**Report to the CCRI Section II on the activity carried out at the ENEA-INMRI on radionuclide measurements in the period 2001-2003**

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## 1. INTRODUCTION

The present report summarizes the 2001-2003 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 | Uncertainty Range (^) (%) | Measurement Range |
|---|---|---|---|---|
| Activity (+) | -n. 2 $4\pi\beta$-$\gamma$ coincidence counting systems | $\beta$ and $\beta$-$\gamma$ emitters | 0.1 - 3 | (1 - 20) kBq |
| | -n. 1 NaI(Tl) well-type sum-peak coincidence counting system | $\gamma$–$\gamma$ emitters | 0.5 - 3 | (1 - 20) kBq |
| | -n. 1 NaI(Tl) well-type $4\pi\gamma$ counting system | $\gamma$ emitters | 0.5 - 3 | (1 - 20) kBq |
| | -n. 1 LS (CIEMAT/NIST) counting system | $\beta$ 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* | $\gamma$ emitters | 0.2 - 3 | $(10 - 2\ 10^4)$ kBq |
| | -n. 3 HPGe $\gamma$-ray spectrometers* | x and $\gamma$ 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 $2\pi$ windowless gas flow proportional counter | $\alpha$ and $\beta$ emitters | 0.5 - 3 | (1 - 20) s$^{-1}$ |

(^) Rounded values for standard combined uncertainties ($1\sigma$).
(*) 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 (2001-2003) 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. DEVELOPMENT OF NATIONAL STANDARDS AND COMPARISONS

Standardisation of $^{152}$Eu and $^{134}$Cs by 4πγ counting method

A new measurement standard, based on the 4πγ high efficiency counting method, was developed in 2001 for absolute standardisation of radionuclides with complex decay scheme. A NaI(Tl) well-type crystal (125 cm x 125 cm) with accurately known geometrical parameters (uncertainty of 0.1 mm) was used. The new standardisation method was firstly applied at ENEA during the $^{152}$Eu BIPM comparison. The single-photon detection efficiency curve, generated by Geant 3.21 Monte Carlo code, was used for calculating the specific nuclide counting efficiency by Winkler's NaJ5EF Fortran program. The ENEA result (570.8+-1.8 kBq/g) was 1.9% lower than the arithmetic mean of the comparison results (582.0 +- 1.4 kBq/g). An extensive re-analysis was then started to find out the explanation of this deviation and improve the measurement accuracy. A trivial mistake was identified indeed in the use of the NAJ5EF Fortran program, in particular the program's input data sections. The corrected $^{152}$Eu activity concentration was then 582.1+/-1.8 kBqg$^{-1}$, in very good agreement with the arithmetic average of the comparison. This last conclusion was communicated to the BIPM as comment to the ENEA result.

Recently, a $^{134}$Cs solution has been standardised by the same method. The resulting combined standard uncertainty is lower than 0.5%. It is intended to submit this standardised $^{134}$Cs solution to the BIPM SIR.

Participation in the BIPM intercomparison on $^{32}$P and $^{204}$Tl activity measurements

The CIEMAT-NIST liquid scintillation efficiency-tracing method was used at ENEA-INMRI to measure the activity concentration of the $^{32}$P and $^{204}$Tl BIPM solutions. The computer programs EFFY-4 were used with $^{3}$H as tracer nuclide. The main corrections applied were: dead-time, background, impurity and decay. A particular effort was required for impurity determination in the $^{32}$P solution. Two pure beta emitters were indeed detected ($^{33}$P and $^{35}$S) the activity concentrations of which were obtained by careful statistical analysis of integral counts repeated over a period of two months. The relative combined standard uncertainties of the ENEA results were 0.61% and 0.49%, respectively for $^{32}$P and $^{204}$Tl. The results of the intercomparison are under evaluation by the BIPM.

Standardisation of $^{65}$Zn and participation in the BIPM intercomparison

In view of the incoming BIPM intercomparison, a new $^{65}$Zn standardisation was started at ENEA in 2001 in cooperation with colleagues of the National Institute of Physics and Nuclear Engineering (IFIN) in Bucharest, Romania.

