

LNE-LNHB progress report related to Radionuclide Metrology (2007-2009)

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1. News

In order to comply with the legislation related to the use and holding of radionuclides at LNHB, a long period of renovation works started in June 2007, ended in April 2009; the different renovation operations were managed in order to maintain the main activities of the laboratory during this period. A new nuclear ventilation is now operational in relocated areas specifically dedicated to source preparation and to radionuclide measurements; the control of ambient conditions has been improved (temperature, hygrometry).

The renovation of the laboratory permits the strengthening of our industrial partnerships: nuclear industries, radiopharmaceutical and environmental fields.

In particular, the development of a laboratory dedicated to the preparation of low level environmental matrices, is underway (vegetables, soils, etc.); this laboratory will include equipments such as pulverizers or a microwave oven. The project corresponds to the demand of the national network for environmental measurements piloted by ASN (Authority for Nuclear Safety) and IRSN (Institute of Radioprotection and Nuclear Safety) to organize proficiency inter-laboratory tests; the objective replies to the need to establish a metrological traceability to national references in the field of environmental measurements of nuclear installations.

2. Research & Development

2.1 Improved energy resolution for x-ray and gamma spectrometry with a metallic magnetic calorimeter

In our development of cryogenic detectors (metallic magnetic calorimeters) for high energy resolution x-ray and γ -spectrometry, we have achieved a substantial progress in energy resolution. The detector consists of a gold photon absorber, a cylinder of 1.1 mm diameter and 350 μ m thickness, thermally coupled to a paramagnetic AuEr temperature sensor. The sensor is magnetized and read out by a superconducting niobium thin film structure in the shape of a meander, coupled to a SQUID magnetometer. Despite its small thickness, the gold absorber offers a detection efficiency larger than 70 % for photon energies between 3 keV and 100 keV.

Previously, the energy resolution was 320 – 350 eV (FWHM) from 3 keV to 160 keV. It was limited by a maximum magnetic field (required for sensor magnetization) much below its optimal value, and additionally by an excess noise of the SQUID based readout system. By modifications of the wiring of the dilution refrigerator providing the detector operating temperature (\sim 15 mK) as well as by an improved shielding of the experimental setup against electromagnetic interference, both of these limiting factors could be significantly improved. By now, the energy resolution is 51 eV at 6 keV and 58 eV at 80 keV, respectively. We believe that by a further enhanced magnetic field and a better magnetic coupling between the AuEr sensor and the meander readout structure the energy resolution can be brought down to \sim 25 eV.

2.2 Development and qualification of a portable LS counter with a Compton spectrometer

A portable LS counter using 6 channel photomultipliers, 1 miniature photomultiplier and a CdTe gamma-ray detector is currently under test. The system allows the measurement for two preset values of detection efficiency and using the Compton efficiency tracing technique previously described. The acquisition system uses a commercial FPGA module under the control of Labview software.

2.3 Development of new liquid scintillators using fluorescent nanoparticles

The goal of this study is to replace the traditional LS medium by a suspension in aqueous medium of fluorescent inorganic nanoparticles with intrinsic high light yields. Several different particles were evaluated: semiconductors core-shell quantum dots and fluorescent oxides nanoparticles. Encouraging results were already obtained with Ga doped LSO nanoparticles. The study is now focusing on the surface functionalization of the nanoparticles in order to increase their concentration in the solution.

2.4 Implementation of digital platform for primary measurements

Due to the growing difficulties to ensure the maintenance and the acquisition of classical analog devices dedicated to primary measurements, the transition to digital systems is in progress at LNHB. The objective is to investigate the capabilities of logical programmable devices for radionuclide standardization instrumentation. For that purpose, two projects are underway; in both cases, the implementation of the counting processing is governed by the long-term experience acquired with home-made modules based on the live-time technique using extendable dead-times.

The first development started in 2007; it concerns the instrumentation dedicated to the $4\pi\gamma$ method using a NaI(Tl) well-type detector. The acquisition card is composed of a PCI mother-board placed inside a PC and fitted with 2 modules. Basically, the first module consists in an analog-to-digital converter (ADC, 200 MHz, 12 bits) followed by a FPGA component for fast data processing (triggering, amplitude and timing measurements). The second module holds a DSP that controls the data transfer between the mother-board and the PC. The quantities of interest such as signal amplitudes and time intervals are stored in the PC; the counting, histogramming and the dead-time processing are implemented off-line using a simple computer algorithm. A first comparison with the classical NIM-based instrumentation has been carried out for counting rates up to $100 \text{ kc}\cdot\text{s}^{-1}$ using a ^{137}Cs source.

In the second development in progress started in 2008, the final objective is to obtain a digital platform dedicated to $4\beta\text{-}\gamma$ anticoincidence counting which can be feed by usual detectors; it has also been added to the specifications the possibility of performing TDCR measurements (Triple to Double Coincidence Ratio) when a liquid scintillation counter is used in the β -channel. The project is based on an on-line processing of the counting according the live-time technique extendable dead-times. In addition, the aim is to incorporate the different functionalities of the surrounding associated with traditional equipment: scalers, preset counters, multi-channel analyzers, time-to-amplitude converters, etc. In order to validate the on-line counting processing implemented in a FPGA-based digital platform, the liquid scintillation counting has been first programmed to enable TDCR measurements. Therefore, the digital system can be considered as an improved MAC3 module with extended possibilities: a variable resolving time for double and triple coincidences is implemented and time intervals between dead-time periods are registered in histograms to give information on the timing of after-pulses.

