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Verification of the linearity of the new SIR using sources of ^{64}Cu and ^{99m}Tc

by Carine Michotte

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Pavillon de Breteuil, F-92312 SEVRES cedex

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Carine Michotte, BIPM

The linearity of the new SIR electronics has been verified by looking for deviations from the exponential decay for several ^{64}Cu and ^{99m}Tc sources. Data analyses are compared with the half-life used as a fixed or free parameter. It is concluded that no significant non-linearity is observed in the new SIR. For comparison, the same method has been applied to analyse existing data from the original SIR.

1. Introduction

Non-linearities of the electronics over the whole range of current measurements (1 pA to 300 pA) is a potential source of uncertainty for the SIR measurements. A common method to check the linearity is to follow the decay of short-lived radionuclides and look for deviations from the exponential decay law [1, 2].

In the frame of the SIR ongoing comparisons, several ampoules of ^{99m}Tc and ^{64}Cu were measured in the new SIR [3] and these measurements were used to check the linearity. Some such measurements are also available for the original SIR, ^{99m}Tc having been measured since 1998, so the linearity of the two systems can be compared.

2. Measurement model

In the SIR, the ionization current is obtained by measuring the time d_i to charge a reference capacitor C_j up to a selected reference voltage ΔV_j, using the Townsend balance principle [1, 3, 4]. Each ampoule measurement (a series of 10 measurements) is preceded and followed by the measurement of one of the five ^{226}Ra reference sources. The current ratio $R_{i,j}(t_i)$ of the ith measurement of the ampoule in the jth series and of the radium source is calculated using (1), taking into account the decay of the short-lived radionuclide during the measurement.

$$R_{i,j}(t_i) = \frac{\left(C_j \Delta V_j / d_i\right) - I_{\mathrm{bg},j}}{\left(\dfrac{T}{d_i \ln(2)}\right)\left(1 - e^{-d_i \ln(2)/T}\right)} \left(I_{\mathrm{Ra},j}(t_0) F_j\right)^{-1} C_{\mathrm{imp}}(t_i) \qquad (1)$$

where T is the half-life of the radionuclide [5];
t_i is the starting time of the ith measurement;
$I_{\mathrm{bg},j}$ is the background ionization current for the jth series of measurements;
$I_{\mathrm{Ra},j}(t_0)$ is the mean current of the preceding and following Ra measurements, evaluated at the reference time of the SIR (1976);
F_j is the tabulated current ratio of the radium source used for the jth series and radium source no. 5;
$C_{\mathrm{imp}}(t_i)$ is the correction for impurity if relevant [6].

3. Data analysis and results

The current ratios $R_{i,j}(t_i)$ measured for four ^{64}Cu different ampoules and one ^{99m}Tc ampoule are plotted versus time and fitted using Origin8 software to an exponential with a fixed half-life [5] taking into account the weights coming from the statistical uncertainties and $u(F_j)$.

For the ^{99m}Tc ampoule, the influence of the ^{99}Mo impurity on the SIR result has been taken into account although it corresponds to only a few parts in 10^5 and has a negligible influence on the non-linearity results.

The plots are shown in Figure 1 and the residuals in Appendix 1. The uncertainty of the fit is negligible compared to the uncertainty of the current ratios. All residuals agree with the fitted curves within two standard uncertainties. However, it may happen that some series of measurements tend to be systematically above or below the curve (see Figure 2). Consequently, the mean and standard deviation of the residuals of each series of ten measurements are calculated and shown in Figure 3 as a function of the current ratio. Any trends or structures in the residuals in Figure 3 would reflect non-linearities of the measurement system. The corresponding non-linearity value for each radium source is taken as the weighted mean of the relative residuals. In fact, the relative residuals plotted in Figure 3 do not show any significant trend or structure which could correspond to a non-linearity. The weighted mean values of the residuals over all measurements of all ampoules are given in Table 1 and are not significantly (k = 2) different from zero.

4. Fixed or fitted half-life

As explained above, the data were fitted to an exponential with a half-life fixed at the recommended value. This was because a half-life taken as a free parameter could help the fit to be closer to the data and so possibly hide part of any non-linearity. In addition, the fitted half-life could then not be compatible with the recommended value (T = 12.701 (2) h [5] for ^{64}Cu). However, the disadvantage of fixing the half-life is that its uncertainty as given in the literature is not taken into account.

