First measurement of ^{177}Lu in the SIR

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^{177}Lu has been measured for the first time in the International Reference System (SIR) for activity comparisons. Two laboratories have participated and the results and the influence of impurities are presented.

The PTB and the NIST have each sent to the BIPM one ampoule of a solution of ^{177}Lu ($T_{1/2}$ = (6.646 ± 0.005) d ; Schötzig, 2001)[a] prepared from the same parent solution. Each ampoule was measured in the ionization chamber (I.C.) of the SIR, at the end of January 2000 for the PTB ampoule and during the second half of February 2000 for the NIST ampoule. The solution contains some ^{177}Lu^{m} ($T_{1/2}$ = 160.4 d) the activity of which has been measured independently in both laboratories and at the BIPM. The correction factor to account for the ionization current produced by this impurity is

$$C = 1 + \frac{A_{177m}}{A} \cdot \frac{A_{\mathrm{e}}}{A_{\mathrm{e},177m}} \cdot D = 1 + R \cdot \frac{A_{\mathrm{e}}}{A_{\mathrm{e},177m}} \cdot D \ , \qquad (1)$$

where A and $A_{177\mathrm{m}}$ (A_{e} and $A_{\mathrm{e},177\mathrm{m}}$) are the activities (equivalent activities), at the reference date, of the main radionuclide and the ^{177}Lu^{m} impurity, respectively, and D is a decay correction (Michotte, 2000). The equivalent activities for ^{177}Lu and ^{177}Lu^{m} are not known experimentally but are determined from the efficiency curve of the SIR

^{177}Lu and ^{177}Lu^{m} equivalent activity calculated from the efficiency curve

The method of calculation of an equivalent activity from the efficiency curve of the SIR is described in (Rytz, 1983). Using the x- and γ-ray emission probabilities from (Schötzig, 2001) and the efficiency curve of the SIR, an equivalent activity was obtained for ^{177}Lu, $A_{\mathrm{e,calc}}$ = (565.8 ± 3.1) MBq. Using the emission probabilities from the Nuclear Data Sheets **68**, 747 (1993), the value obtained for $A_{\mathrm{e},177\mathrm{m}}$ is (17 010 ± 160) kBq, taking into account both the γ and β decay of ^{177}Lu^{m}, and the decay of the daughter ^{177}Lu (assuming equilibrium).

SIR measurements of the PTB ^{177}Lu ampoule

The PTB measured the specific activity of the solution by the coincidence method using a proportional counter and by the CIEMAT/NIST method. The two results agreed within the uncertainties (Schötzig, 2001).

The first SIR measurement of the ^{177}Lu solution took place one week after the PTB reference date ($t_{\mathrm{R,PTB}}$ = 19 Jan. 2000) and was repeated twice. Using $A_{\mathrm{e,calc}}$ = (565.8 ± 3.1) MBq and the PTB measured impurity content R_{PTB} = (6.9 ± 0.6)×10^{-5} in the correction factor C, the following SIR results were obtained:

[a] all uncertainties given are standard uncertainties.

Measurement date	Equivalent activity / kBq	Uncertainty / kBq
26-01-2000	559 000	2 720
31-01-2000	559 040	2 780
07-02-2000	558 820	2 940

Table 1: PTB SIR results, using $A_{e,calc}$ and R_{PTB} in the correction factor C.

The impurity content was also measured at the BIPM, giving $R_{GeLi,\,1} = (6.7 \pm 0.3)\times10^{-5}$, which is in very satisfactory agreement with the PTB value. Figure 1 shows that the PTB SIR results are almost insensitive to the impurity content value.

The SIR results are very sensitive to the value used for the half-life of ^{177}Lu. The stability of the SIR results over 12 days shows that the decay corrections using the PTB value for the half-life of ^{177}Lu [$T_{1/2}$ = (6.646 ± 0.005) d] are correct. On the contrary, when the previously recommended half-life of (6.734 ± 0.012) d (NDS **68**, 747 (1993); Bé, 1998) is used, the results are much less coherent (Figure 1), suggesting that the latter value should be wrong.

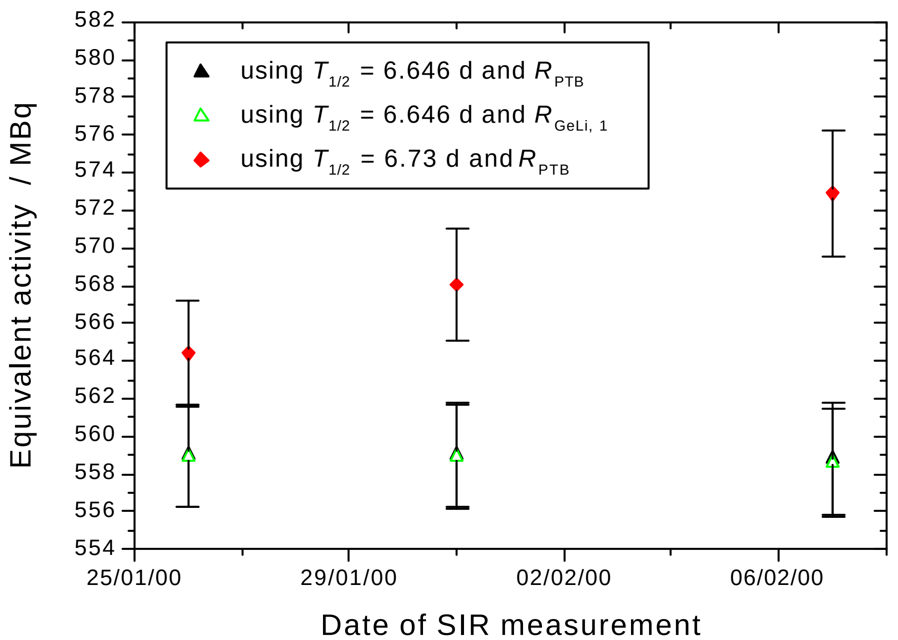

Figure 1. PTB ^{177}Lu SIR results using different values for the $T_{1/2}$ and R quantities.

