Report to the CCRI Section II on the activity carried out at the ENEA-INMRI on radionuclide measurements in the period 1999-2001

P. De Felice
Istituto Nazionale di Metrologia delle Radiazioni Ionizzanti
ENEA, C. R. Casaccia
c.p. 2400, 00100 AD Roma (Italy)

1. INTRODUCTION

The present report summarizes the 1999-2001 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 measurement are reported in the following table.

National standards maintained at the Enea-Inmri (Italy) in the field of radionuclide measurements

Quantity	Standard	Radionuclide	Accuracy Range (%)	Measurement Range
Activity (+)	-n. 2 $4\pi\beta$(PC)-γ coincidence counting system	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 $4\pi\gamma$ 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 High press. ion chamber*	γ emitters	0.2 - 3	(10 - 2 10^4) kBq
	-n. 3 HPGe γ-ray spectrometers*	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

N.B. 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 MBq g^{-1} (aqueous solutions in standard ampoule or in flask of different size) and in the activity range from 1 Bq to 20 MBq (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 (1999-2000) 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. The main specific activities carried out at ENEA-INMRI in this field are summarised below.

2. STANDARDS DEVELOPMENT AND COMPARISONS

2.1 Development of a new radon-in-water standard

A project to develop a national standard for radon-in-water measurements was initiated in Italy by the growing demand of calibration arising from a number of laboratories in the Country. The new standard, developed in collaboration with the CMI (CZ), is based on a radon-in-water generator and a measurement system.
The radon-in-water generator is used to prepare a water solution with an amount of homogeneously dissolved radon. The generator is based on a solid radon emanator, with high emanation power (99.9%), made of a ^{226}Ra solid source. The radium source is immersed in a water bath sealed in a glass flask. A radon- and air-tight circuit is connected to this flask to allow full filling of the flask and homogenisation of the solution. A particular procedure was also developed for quantitative transfer of the radon-in-water solution from the generator into appropriate sample holders, keeping radon losses at negligible levels. The radon-in-water generator was designed to allow the preparation of up to 4 l of solution with ^{222}Rn activity concentration from 200 to 10000 Bq/l and with negligible radium impurities.
The measurement system is needed for reproducibility and stability checking and for calibration of the radon-in-water generator. Radon-in-water samples taken from the generator were measured by gamma-ray spectrometry and liquid scintillation counting. The type of liquid scintillator, the chemical composition of the solutions, the source geometries and the measurement procedures were adapted to the peculiar chemical-physical properties of radon and its decay products.
Calibration of the measurement system was carried out by a ^{226}Ra standard solution traceable to the radium standard maintained by ENEA-INMRI. Preliminary results showed that both measurement methods have reproducibility levels better than 1%. The relative combined standard uncertainty of the activity concentration of the radon-in-water solution resulted to be better than 2%. A bilateral comparison is in course, between ENEA and CMI on their radon-in-water standards.

2.2 Development of a $4\pi\gamma$ well-type NaI(Tl) counting system

A new standard was developed, based on the $4\pi\gamma$ high efficiency measuring method, for absolute measurement of activity of radionuclides with complex decay scheme. To this purpose a NaI(Tl) well-type crystal was obtained with accurately measured geometrical parameters (uncertainty of 0.1 mm): crystal diameter= 124.9 mm, crystal height= 125.0 mm, well diameter 32.0 mm, well depth=69.8 mm. An integral discrimination threshold was set at 22.1 keV (x-ray emission from a ^{109}Cd point source). The single-photon detection efficiency curve was generated by Geant 3.21 Monte Carlo code. The counting efficiency for several complex decay scheme nuclides were calculated by the NaJ5EF program (by dr. G. Winkler). The new standard was compared for the first time in the frame of the ongoing ^{152}Eu BIPM full-scale comparison.

2.3 Participation in the BIPM full-scale intercomparison on ^{152}Eu activity measurements

In this comparison the $4\pi\gamma$ high efficiency measuring method was used at ENEA-INMRI. A few details on the experimental equipment are reported above. The main corrections taken into account were: dead-time, background, impurity (^{154}Eu), adsorbtion and decay. The ^{154}Eu activity was determined by HPGe γ-ray spectrometry. The combined standard uncertainty of the ^{152}Eu activity concentration was 0.31% with the main contributions due to determination of the detection efficiency curve (0.2%) and knowledge of the decay scheme parameters (0.2%). Unfortunately, the measuring method used in this comparison was introduced very recently at ENEA-INMRI and it was not possible to test it before the comparison. A critical examination of the ENEA-INMRI result will then allow to identify and correct for possible sources of errors.

2.4 Participation in the BIPM full-scale intercomparison on ^{89}Sr activity measurements

The CIEMAT-NIST liquid scintillation efficiency-tracing method was used at ENEA-INMRI to measure the activity concentration of the ^{89}Sr solution. The computer program EFFY-4 was used with ^{3}H as tracer nuclide to obtain the counting efficiency of the liquid scintillation system. The main corrections taken into account were: dead-time, background, impurity (^{85}Sr) and decay. The ^{85}Sr activity was determined by HPGe γ-ray spectrometry. After a specific investigation, no ^{90}Sr impurity was detected in the solution. The relative combined standard uncertainty of the ENEA result was 0.57% with the main contributions due to quenching correction (0.3%), nuclear data (0.3%), half life (0.2%) and ionisation quench (0.2%). The results of the intercomparison are under evaluation by the BIPM.

