Update on the international reference system (SIR) for gamma-ray emitting radionuclides

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1. The SIR and activity measurements

The SIR facility is now housed in a new laboratory area with temperature and humidity control. The reinstallation was successful, with stability maintained and the facility operational within three weeks.

Since the last CCRI(II) meeting in 2001, the BIPM has measured seventeen ampoules from eight laboratories: the BEV (Austria), BNM-LNHB (France), ČMI-IIR (Czech Republic), IRA (Switzerland), NIST (USA), NMIJ (Japan), OMH (Hungary) and PTB (Germany). Fifteen different radionuclides were submitted: ^{18}F, ^{22}Na, ^{51}Cr, ^{54}Mn, ^{59}Fe (2 results), ^{60}Co, ^{65}Zn, ^{67}Ga, ^{85}Sr, ^{88}Y (2 results), ^{110}Ag^{m}, ^{111}In, ^{131}I, ^{137}Cs and ^{152}Eu, giving seventeen new results.

There is one new radionuclide entry, ^{18}F ($T_{1/2}$ = 1.829 h ; u = 0.001 h). The half-life of ^{18}F is so short that the algorithm to determine the SIR equivalent activity A_e had to be modified to take into account the decay during the measurement. This introduced a relative change of about 5×10^{-3}, depending on the measurement conditions. The measured value of A_e is in agreement within the uncertainty with the value estimated from the efficiency curve of the SIR.

During 2002, the BIPM measured sixteen ampoules from thirteen laboratories: the BARC (India), BEV, BNM-LNHB, CIEMAT (Spain), ČMI-IIR, CSIR-NML, ININ (Mexico), IRA, NIST, NMIJ, OMH, PTB and the VNIIM (Russia). Sixteen different radionuclides were submitted: ^{18}F, ^{54}Mn, ^{57}Co, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{67}Ga, ^{85}Kr, ^{88}Y, ^{99}Tc^{m}, ^{125}I, ^{131}I, ^{134}Cs, ^{166}Ho^{m}, ^{222}Rn and ^{241}Am giving sixteen new results.

In April 2002, the twenty-two ampoules for the comparison of activity measurements of ^{65}Zn were measured in the SIR. In May 2002, seven ampoules prepared from the undiluted solution of ^{32}P of specific activity of about 70 MBq/g and filled with increasing mass of solution (from 1 g to 4.2 g) by the PTB were sent also for measurement in the SIR. After completion of this comparison and an evaluation of the activity concentration, these measurements will be used to improve the characterization of the SIR chamber response to the bremsstrahlung produced by the β-emission of ^{32}P. In September, two ampoules were measured for the ^{241}Am international comparison. Finally, twenty ampoules for the ^{192}Ir international comparison were measured in October.

Since the beginning of 2003, the BIPM has measured five ampoules from five laboratories: the CNEA (Argentina), CIEMAT, IRA, NMIJ and NPL (UK). Five different radionuclides were submitted: ^{18}F, ^{60}Co, ^{67}Ga, ^{137}Cs and ^{222}Rn, giving five new results. In addition, a solid source of ^{67}Ga sent by the NMIJ has been measured as part of a laboratory verification.

The cumulative number of ampoules measured since the introduction of the SIR in 1976 up to the present is now 840, corresponding to 611 independent results. The participants have approved all the results that have been measured. Indeed, since the beginning of the SIR only 39 results have been withdrawn, which represents 6.4 % of the number of results registered. The number of different radionuclides measured in the SIR is now 62.

Following the meeting of the Key Comparison Working Group in September 2002, significant progress has been made in publishing the results of the SIR comparisons in the key comparison database. It is planned to have all the past comparisons published by the end of 2003.

2. The SIR efficiency curve

For the determination of the gamma efficiency curve for a given radionuclide, the selection of experimental equivalent activity values to be included is based mainly on the following criteria: one result per laboratory, or a minimum 10-year interval between two results from the same laboratory; and, for low-energy γ-ray emitters, a low acid and/or carrier concentration of the radioactive solution..

The SIR gamma efficiency curve is determined by fitting functions to the experimental data for radionuclides in liquid solutions. The function used to fit the response was changed to a 6th-order

polynomial up to 1 MeV with an exponential decrease fitted at higher energies. The resulting gamma efficiency curve has a relative standard uncertainty less than 10^{-2} at energies above 65 keV and is often used to evaluate the SIR response to impurities. However, few data are available at low energies and this limits the precision of the fitted curve. There are other difficulties in that the curve is not directly applicable to annihilation gamma rays nor to gases for which the self-attenuation is different. The IRA has simulated the response of the ionization chamber (IC) of the SIR using the GEANT Monte Carlo code. The simulation seems to reproduce the measurements better than the efficiency curve functions, except at each end of the energy range (below 40 keV and above 1.8 MeV). This is under investigation. Using the Monte Carlo model developed, a study of the influence of the wall thickness of the glass ampoules and of the density of the solution on the response of the IC is in progress.

An additional data point for the beta efficiency curve of the SIR was obtained by measuring the undiluted key comparison solution of ^{89}Sr. An exponential curve fit yielded a relative uncertainty of about 5×10^{-2}, an acceptable value as the beta response is a second-order effect. The simulation of the response of the IC to beta-rays is also in progress at the IRA. As an input to this, a FORTRAN program has been developed at the BIPM to calculate beta spectra shapes for allowed and forbidden transitions taking into account Coulomb and screening effects. The program, based on the tables of Behrens and Jänecke, is being compared with other programs, in collaboration with the BNM-LNHB.

A project has been started in collaboration with the NPL to determine the mathematical function that best fits the curve and which is based on the various measurement equations. As the project develops, it will also take the beta efficiency into account.

3. Gamma spectrometry and the measurement of impurities

In addition to the regular stability checks of the Ge(Li) spectrometer using ^{60}Co and ^{137}Cs, impurity checks and activity measurements were made for ^{57}Co, ^{58}Co, ^{89}Sr, ^{110}Ag^{m}, ^{139}Ce, ^{177}Lu ^{18}F, ^{32}P, ^{131}I, ^{152}Eu and ^{238}Pu SIR ampoules. The results agreed within two standard uncertainties with the values of the participating NMIs.

The energy resolution of the HPGe spectrometer has been measured and shows a quadratic dependency. The resolution is similar to the Ge(Li) spectrometer at low energy, but is improved at high energy. In addition, the peak shape is nearly gaussian over the whole energy range, which will facilitate spectral analysis once the calibration of the HPGe is complete. The linearity of the electronic chain of the HPGe spectrometer has been improved by the construction of a linear gate based on a recent analogue switch integrated chip, with no deterioration of the energy resolution. The dynamic range remains large allowing measurements from 12 keV to 1.85 MeV.

4. Publications

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