2.5 Modeling of a TDCR liquid scintillation counter using the Monte Carlo code GEANT4

The modelling of the TDCR counter used in the β -channel of a $4\pi(\text{LS})\beta\text{-}\gamma$ anticoincidence system; the stochastic calculation of coincidences between photomultipliers is implemented using the Monte Carlo code GEANT4. The objective is to simulate the different modes of light emission in a TDCR counter. The first results confirm the hypothesis of Cherenkov photons emitted in the photomultiplier widows induced by Compton diffusion.

3. Radionuclide Measurements and Evaluation of Nuclear Decay Data

- Participations to international comparisons: Eu-152, Co-57, Rn-222, Sb-124, Tc-99m, Se-75, U-233, Kr-85.
- IAEA-CU-2007-06-CCRI(II)-S5: Supplementary comparison for the determination of technically enhanced naturally occurring radionuclides (TENORM) in phosphogypsum.
- **Sb-124:** The Euromet 907 comparison coordinated by LNE-LNHB, is completed (8 laboratories participated); it has been proposed as a BIPM supplementary comparison (EUROMET.RI(II)-S5.Sb-124). A measurement questionnaire has been filled by each participant and received at LNHB by March 2008; a first draft has been reviewed by the participants and the final version is under preparation.
- **Sn/Sb-126:** This nuclide was provided by CEA Cadarache; the mass concentration of the sample was carried out by CEA Cadarache and the activity of Sn/Sb-126 was determined at LNHB by means of γ -spectrometry measurements. The Sn-126 half-life has been deduced and the γ -ray intensities were also measured in order to improve the decay scheme data.
- A fourth volume of the Monographie BIPM 5 has been published.
- A specific volume dedicated to alpha and photon emissions, sorted by increasing energy, was published. It includes the energy and intensities of alpha or gamma emissions for about 400 nuclides. It can be sent on request.

4. International Activities

- CCRI(II) working groups.
- Liquid scintillation ICRM working group.
- VERMI workshops: Coincidence (LNHB; March 2007); Source Preparation (PTB; November 2007).
- EXSA (European Xray Spectrometry Association); organization of a workshop at LNHB (October 2008).
- DDEP (Decay Data Evaluation Project).
- Conferences: ICRM07 (Cape Town, September 2007); LSC2008, Advances on Liquid Scintillation (Davos, May 2008); EXRS2008 (June 2008); LTD12, Low Temperature Detectors (July 2007).

5. Miscellaneous

- In progress: test of the metrology beam line at SOLEIL (monochromatic in the 100 eV to 35 keV energy range).

- Development of a software (ACORES) for fitting efficiency curves for γ -spectrometry taking into account correlations between input data.

6. Publications

5.1 2007

M.-M. Bé, V.P. Chechev, R.Dersch, O.A.M. Helene, R.G. Helmer, M. Herman, S. Hlavac, A. Marcinkowski, G.L. Molnar, A.L. Nichols, E.Schönfeld, V.R. Vanin, M.J. Woods, "Update of X ray and gamma ray decay data standards for detector calibration and other applications", IAEA, Vienna (2007) in two volumes, ISBN92-0-113606-4.

A.J. Koning, et al. M.M. Bé, et al., "The JEFF evaluated nuclear data project". Proceedings Int. Conf. on Nuclear Data for Science and Technology (ND2007).

C. Dulieu, M.M. Bé, V. Chisté, "Tools and publications for reference decay data", Proceedings Int. Conf. on Nuclear Data for Science and Technology (ND2007).

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R. Broda, P. Cassette and K. Kossert, "Radionuclide metrology using liquid scintillation counting", Metrologia 44 (2007) S36-S52.

P. Bienvenu, P. Cassette, G. Andreoletti, M.M. Bé, J. Comte and M.C. Lépy, "A new determination of ^{79}Se half-life", Applied radiation and Isotope, Vol 65 (2007) 355-364.

M.-G. Iroulart, "Thermodynamic stability of radioactivity standard solutions", Monographie BIPM-6. Bureau International des Poids et Mesures, ISBN 92-822-2220-9, 2007.

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C. Bobin, "Primary standardization of activity using the coincidence method based on analogue instrumentation", Metrologia 44 (2007) S27-S31.

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M.-C. Lépy, J. Plagnard, "Development of low-energy X-ray spectrometry at the LNHB", Revista Mexicana de Física S53 (3) (2007) 68-73.

5.2 2008

R. Collé, B.E. Zimmerman, P. Cassette, L. Laureano-Perez, " ^{63}Ni , its half-life and standardization: Revisited", Applied Radiation and Isotopes 66 (2008) 60-68

M. C. Lépy, J. Plagnard, L. Ferreux, "Determination of ^{241}Am L X-ray emission intensities", Applied Radiation and Isotopes 66 (2008) 715-721

J. Plagnard, C. Hamon, M. C. Lépy, "Study of scattering effects in gamma-ray spectrometry", Applied Radiation and Isotopes 66 (2008) 769-773

T. Branger, C. Bobin, M-G. Iroulart, M-C. Lépy, I. Le Garrères, S. Morelli, D. Lacour, J. Plagnard, "Comparative study of two drying techniques used in radioactive source preparation: freeze-drying and evaporation using hot dry nitrogen jets", Applied Radiation and Isotopes (2008) 685-690

V. Gorozhankin, M.M. Bé, "Assessment of internal conversion coefficients for anomalous electric dipole transitions", Applied Radiations and Isotopes 66 (2008) 722

P. Cassette, Phuc Do, "The Compton source efficiency tracing method in liquid scintillation counting: A new standardization method using a TDCR counter with a Compton spectrometer", Applied Radiation and Isotopes 66 (2008) 1026-1032

A.C. Razdolescu, P. Cassette, M. Sahagia, "Measurement of ^{55}Fe solution activity by LSC-TDCR method", Applied Radiation and Isotopes 66 (2008) 750-755

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