2.5 Bilateral comparison with CMI (CZ) on ^{226}Ra activity measurements

A bilateral comparison on ^{226}Ra activity measurement was carried out between ENEA-INMRI and CMI (CZ). It was thought that such a comparison should be carried out before performing the in-course comparison on radon-in-water standards. This is because of the important role played by the ^{226}Ra standards in the traceability chains for radon metrology developed by the two Institutes. The result of the comparison was very encouraging. The relative deviation was well within the combined standard uncertainty reported by the two institutes (about 0.5%).

2.6 Bilateral comparison with OFMET-IRA (CH) on activity measurement of a mixture on gamma-ray emitting radionuclides

A bilateral comparison was carried out by ENEA-INMRI and IRA-OFMET (CH) on activity concentration measurement of a liquid solution spiked with a mixture of gamma emitters (^{109}Cd, ^{57}Co, ^{139}Ce, ^{137}Cs, ^{88}Y, ^{60}Co). This is a typical mixture utilised by the two laboratories in the QA national programmes on radioactivity measurements organised in their respective Countries. The result of the comparison was satisfactory, being the deviations of the two results within the combination of the individual uncertainties (0.5-3%).

3. QA NATIONAL PROGRAMME AND CALIBRATION ACTIVITY

3.1 A new calibration campaign for the radioactivity surveillance network

The main Quality Assurance program conducted by ENEA-INMRI in the field of radioactivity measurements regarded the national network for environmental radioactivity surveillance. The programme is based on periodical calibration and intercomparison campaigns carried out by ENEA-INMRI under request of the National Agency for Environmental Protection. This programme started more than 15 years ago. Beta counting and γ-ray spectrometry in environmental samples are the main objects of the programme. This QA programme was effective in reducing to about 10% the maximum deviation of the results among the network laboratories. A new national calibration campaign was carried out for γ-ray spectrometry measurements. To check procedures for self-absorbtion corrections, a solution with density (1.4 g/cm^3) and chemical composition different by water was used.

3.2 A national campaign for application of coincidence-summing corrections in γ-ray spectrometry

A national campaign was organised by ENEA-INMRI for application of coincidence-summing correction in γ-ray spectrometry. About ten laboratories, belonging to the national environmental radioactivity network, took part in the campaign. These laboratories are equipped with γ-ray spectrometry systems based on p- and n- types HPGe detectors with relative efficiency values ranging from 15% to 80%. All the laboratories regularly take part in the QA programme carried out for the network. In all of these comparisons it was clear that the major source of errors in laboratory measurements was due to true summing effects. The magnitude of the errors varied from a few percent up to 30% with high efficiency detectors. It was then decided to perform a national exercise to introduce coincidence-summing corrections among the network laboratories. A fast and simplified correction procedure was obviously needed. It was selected among a set of three different procedures developed and tested by ENEA-INMRI in previous years. The selected procedure is based on the usual theoretical expressions of the correction factors with the total efficiency curve determined by the assumption of a constant relation between the peak-to-total efficiency ratios and the photoelectric-to-total cross-section ratios. This procedure require a single-photon, single-nuclide source to measure the above ratio at one photon energy value. The counting geometry of 1 L Marinelli beaker filled with aqueous solution was chosen.
All the laboratories were given a ^{137}Cs standard source, for total efficiency determination, and a ^{134}Cs source in the same geometry whose activity they had to determine. The average deviation of the laboratory results from the ENEA-INMRI reference value changed from 10-15% to a few percent after correction. The results of the campaign will be presented at the incoming ICRM2001 Conference.

3.3 Radionuclide activity calibration service

Radioactive standards (liquid solutions, point sources, gas sources, paper filters and spiked reference materials for a total of about 100 sources per year) were provided for calibration of radioactivity measurement instruments in the country and for organisation of bilateral comparisons for QA.

3.4 Surface contamination monitor calibration service

Calibration of surface contamination monitors are performed according to ISO standards. In the 1999-2000 period about 50 instruments were calibrated mainly with ^{241}Am, ^{90}Sr, and ^{14}C sources.

4) ACTIVITY IN METROLOGICAL AND STANDARDS ORGANISATIONS

Part of the time was devoted to activity in metrological and standardisation organisations: ICRM, BIPM/CCRI-II (observer), IEC/TC45, ISO/TC85/SC2, UNI (National Standardisation Organisation). P. De Felice was member of the Scientific Committee of the ICRM Low-Level Radioactivity Measurement Techniques Conference, Mol, October 1999.

5. STAFF AT THE ENEA-INMRI INVOLVED IN ACTIVITIES RELATED TO RADIONUCLIDE MEASUREMENTS

Istituto Nazionale di Metrologia delle Radiazioni Ionizzanti
ENEA, C. R. Casaccia
c.p. 2400, 00100 AD Roma (Italy)

Director, R. F. Laitano

Fax: ++39 06 3048 3558
Phone: ++39 06 3048 (EXTENSION)
E-mail: username@casaccia.enea.it

STAFF (3)	**E-mail username**	**Phone extension**
Scientists		
Dr. P. De Felice	defelice	3580
Dr. M. Capogni(1)	marco.capogni	6628
Technicians		
Mr. A. Fazio	fazio	3557
Mr. R. Biagini(2)	biagini	3354
Mr. A. Manzotti (50%)(4)	manzotti	4563
Mr. L. Ceravolo	ceravolo	3556
Mr. M. Quini (20%)(4)	quini	6626

(1) Since April 2000.
(2) Retired in April 2001.
(3) Some activities at the ENEA-INMRI in the period 1999-2001 have been carried out with the collaboration of guests (3 fellowships) and students (2 student).
(4) Due to the shortage of personnel some technicians share their work (e.g., mechanical workshop) among the different sections of the Institute.
(5) Administrative service and technical assistance for maintaining and repair are supplied by the CR Casaccia central service.

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