As a test, a global fit was carried out by exponential fitting of the data from all the ^{64}Cu ampoules in a single minimization procedure with a common half-life to be fitted for all data. The ^{99m}Tc ampoule was tested separately (see below). As expected the “non-linearity” deduced from this test for radium 1 is generally smaller than with a fixed half-life. In fact no significant (k = 2) non-linearity is obtained whether the half-life is fixed or not in the fit (see Table 1). The interesting result is that the half-life obtained from this global fit is 12.7033 (8) h which is in agreement with [5].

When deducing the half-life of ^{99m}Tc from a fit of the NPL 2008 data with the radium source 1, the result T = 6.012 (15) h is in agreement with [5] (T = 6.0067 (10) h), as in the case of ^{64}Cu.

The data thus seem to be consistent and the values in Table 1 reliable.

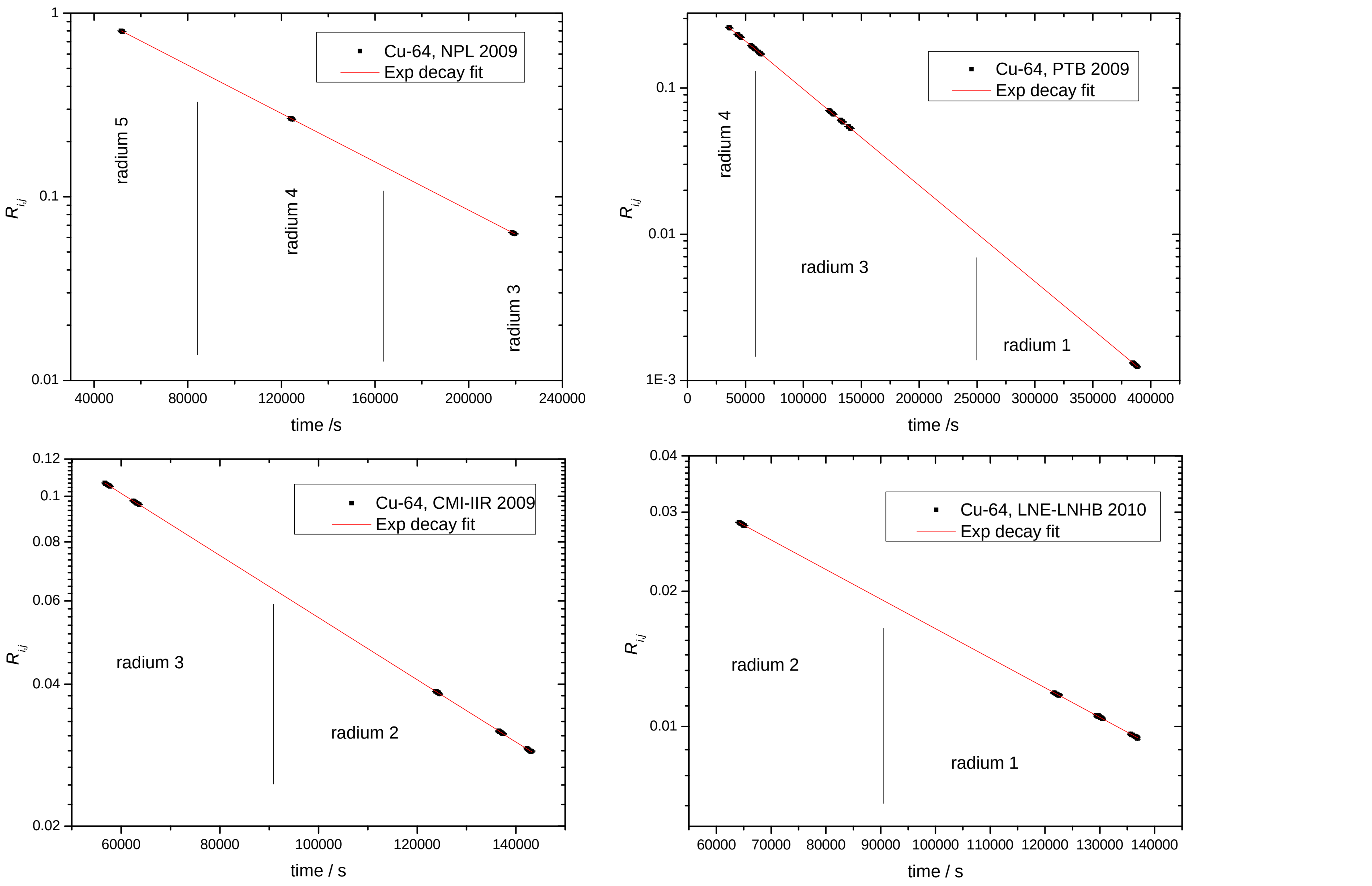

Figure 1 – Continued overleaf

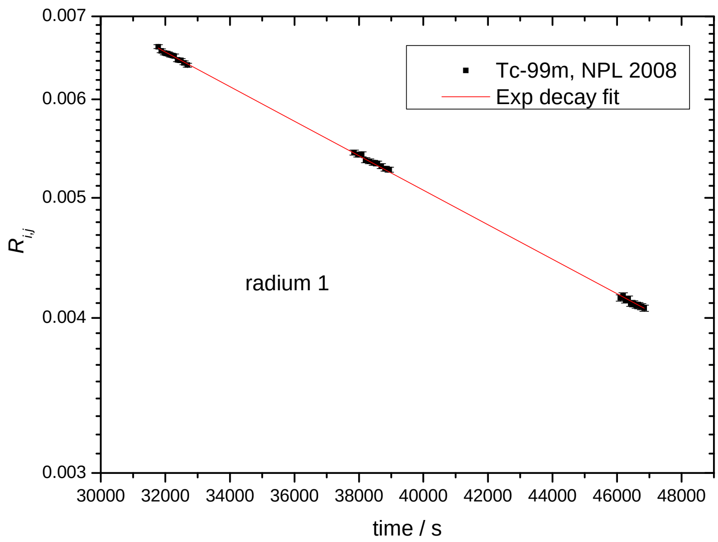

Figure 1. Fitted exponential decay curves for the five SIR ampoules used to evaluate the SIR linearity.

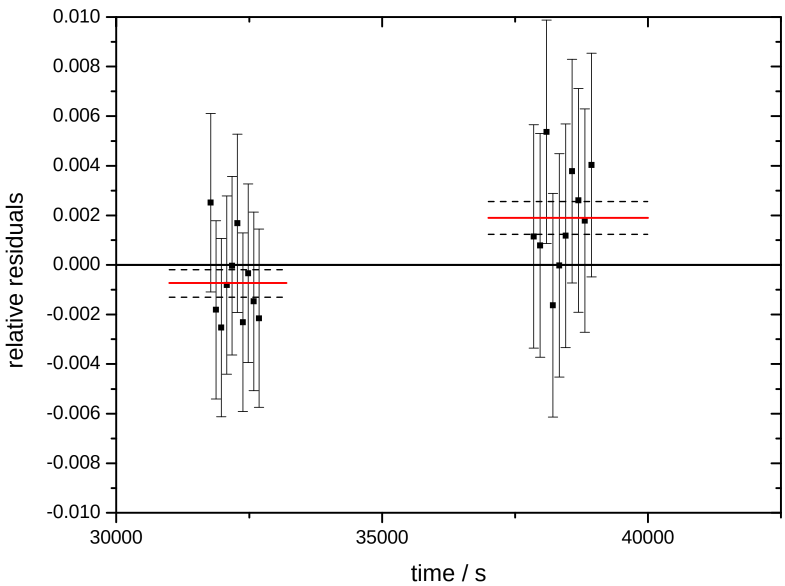

Figure 2. Example of relative residuals obtained for two series of measurements with the new SIR and a ^{99m}Tc ampoule at low current (radium source 1). The red lines and dotted lines correspond to the mean relative residuals and the standard deviation of the mean. On the left no significant (k = 2) deviation from zero is observed. On the right the residuals are systematically above zero.

