Conclusions of the CCRI(II) Activity Comparisons Workshop
BIPM, 25 and 26 November 2004

1. Summary findings for the ^{192}Ir comparison

1.1 Low carrier concentration (20 μg g^{-1}) has resulted in more coherent results.

1.2 Some uncertainties seem to be slightly underestimated
- in particular from extrapolation (due to the EC branch) and
- from weighing for which the uncertainties should not be just the calibration of the balance. Sometimes it is better to use a dilution to decrease the uncertainty due to the weighing and work with source masses of about 100 mg. Uncertainty in the dilution may produce correlations that should then be taken into account.

1.3 Sources with high beta/EC efficiency are essential to see the change of slope in the extrapolation (which appears for certain gamma windows). This is relevant for both (P)PC and LS counting. Using PPC + Ge gives results showing clearly that the change of slope is due to the EC branch (see Y. Hino and H. Ohgaki, 1998, ARI **49**, 1179-1183).

1.4 PPC may be better than PC (more efficient, more linear).

1.5 The gamma windows should be set carefully in order to minimize the risks of underestimating the results (see D.G.F.Reher et al, NIM **A312** (1992) 263 and E.L.Grigorescu et al, NIM **A369** (1996) 414).

1.6 A simulation of the detector response is sometimes easier and more reliable than a deduction of the of the efficiencies from the equation. However, simulations at low energy can produce erroneous results.

2. Summary findings for the ^{90}Y comparison

2.1 Due to the short half-life, it is difficult to check for stability of the cocktail and for impurities. A recommendation is to prepare fresh cocktails when remeasuring the sample after some delay.

2.2 In relation to the short half-life, the first measurements may suffer from high count-rate, and it was noted that LSC dead-time corrections of some commercial counters are not reliable above some 10^3 counts per second.

2.3 The wall effect correction is not needed for high-energy emitters such as ^{90}Y.

2.4 Some LS counters have anti-coincidence shielding around the vial and this reduces the detection efficiency for gamma emitters and high energy beta emitters (bremsstrahlung).

2.5 Addition of acid helps to stabilize the sources used in liquid-scintillation counting.

3. Summary findings for the ^{204}Tl comparison

3.1 A low carrier concentration (~ 30 μg g^{-1}) has resulted in more coherent results.

3.2 The spread of the TDCR results is larger than the spread of the CIEMAT/NIST results and this should be investigated.

3.3 HNO_3 solutions are preferable when producing ^{204}Tl solid sources. Subsequent UV irradiation seems to increase the quality of the source by precipitating Tl into a metallic form.

3.4 No evidence of photosensitivity of ^{204}Tl cocktails could be shown.

3.5 $4\pi(x,e)$-x_K is a possible alternative method although used only by one participant.

3.6 The efficiency tracing with ^{60}Co gives results which are sometimes too low. The shape of the β spectrum of the tracer should be similar to that of the radionuclide to be measured.

4. Summary findings for the ^{32}P comparison

4.1 No clear explanation for the two groups of results could be found.

4.2 As the ^{33}P/^{32}P activity ratio increases rapidly with time, the organization of the source transport in advance is essential to allow all NMIs to measure the solution as close as possible to the reference date.

4.3 The various activity values reported for the impurities may be due to different threshold settings.

4.4 As a comparison using a pure ^{32}P solution would not reflect the capabilities of NMIs to measure commercially available solutions that always contain impurities, the recommendation for the new comparison is to use a commercially available solution.

4.5 Water can be used to stabilize the samples used for the liquid scintillation.

5. Summary findings for the ^{65}Zn comparison

5.1 The participants asked that in the report, the results of this comparison are compared with the results of the comparison carried out in the 1970's.

5.2 The low gamma-ray counting rate added to the difficulty of measurement.

5.3 It was recalled that when using the 4π(PC)-γ method a correction to the result must be applied as only some of the low-energy x-rays and Auger electrons are detected (see Funck, 1983, IJARI **34**/3, 565-569).

5.4 The use of a PPC is recommended due to its higher detection efficiency.

5.5 In the coincidence method the linearity conditions can be reached by modulating the photon efficiencies with some lead absorber. (e.g. Sahagia et al, 2004, *Appl. Rad. Isot*.**60,** 423-428.)

6. Summary findings for the ^{241}Am comparison

6.1 The presence of ^{241}Pu in the solution, which is not surprising in view of the source production mode, was detected by three laboratories. All the participants are recommended to measure this impurity if at all possible to enable a more robust evaluation of the impurity content. They should also consider the effect of this on their measurement technique and where appropriate evaluate the correction that should then be applied to their reported results. This may allow the BIPM and the KCWG to produce a more robust KCRV. However the result reported originally will be used in the KCDB for degrees of equivalence. As ^{241}Pu decays to ^{241}Am, the effect of that impurity may be indirect. This may also affect the ^{241}Am decay correction. However, in view of the long half-life of ^{241}Pu, the amount of ^{241}Am produced by its decay is negligible (less than 6×10^{-5} of the main ^{241}Am).

6.2 Some methods used by the participants were described as CIEMAT/NIST or TDCR when this was not strictly the case. Indeed they assumed 100 % α efficiency. It was recommended to identify another terminology.

6.3 Alpha particles in a LS cocktail generally produce very long after-pulses.

6.4 Small differences in the results were observed by some participants, depending on the cocktail or on the LS counter that was used, although this was not always the case.

6.5 For the coincidence method using a PC, no extrapolation is needed because on the α plateau, the electron/photon efficiency is negligible. This can be checked easily, experimentally, by trying to detect a beta emitter in the same counter conditions.