The 4πPC-γ efficiency extrapolation method was used with different measurement conditions for the beta channel: detection of the $e_K/\beta^+$ or $(e_K+e_L)/\beta^+$ events. Gamma windows and photon lead absorbers were used for accomplishment of the linearity condition: $\varepsilon_{\gamma 511} = 0.25\ \varepsilon_{\gamma 1115}$. Foil extrapolation was performed. The combined standard uncertainty was 0.6%. The activity concentration of the solution was also in good agreement with ionisation chamber measurements traceable to PTB, OMH and previous ENEA standards (based on the CIEMAT/NIST method). The results of the campaign will be presented at the incoming ICRM2003 Conference. The BIPM solution for the in course intercomparison was measured in 2003 by the ionisation chamber (by using the last calibration entry) and the result was communicated to the BIPM.

Bilateral comparison with CMI (CZ) on $^{222}$Rn in water activity measurements

A project for development of a new national standard for radon-in-water measurements was terminated in 2001. The new standard is based on a radon-in-water generator, used to prepare a water solution with an amount of homogeneously dissolved radon, and on measurement systems based on liquid scintillation counting and on gamma-ray spectrometry. Calibration of both measurement systems was carried out by a $^{226}$Ra standard solution traceable to the radium standard maintained by ENEA-INMRI. The relative combined standard uncertainty of the activity concentration of the radon-in-water solution was better than 2%.

The project was terminated with an intercomparison with the CMI (CZ). A number of ready-to-measure liquid scintillation vials with radon-in-water solution were prepared, exchanged and measured by the two laboratories. Results were well within the stated uncertainties.

## 3. DEVELOPMENT OF RADIOACTIVITY MEASUREMENT METHODS

New detectors for radon measurements

Investigations on new detectors for radon measurement in geophysical field were carried out in cooperation with the Rome University “Roma3”. The scintillation properties of a Ce-doped yttrium aluminium oxide perovskite monocrystals (YAP) optically coupled to a Hamamatsu H5784 photomultiplier with standard bialkali photocatode have been studied in particular for radon and radon daughters gamma-ray spectrometry. Tests in water up to a temperature of 100 °C and in HCl (37%), $H_2SO_4$ (48%) and $HNO_3$ (65%) acids solutions have been performed to simulate environments of geophysical interest such as geothermal and volcanic areas. Measurements with standard radon sources emphasized the non-hygroscopic properties of the crystal and a small dependence of the light yield on temperature and $HNO_3$ treatment. It was then found that YAP crystals can allow high response stability for radon gamma-ray spectrometry in environments with large temperature gradients and high acid concentrations.

## 4. QA NATIONAL PROGRAMME AND CALIBRATION ACTIVITY

Intercomparison on measurements of natural radionuclides in building materials

The role of building material in the exposure of population due to gamma radiation and radon indoors has been shown in the past in several Countries. The European Commission recently published a guide aiming at the limitation of the activity concentration in building material and to the harmonisation of the radiation protection approach to the problem. However, gamma-ray spectrometric determination of natural radionuclides in building material is particularly complex due to spectral interference, choice of nuclear data, radioactive equilibrium condition, coincidence-summing and self-absorption effects. To study these aspects a group of European Institutes working in the field of gamma spectrometry organised a preliminary intercomparison of gamma-ray spectrometric determination of natural radionuclides in environmental samples, such as building materials. The exercise was carried out using samples of ash, sand and tuff. The participating laboratories were: the National Technical University of Athens (NTUA) in Greece, the National Institute of Health (ISS) in Italy, the Radiation and Nuclear Safety Authority (STUK) in Finland and the Kernfysisch Versneller Instituut (KVI) in the Netherlands. The results from all collaborating laboratories were collected and analysed by the ENEA-INMRI also participating in the intercomparison.

The results of this exercise were presented in the Natural Radiation Environment-VII Symposium in Greece. The experience gained may be used for suggesting future exercises with wider participation and/or the drafting of an international protocol for standardisation of these measurements.

Calibration service: radon-in-air and radon-in-water

New calibration services were introduced in 2001 for the growing demand arising from a number of laboratories in the Country in the field of radon measurements. The ENEA radon chamber and the radon-in-water standard are used for this purpose.

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.

Surface contamination monitor calibration service

Calibration of surface contamination monitors are performed according to ISO standards. In the 2001-2003 period about 50 instruments were calibrated mainly with $^{241}Am$, $^{90}Sr$, and $^{14}C$ sources.

## 5. 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). Since 2001 P. De Felice was elected member of the ICRM Executive Board as Secretary.

## 6. STAFF

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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 2001-2003 have been carried out with the collaboration of guests (3 fellowships) and students (2 students).

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