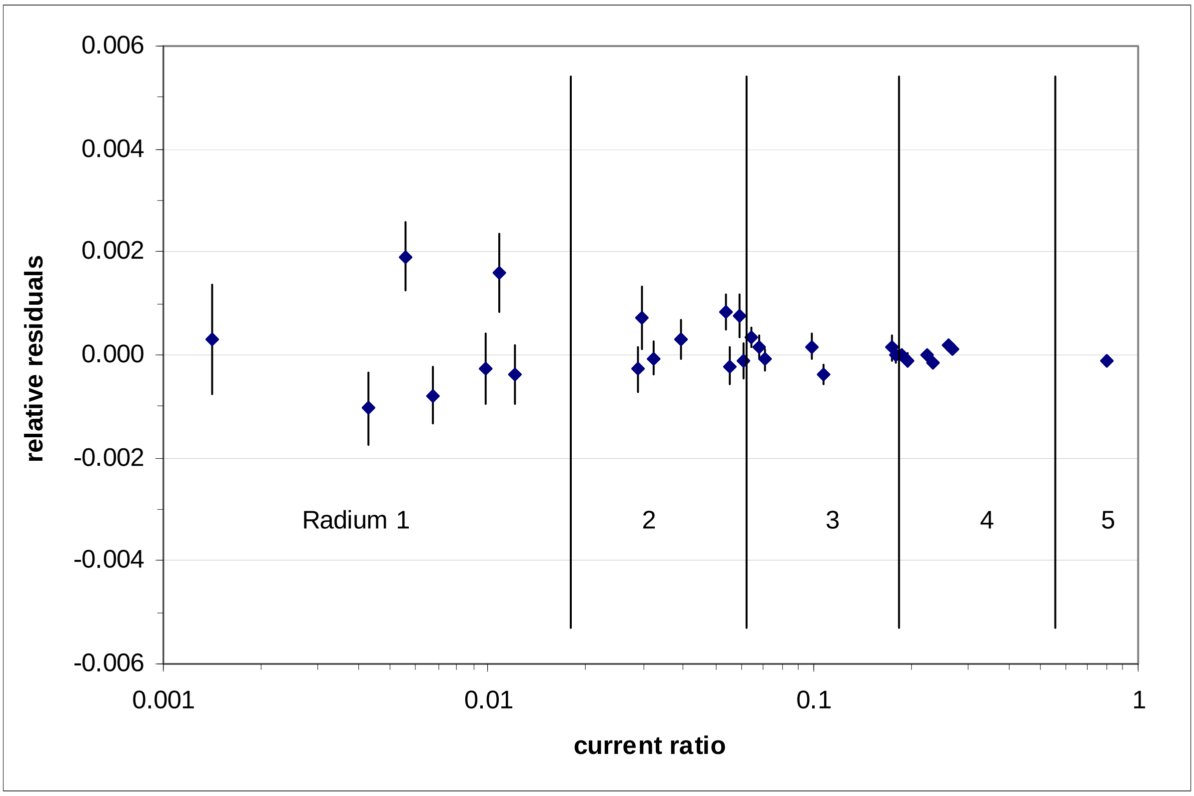

Figure 3. Mean relative residuals (one data point per series of ten measurements) of the fit to an exponential of the decaying current ratio for the new SIR.

5. Comparison with the original SIR

For comparison, the same procedure (with a fixed half-life) has been applied to ^{99m}Tc measurements in the original SIR (ampoules from LNE-LNHB 1998; BEV 2002; PTB 2005 and NPL 2005). Again, the influence of the ^{99}Mo impurity on the SIR results is at most a few parts in 10^5 and has been taken into account. In contrast to the new SIR, significant (k = 2) non-linearities are observed and for large currents: -2.3(4) × 10^{-4} for radium source 5 and 2.2(9) × 10^{-4} for radium source 2 as given in Table 1 and on the graph of residuals in Figure 4. However, it should be noted that the result for radium source 5 is based on a single series of measurements. Besides, the mean non-linearity value for radium source 2 is small compared with other sources of uncertainty in the SIR measurement. Indeed, for low currents such as those produced by the radium source 2, the statistical uncertainty is about (4 to 9) × 10^{-4} in relative terms. In addition, the relative uncertainty associated with F (normalization ratio to radium source 5) is 4 × 10^{-4} for this source. Finally, it is important to note that the five first data points in Figure 4 correspond to ionization currents up to a factor of five lower than the normal measuring conditions in the SIR.

When making a global exponential fit of all the ^{99m}Tc data with the half-life as a common free parameter as in section 4, the half-life obtained is T = 6.006 85(24) h which agrees with [5] (T = 6.0067(10) h). In this case, the weighted mean residuals obtained from the global fit are similar to the values given above, although slightly smaller (see Table 1). This indicates that the non-linearities may not be related to the half-life value. More data would be necessary to confirm the presence of non-linearities in the original SIR. However, this system is no longer in use at the BIPM.

Table 1. Weighted mean value and standard uncertainty of the relative residuals. Results significantly different from zero are highlighted.