7. Summary findings for the ^{54}Mn comparison

7.1 The results show a larger spread than the SIR data.

7.2 The CIEMAT/NIST and TDCR results show a large spread but this is not surprising for such a high-energy gamma emitter. The efficiency calculation is very sensitive to differences in the *kB* values used. To increase efficiency, one laboratory used flat-faced cylindrical glass samples, and acid in the cocktail to minimize adsorption in glass.

7.3 The results of the comparison obtained by 4π(P)PC- γ (anti-)coincidence methods are consistent. However the results as a whole seem to show some correlation with the method used. It will be interesting to compare the results with the SIR data, when the link is calculated.

7.4 Several participants have indications for the presence of an impurity in the solution. It was concluded that the impurity is most likely to be a pure beta emitter with a half-life close to that of ^{54}Mn. The PTB will investigate the source production process in order to propose possible candidates. In consequence, the use of the results of this comparison for equivalence purposes was questioned. It was proposed that in the report of the comparison, the reference value obtained in this comparison is compared with that obtained in the previous comparison of ^{54}Mn made in 1965.

8. General comments

8.1 $4\pi\gamma$ (NaI) counting: it is important to check the Monte Carlo model of the detector by using a mono-gamma source of known activity.

8.2 It was recalled that the measurement of the impurities is part of the comparison and that impurities must be determined even when the standardization method is such that impurities would have negligible influence of the final result. This is of particular importance when the solution is measured in the SIR to make a link with other comparisons. For future comparisons, a new paragraph in the KC guidelines should describe the procedure related to the presence of impurities in the KC solutions.

8.3 A “Pomplot” may be useful for a given laboratory to see whether they over or under estimate their uncertainties in general but should not be used in relation to the MRA degrees of equivalence.

8.4 There was a request for a more detailed BIPM Report to include details of equipment settings used by participants; this could be facilitated by the introduction of electronic reporting sheets, a project that is in hand for 2005.

8.5 It would be useful to have graphs of degrees of equivalence expressed relatively to the KCRV. This could be tried in parallel with the present method for the next reported comparison.

8.6 There was a suggestion for a supplementary comparison to test dilution capability to support the activity ranges quoted in the CMCs. However, dilutions normally have negligible uncertainties compared to other sources of uncertainties.

Coincidence method

8.7 For the coincidence method, it is important to start by writing the complete coincidence equations and deduce which gamma gates will produce linear extrapolation. Monte Carlo simulation of PC can help when the equations are complex.

8.8 When making an extrapolation in the coincidence method, the correlation between the abscissa $(1 - \varepsilon_\beta)/\varepsilon_\beta$ and the ordinate $(N_\beta/\varepsilon_\beta)$ should be taken into account.

8.9 Coincidence efficiency tracing is a delicate method because its success depends on many parameters (chemistry/source production, energy spectra of the nuclide and the tracer, for example). Other measurement methods should be used when possible.

Liquid scintillation

8.10 Some LS counters make an automatic internal re-setting if they are left switched on for a long period. This may produce variations in the results.

8.11 When reporting LSC results, it is important to mention the code that was used to calculate the efficiency.

8.12 The LS producers may change their cocktail composition without warning and this may lead to error in the efficiency calculation. The production of a "home-made" scintillator is a satisfactory alternative.

8.13 Concerning the use of an external ^{3}H standard:

- the quench range of ^{3}H should be larger than that of the sample;
- the influence of the ^{3}H standard is very limited in the case of nuclides with high efficiency detection but plays an important role when the detection efficiency is low;
- in order to minimize the uncertainty on the quenching parameter, it may be better to have identical chemistry for the ^{3}H and sample cocktails - this is most important for radionuclides with low-energy β emission;
- the measurement of a fresh ^{3}H cocktail is also advisable.

8.14 TDCR: even when the 3 PM tubes are not matched, it is still possible to analyse the measurements. See for example the program of R. Broda "B4" available from the ICRM LSWG web site.

8.15 TDCR: to reduce the influence of after-pulses and random coincidences, need to use longer dead-times and short resolving times.

8.16 In the LS techniques, the Monte-Carlo simulation of the photon response is crucial above 20 keV. A current ICRM exercise is comparing simulation results in three different geometries.

Uncertainties

8.17 The relative uncertainty of weighing small drops (≤ 10 mg) is high so it may be better to dilute the solution first, although this also introduces some uncertainty.

8.18 In an uncertainty budget, the reported uncertainty on weighing may be the remaining Type B uncertainty, the Type A uncertainty being included in the counting uncertainty through the mean of several measurements of different sources. The same argument is valid for dead-time and background uncertainties. A BIPM monograph on the uncertainty in weighing would be welcomed by the participants.

8.19 In the coincidence method, the uncertainty of the extrapolation is significantly different from one NMI to the other and this should be investigated.

8.20 In the CIEMAT/NIST method, an uncertainty on the ^{3}H standard of 10^{-2} generally produces an uncertainty of about 10^{-4} on the nuclide activity measurement. In other words, the sensitivity coefficient is about 10^{-2}, depending on the nuclide.

8.21 The Uncertainties Working Group of the CCRI(II) will produce templates for uncertainty budgets for different measurement methods to help with coherence in uncertainty estimation between the participants.

8.22 It was proposed for a future Workshop to organize a comparison of data analysis and uncertainty evaluation by distributing sample data and comparing the final result and uncertainty deduced by each participant.

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