ispra under a scientific collaboration between the ENEA-INMRI and the Institute for Health and Consumer Production. The isotope has been produced by the Scanditronix MC40 Cyclotron of the EC-JRC and standardised at the production site. The new national standard has been used to calibrate the ENEA-INMRI portable well-type ionisation chamber (IC), used as Secondary Standard Measurement System (*SSMS*) and then easily transportable in Nuclear Medicine centres or in the other production sites to calibrate local instrumentation by a simpler comparison. The identification of pure β-impurities in the ^{64}Cu solution, as ^{61}Cu and ^{65}Zn, requested a particular effort. The analysis of the collected experimental data is in progress.

2.c Standardisation of ^{188}Re by 4πβ liquid scintillation efficiency-tracing method

A scientific collaboration between ENEA-INMRI, FIS ION (TRIGA Reactor) and the Nuclear Medicine department of S. Eugenio Hospital (Rome) was established to develop a ^{188}Re national primary standard and suitable calibration procedures for ^{188}Re measurement instrumentation . This isotope has a radiochemical behaviour which is similar to that of the well known diagnostic radionuclide ^{99m}Tc. A wide application of the ^{188}Re in therapeutic nuclear medicine can be foreseen. The isotope is eluted from a ^{188}W/^{188}Re alumina column, by using 5-20 ml of physiologic solution and currently used in the Nuclear Medicine Dept. of S. Eugenio Hospital for the preparation of ^{188}Re sulphide, ^{188}Re-labelled tin colloid, ^{188}Re-labelled antimony sulphide colloid in micro and nanoparticle form, for injective and non-injective brachytherapy, ^{188}Re-DMSA(V), ^{188}Re-DTPA. ^{188}Re-EDTA.

The CIEMAT/NIST method has been applied for the measurement of the activity concentration of a ^{188}Re solution. The ENEA-INMRI portable well-type Ionization Chamber (IC) then will be calibrated and used as a Secondary Standard Measurement System (*SSMS*). The data analysis is in progress; the preliminary results show the presence in the ^{188}Re solution of a little but non negligible impurity. This means that both a radioactivity purity check of the mother radioactive solution by high resolution gamma-ray spectrometry and a measurement of the activity by an other absolute method, like the 4πβ–γ coincidence method in the efficiency tracing variant, are necessary. A calibration test with uncertainty lower than 2% has been performed for the IC of S. Eugenio Hospital in Rome by using the preliminary results of the ^{188}Re activity absolute measurement.

2.d Participation in the BIPM intercomparison on ^{55}Fe activity measurements

The ^{55}Fe isotope is a pure electron-capturing nuclide, emitting X-rays and Auger electrons of very low energy (0,6 to 6,5 keV). The photons emitted by ^{55}Fe are close in energy to the low energy threshold of the many radiation measurement equipments such as surface contamination monitors, so a check using a ^{55}Fe source is a sensitive method to identify any faults with these equipments (eg., drifts in the high voltage applied to the detector). For these reasons it is important to have a ^{55}Fe standard, although the ^{55}Fe standardisation is very difficult because of the particular decay scheme of this isotope. At the ENEA-INMRI a ^{55}Fe BIPM solution have been standardised by applying the CIEMAT-NIST method. A set of ^{55}Fe sources for scintillation counting have been prepared in 20-ml low-potassium-glass vials, with 10 ml of ULTIMA GOLD liquid scintillator. Quenching variation have been carried out by adding different aliquots (from 0 to 100 μl) of quenching agent CCl_4 (corresponding to a value interval for the experimental quench parameter, tSIE, ranging from 700 to 200). The experimental data have been corrected for: dead-time, background, impurity and decay. The code Minerva (P. De Felice et al., "Standardisation of ^{90}Sr, ^{63}Ni and ^{55}Fe by the 4πβ liquid scintillation spectrometry method with ^{3}H-standard efficiency tracing", ARI **51,** 1999, 85-92) has been used to compute the counting efficiency. An unexpected deviation of the ENEA-INMRI results among the average value of the comparison has been communicated by the BIPM and it is under investigation.

2.e Primary standard of ^{222}Rn

The ENEA-INMRI primary standard for radon-in-air measurements, in operation since 1995, is based on the extraction of radon from a ^{226}Ra standard solution that is transferred into a

closed known volume circuit. An additional radon reference monitor, based on a Genitron AlphaGuard, is installed in the system gas circuit and directly calibrated by the primary measurement system. The AlphaGuard monitor, thanks to its notable stability of response and low humidity dependence, allowed recalibration of the radon reference atmosphere with lower uncertainty.