	New SIR		Original SIR	
	fixed *T*	fitted *T*	fixed *T*	fitted *T*
Radium 5	-1.2(9) × 10^{-4}	-0.2(9) × 10^{-4}	-2.3(4) × 10^{-4}	-2.2(4) × 10^{-4}
Radium 4	0.7(50) × 10^{-5}	0.2(5) × 10^{-4}	0.9(7) × 10^{-4}	0.9(7) × 10^{-4}
Radium 3	0.3(9) × 10^{-4}	-0.7(8) × 10^{-4}	0.2(7) × 10^{-4}	0.2(7) × 10^{-4}
Radium 2	2.0(16) × 10^{-4}	0.9(15) × 10^{-4}	2.2(9) × 10^{-4}	1.7(9) × 10^{-4}
Radium 1	0.6(43) × 10^{-4}	-0.4(44) × 10^{-4}	-1.3(2.3) × 10^{-4}	-1.9(23) × 10^{-4}

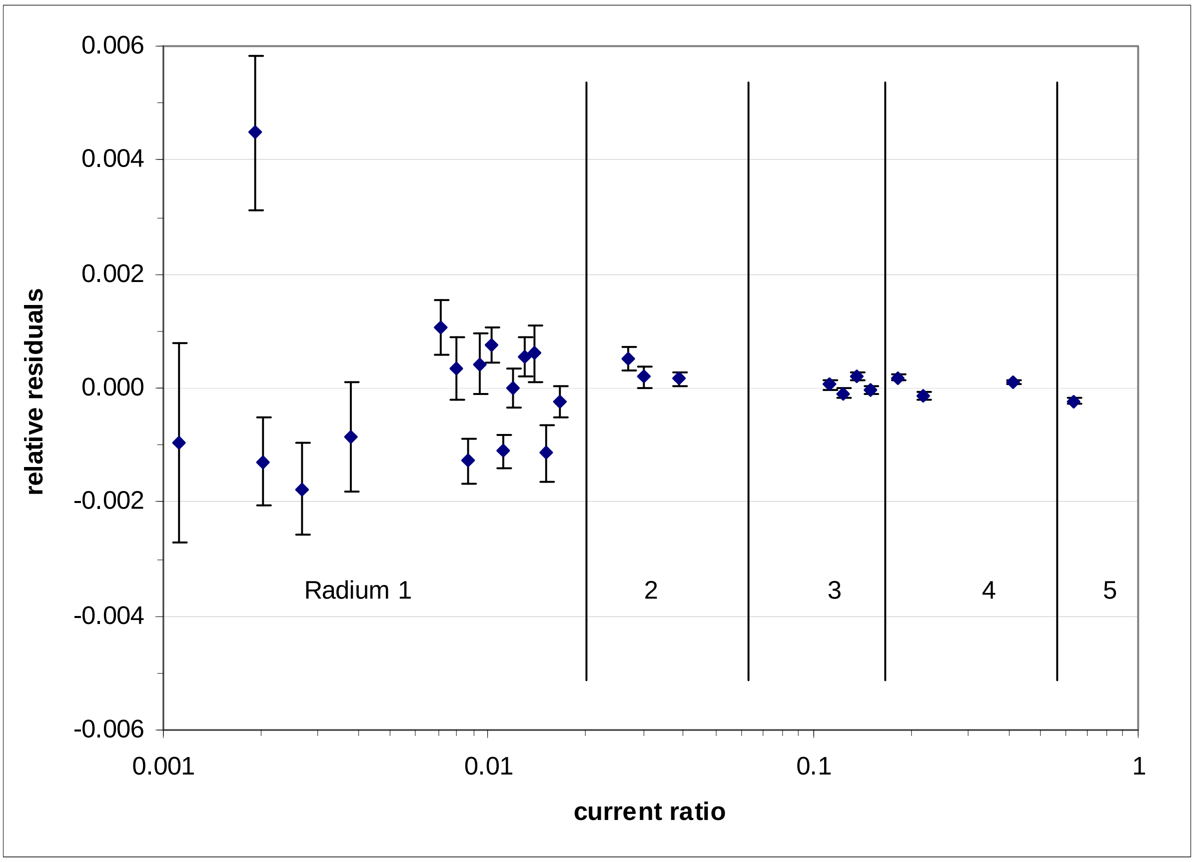

Figure 4. Mean relative residuals (one data point per series of ten measurements) of the fit to an exponential of the decaying current ratio for the original SIR.

7. Conclusion

The non-linearities observed for the original SIR, if they could be confirmed, are one order of magnitude smaller than the typical standard uncertainties quoted by the NMIs for their primary measurements. Thus the reliability of the SIR results based on the original SIR is not in question. A global fit of all the ^{99m}Tc data gives a half-life T = 6.006 85(24) h, in agreement with [5].

No significant non-linearity could be observed in the new SIR for either of the fitting methods, which is a satisfactory outcome of the tests. The global fit of all the ^{64}Cu data gives a half-life T = 12.7033 (8) h, in agreement with [5].

In both the original SIR and the new SIR, the results conform to the expected values for high-quality instruments, that is a linearity better than 1 part in 10^3 [1].

Acknowledgements

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References

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Appendix 1. Value and standard uncertainty of the relative residuals from the fit to exponential decay with a fixed half-life for the new SIR

Ampoule from NPL, 2009

Time /s	Rel. residual	*u*
51528	-4.02E-04	4.16E-04
51605	-4.14E-04	4.16E-04
51683	-2.08E-04	4.16E-04
51759	-1.28E-04	4.16E-04
51836	2.35E-04	4.16E-04
51914	-4.78E-04	4.16E-04
51991	-2.26E-04	4.16E-04
52069	1.17E-04	4.16E-04
52146	3.08E-04	4.16E-04
52224	-1.46E-05	4.16E-04
123777	2.39E-04	5.57E-04
123886	7.19E-05	5.57E-04
123996	1.18E-04	5.57E-04
124106	-2.66E-04	5.57E-04
124216	1.96E-04	5.57E-04
124326	4.10E-04	5.57E-04
124435	6.35E-04	5.57E-04
124546	1.03E-04	5.57E-04
124657	-3.02E-04	5.57E-04
124768	-6.21E-06	5.57E-04
218430	-1.65E-04	1.04E-03
218579	-8.21E-05	1.04E-03
218728	5.90E-04	1.04E-03
218878	4.83E-04	1.04E-03
219028	-1.20E-04	1.04E-03
219178	4.25E-04	1.04E-03
219329	1.40E-03	1.04E-03
219479	7.27E-04	1.04E-03
219631	-7.17E-04	1.04E-03
219782	9.66E-04	1.04E-03

Ampoule from PTB, 2009

Time /s	Rel. residual	*u*
35622	-3.76E-04	6.47E-04
35734	1.97E-04	6.47E-04
35847	1.49E-04	6.47E-04
35960	5.79E-05	6.47E-04
36073	4.32E-04	6.47E-04
36186	9.20E-04	6.47E-04
36298	-1.07E-04	6.47E-04
36412	4.94E-04	6.47E-04
36525	1.39E-04	6.47E-04
36639	-1.12E-04	6.47E-04
42779	-2.83E-04	4.64E-04
42903	7.84E-05	4.64E-04
43027	-1.52E-04	4.64E-04
43152	7.83E-05	4.64E-04
43277	4.32E-05	4.64E-04
43402	-4.18E-04	4.64E-04
43527	-6.88E-04	4.64E-04
43651	-3.04E-04	4.64E-04
43777	8.67E-05	4.64E-04
43903	1.08E-04	4.64E-04
45277	-6.12E-05	5.55E-04
45406	8.29E-04	5.55E-04
45535	-9.31E-05	5.55E-04
45664	-2.51E-04	5.55E-04
45793	3.92E-06	5.55E-04
45922	-3.54E-04	5.55E-04
46052	1.00E-04	5.55E-04
46181	5.40E-05	5.55E-04
46312	-1.15E-04	5.55E-04
46442	-2.52E-04	5.55E-04
54556	1.04E-04	6.47E-04
54702	-8.07E-05	6.47E-04
54848	-1.03E-03	6.47E-04
54995	3.90E-04	6.47E-04
55143	3.27E-04	6.47E-04
55291	-6.49E-04	6.47E-04
55438	-2.42E-04	6.47E-04
55586	-2.84E-05	6.47E-04
55734	2.44E-04	6.47E-04
55883	-2.94E-05	6.47E-04

Ampoule from PTB, 2009 – Continued

Time /s	Rel. residual	*u*
57220	3.64E-04	5.55E-04
57373	4.04E-04	5.55E-04
57524	-5.26E-04	5.55E-04
57677	3.47E-04	5.55E-04
57830	-6.26E-04	5.55E-04
57983	1.98E-04	5.55E-04
58137	-6.02E-05	5.55E-04
58290	-3.37E-05	5.55E-04
58445	5.10E-05	5.55E-04
58599	6.91E-05	5.55E-04
61241	-6.00E-04	8.22E-04
61301	3.30E-04	8.22E-04
61362	4.15E-04	8.22E-04
61422	-5.27E-04	8.22E-04
61483	-1.52E-04	8.22E-04
61543	-2.14E-04	8.22E-04
61603	-5.72E-05	8.22E-04
61664	-5.67E-04	8.22E-04
61725	3.72E-04	8.22E-04
61786	9.01E-04	8.22E-04
63114	1.09E-03	1.19E-03
63175	8.42E-04	1.19E-03
63237	4.75E-04	1.19E-03
63299	4.89E-04	1.19E-03
63361	-4.15E-04	1.19E-03
63423	-1.16E-03	1.19E-03
63486	-2.82E-05	1.19E-03
63548	1.08E-03	1.19E-03
63609	-8.40E-04	1.19E-03
63671	-1.96E-04	1.19E-03
122211	-6.04E-04	1.13E-03
122348	1.18E-03	1.13E-03
122485	1.20E-03	1.13E-03
122622	-3.44E-04	1.13E-03
122759	-7.51E-04	1.13E-03
122897	-3.03E-04	1.13E-03
123035	4.44E-05	1.13E-03
123173	-6.27E-04	1.13E-03
123312	-3.45E-04	1.13E-03
123450	-3.81E-04	1.13E-03