2.f Reference Climate Radon Chamber

The ENEA-INMRI 1 m^3 climate radon chamber allows fully automated conditioning cycles. The chamber was improved to obtain variable environmental conditions for calibrating radon and radon progeny measuring instruments. A new control and data acquisition equipment has been developed to automate the multitasking management of different sets of radon monitors and climate sensors.
The new monitoring apparatus (named Radotron) has been set up and it is currently under test. Radotron is a multifunction system that provides control functions with the capability to correlate data from different radon monitors and climate sensors.
The uniformity and stability of radon concentration during a calibration run have been accurately assessed. The radon concentration was enough uniform so that the time integrated concentration between the maximally and minimally exposed monitors differs by less than 5%.

2.g The alpha track detectors system (ATDs)

A new measurements apparatus based on a set of passive integrating monitors was developed at the ENEA-INMRI. The monitors are based on a new concept of passive time-integrating alpha track detector (ATD) fitted with a mobile wall of a closed cylindrical conductive cup.
This device, called Piston Radon Dosimeter (PRD), produces the syringe effect and the on-off switching of detectors exposure. The major features of the new ENEA PRD are fast sampling, negligible post-exposure effect and control of exposure interval.
Prototypes have been made to be fitted on the three coupling flanges of the door of the exposure facility to standardize the reference atmosphere of the ENEA-INMRI climate radon chamber

3. QA NATIONAL PROGRAMME AND CALIBRATION ACTIVITY

3.a Development of a ^{64}Cu transfer standard

A new portable well-type ionisation chamber was calibrated with uncertainty lower than 2% by the ENEA-INMRI, using the newly developed ^{64}Cu primary standard. This new chamber can be used as a transfer secondary standard measurement system. It can be transported in Hospitals or in ^{64}Cu production centres where the local instrumentation can be calibrated by comparison.

3.b Calibration of radioactive sources

Calibration of radioactive sources by spectrometric methods was strongly reduced for extensive restructure works in the radioactivity measurements rooms. New calibration service for radon measurements has been introduced due to the increasing demand arising from a

laboratories in the Country in the field of radon-in-air and radon-in-water measurements. The calibration facilities for measurements of radon-in-water are temporarily not available for the above reasons.

3.c Calibration of radionuclide activity measurement instruments

The measurement instrument calibration service was strongly reduced due to the same reason explained above. Nevertheless :

1. an extended calibration was made for the ^{99m}Tc measurement instruments of the children's Hospital "Bambin Gesù" in Rome
2. the ENEA-INMRI radon chamber of respectively 137, 216 and 1027 litres have been used for calibrating passive radon dosimeters and radon active monitors.
3. about 20 surface contamination monitors were calibrated according to ISO standards in the 2005-2007 period by using mainly ^{241}Am, ^{90}Sr and ^{14}C sources.

3.d Preliminary Test for calibrating ^{18}F measuring systems

Due to the [^{18}F]FDG increasing demand new production sites started in Italy during the last two years. A growing request for calibration of ^{18}F measuring system (radionuclide calibrators) is coming from the different production centres and the Nuclear Medicine Department in our country. The Secondary Standard Measurement System (SSMS) was then used for calibrating many radionuclide calibrators used in Nuclear Medicine Departments. The preliminary data of this test show a good agreement of the results with the request of the European Farmacopea for the ^{18}F radioisotope, particular applied for the PET diagnostic.

3.e Quality assurance programme for the national radioactivity surveillance network

The Italian radioactivity surveillance network is made of about 50 laboratories located over the national territory and coordinated by the national agency for environmental protection (APAT). A national intercomparison of passive radon detectors was carried out in cooperation with the APAT. 26 laboratories sent to the ENEA-INMRI about 1500 radon passive dosimeters based on nuclear track or electret detectors. The dosimeters were exposed in the ENEA-INMRI climate radon chamber of 1m^3 at four different exposure level (measured in kBq h m^{-3} units). The dosimeters readings coming from the participating laboratories have been compared with the reference exposure values. An average deviation lower than 10% has been observed between the measurements and the reference value for the 4 exposure level of 1938, 1217, 987 and 217 kBq h m^{-3}.

4. PARTICIPATION IN METROLOGICAL AND STANDARDISATION ORGANISATIONS

Part of the time was devoted to activity in metrological and standardisation organisations: ICRM, BIPM/CCRI-II, IEC/TC45, ISO/TC85/SC2, UNI (National Standardisation Organisation). P. De Felice has continued his office as Secretary of the International Committee of Radionuclide Metrology.

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(1) Due to the shortage of personnel some technicians share their work (e.g., mechanical workshop) among the different sections of the Institute.

(2) Administrative service and technical assistance for maintaining and repair are supplied by the CR Casaccia central service. Some activities at the ENEA-INMRI in the period 2005-2007 have been carried out with the collaboration of some students.

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