Time /s	Rel. residual	*u*
124922	-4.36E-05	1.19E-03
125063	1.29E-03	1.19E-03
125206	6.41E-04	1.19E-03
125348	7.20E-04	1.19E-03
125491	4.20E-04	1.19E-03
125634	-5.03E-04	1.19E-03
125777	-1.14E-03	1.19E-03
125921	6.45E-04	1.19E-03
126065	1.63E-04	1.19E-03
126209	-5.24E-04	1.19E-03
132126	5.45E-04	1.57E-03
132186	6.31E-04	1.57E-03
132246	-1.27E-03	1.57E-03
132306	2.39E-04	1.57E-03
132365	2.20E-05	1.57E-03
132425	1.97E-03	1.57E-03
132485	-8.63E-04	1.57E-03
132545	-1.73E-03	1.57E-03
132606	-4.92E-04	1.57E-03
132666	-2.41E-04	1.57E-03
133873	1.01E-03	1.76E-03
133934	-2.77E-04	1.76E-03
133995	1.26E-03	1.76E-03
134056	2.64E-04	1.76E-03
134118	2.27E-03	1.76E-03
134179	-1.98E-03	1.76E-03
134240	2.32E-03	1.76E-03
134302	3.43E-04	1.76E-03
134363	3.41E-04	1.76E-03
134424	2.09E-03	1.76E-03
138695	-8.10E-04	2.13E-03
138760	5.81E-05	2.13E-03
138825	1.53E-03	2.13E-03
138890	9.77E-04	2.13E-03
138955	-1.73E-03	2.13E-03
139020	3.06E-04	2.13E-03
139086	6.76E-04	2.13E-03
139151	-1.07E-03	2.13E-03
139216	1.43E-05	2.13E-03
139282	-2.07E-03	2.13E-03

Ampoule from PTB, 2009 – Continued

Time /s	Rel. residual	*u*
140469	1.74E-03	2.07E-03
140535	-9.91E-04	2.07E-03
140602	2.45E-03	2.07E-03
140668	1.09E-03	2.07E-03
140735	1.37E-03	2.07E-03
140801	-6.94E-05	2.07E-03
140868	-7.44E-04	2.07E-03
140935	8.96E-04	2.07E-03
141002	1.21E-03	2.07E-03
141069	1.33E-03	2.07E-03
384294	5.02E-04	4.89E-03
384732	-4.35E-03	4.89E-03
385171	3.69E-03	4.89E-03
385612	4.47E-03	4.89E-03
386056	5.20E-03	4.90E-03
386504	-8.01E-04	4.90E-03
386954	-1.84E-03	4.90E-03
387410	6.41E-04	4.90E-03
387868	-3.90E-04	4.90E-03
388325	-4.05E-03	4.90E-03

Ampoule from CMI-IIR, 2009

Time /s	Rel. residual	*u*
56687	1.26E-04	9.83E-04
56808	-1.63E-03	9.83E-04
56931	-3.49E-04	9.83E-04
57053	-2.28E-04	9.83E-04
57176	-9.85E-04	9.83E-04
57299	-9.56E-04	9.83E-04
57422	-1.03E-04	9.83E-04
57545	2.07E-04	9.83E-04
57668	1.41E-04	9.83E-04
57792	7.35E-05	9.83E-04
62472	1.30E-03	1.01E-03
62604	3.97E-04	1.01E-03
62736	-2.97E-04	1.01E-03
62869	2.26E-04	1.01E-03
63002	2.57E-04	1.01E-03
63135	-9.01E-04	1.01E-03
63269	-6.31E-04	1.01E-03
63403	-4.69E-04	1.01E-03
63536	1.32E-03	1.01E-03
63670	4.15E-04	1.01E-03

Ampoule from CMI-IIR, 2009 – Continued

Time /s	Rel. residual	*u*
123701	-2.75E-04	1.46E-03
123789	2.47E-05	1.46E-03
123876	1.57E-03	1.46E-03
123963	7.86E-04	1.46E-03
124051	1.49E-04	1.46E-03
124139	4.78E-04	1.46E-03
124227	-8.41E-04	1.46E-03
124315	1.78E-03	1.46E-03
124403	1.59E-03	1.46E-03
124492	-2.13E-03	1.46E-03
136491	1.07E-03	2.13E-03
136595	-1.40E-03	2.13E-03
136699	-1.06E-03	2.13E-03
136804	1.17E-03	2.13E-03
136908	1.07E-03	2.13E-03
137013	-4.80E-04	2.13E-03
137117	1.03E-03	2.13E-03
137222	-7.51E-04	2.13E-03
137328	-1.09E-03	2.13E-03
137433	-2.19E-04	2.13E-03
142225	1.71E-03	2.69E-03
142338	6.59E-05	2.69E-03
142450	3.88E-03	2.69E-03
142563	-5.02E-04	2.69E-03
142676	1.13E-03	2.69E-03
142789	-1.31E-03	2.69E-03
142903	-2.31E-03	2.69E-03
143017	9.93E-04	2.69E-03
143129	6.68E-05	2.69E-03
143244	0.00346	2.69E-03

Ampoule from LNE-LNHB, 2010

Time /s	Rel. residual	*u*
64146	7.11E-04	1.87E-03
64261	1.21E-03	1.87E-03
64376	-2.63E-04	1.87E-03
64492	-8.11E-05	1.87E-03
64608	1.04E-03	1.87E-03
64723	-2.03E-04	1.87E-03
64839	-2.92E-04	1.87E-03
64955	-3.34E-03	1.87E-03
65072	-1.91E-03	1.87E-03
65189	4.09E-04	1.87E-03
121619	-2.28E-03	3.01E-03
121729	1.23E-03	3.01E-03
121839	8.09E-04	3.01E-03
121950	-3.10E-03	3.01E-03
122061	8.59E-05	3.01E-03
122172	3.46E-04	3.01E-03
122283	-1.48E-03	3.01E-03
122394	-1.36E-04	3.01E-03
122505	-0.00182	3.01E-03
122617	2.61E-03	3.01E-03
129374	7.45E-04	3.26E-03
129497	4.43E-04	3.26E-03
129619	-1.25E-03	3.26E-03
129742	6.69E-03	3.26E-03
129865	1.88E-03	3.26E-03
129988	3.69E-03	3.26E-03
130112	-7.35E-04	3.26E-03
130236	1.90E-03	3.26E-03
130359	2.83E-03	3.26E-03
130483	-3.74E-04	3.26E-03
135650	1.21E-03	3.23E-03
135783	-3.25E-03	3.23E-03
135918	4.39E-05	3.23E-03
136051	1.13E-03	3.23E-03
136185	-1.79E-03	3.23E-03
136320	1.44E-03	3.23E-03
136455	-7.49E-04	3.23E-03
136589	2.35E-03	3.23E-03
136725	1.26E-03	3.23E-03
136859	-4.23E-03	3.23E-03

Ampoule from NPL, 2008 (^{99m}Tc)

Time /s	Rel. residual	*u*
31776	2.44E-03	3.58E-03
31875	-1.87E-03	3.58E-03
31976	-2.59E-03	3.58E-03
32077	-8.75E-04	3.58E-03
32178	-9.45E-05	3.58E-03
32279	1.62E-03	3.58E-03
32379	-2.37E-03	3.58E-03
32481	-3.93E-04	3.58E-03
32583	-1.53E-03	3.58E-03
32686	-2.21E-03	3.58E-03
37851	1.15E-03	4.51E-03
37970	7.97E-04	4.51E-03
38090	5.38E-03	4.51E-03
38209	-1.62E-03	4.51E-03
38329	-4.51E-06	4.51E-03
38450	1.19E-03	4.51E-03
38572	3.79E-03	4.51E-03
38693	2.62E-03	4.51E-03
38814	1.81E-03	4.51E-03
38937	4.05E-03	4.51E-03
46106	-4.20E-03	5.75E-03
46187	2.22E-03	5.75E-03
46268	-2.85E-03	5.75E-03
46349	1.77E-03	5.75E-03
46430	-3.67E-03	5.75E-03
46512	-2.00E-03	5.75E-03
46594	-1.96E-03	5.75E-03
46676	-1.36E-04	5.75E-03
46758	3.86E-04	5.75E-03
46840	1.43E-04	5.75E-03