SIR measurements of the NIST ^{177}Lu ampoule

The NIST measured the specific activity of the solution by the CIEMAT/NIST method. A measurement by the 4π(NaI(Tl))γ method gave a result in agreement within the uncertainties. However, as its uncertainty is much larger, it has not been considered for submission to the SIR (Zimmerman, 2001)

Half-life measurements carried out at the NIST (Zimmerman, 2001) give a value of (6.65 ± 0.01) d for ^{177}Lu, in agreement with the PTB . However, the more precise PTB value is used in the following.

The first SIR measurement of the NIST ampoule took place two weeks after the NIST reference date ($t_{R,NIST}$ = 1 Feb. 2000, 17h00 UT) and was repeated twice. The impurity content given by the NIST is R_{NIST} = $(2.2 \pm 0.6)\times10^{-4}$ and is in agreement within the uncertainties with the BIPM value of $R_{GeLi,\,2}$ = $(2.71 \pm 0.05)\times10^{-4}$. The SIR results using both impurity contents are shown in Figure 2.

The impurity contents $R_{GeLi,\,1}$ ($t_{R,PTB}$) and $R_{GeLi,\,2}$ ($t_{R,NIST}$) were obtained by measuring independently[b] the PTB and NIST ampoules, respectively. These ampoules contain a ^{177}Lu solution coming from the same parent solution, and having, in consequence, the same impurity content. Indeed, applying a correction for decay, we have $R_{GeLi,\,1}$ ($t_{R,NIST}$) = $(2.64 \pm 0.12)\times10^{-4}$, in agreement with $R_{GeLi,\,2}$ ($t_{R,NIST}$). It is interesting to note that the impurity content measured at the PTB and evaluated at the NIST reference date gives R_{PTB} ($t_{R,NIST}$) = $(2.72 \pm 0.24)\times10^{-4}$, which is in agreement with $R_{GeLi,\,2}$ within the uncertainty limits.

At the time of the measurements of the NIST ampoule in the SIR, the ^{177}Lu had decayed much more than the ^{177}Lu^{m} impurity. The SIR results are consequently more sensitive to the impurity than they were for the PTB measurements. Figure 2 shows that coherent results are obtained using the impurity content $R_{GeLi,\,2}$ and it has been agreed to use this value for the determination of the final NIST SIR result.

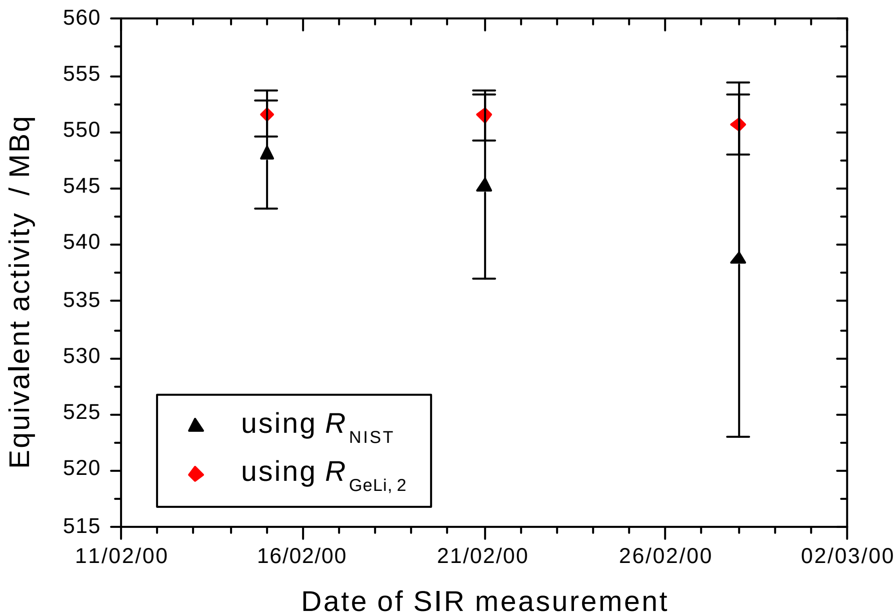

Figure 2. NIST SIR results using NIST and BIPM impurity contents and $A_{e,calc}$ for the correction factor C.

Final SIR results for ^{177}Lu

It is now possible to re-evaluate all the SIR results for ^{177}Lu using an experimental value for the equivalent activity of ^{177}Lu in the correction factor C. Using the mean of the PTB and NIST preliminary results, A_e = 555 MBq, gives the values shown in Table 2.

[b] These measurements were made using the same Ge(Li) detector but with a different ampoule-to-detector distance and at a different date (one month interval).

	Measurement date	A_e / kBq	$u(A_e)$ / kBq	^{177}Lu^m
PTB	26-01-2000	558 930	2 785	R_{PTB}
	31-01-2000	558 920	2 940	
	07-02-2000	558 495	3 320	
NIST	15-02-2000	551 210	2 060	$R_{GeLi, 2}$
	21-02-2000	550 820	2 230	
	28-02-2000	549 440	2 640	

Table 2: Final SIR results for ^{177}Lu. The highlighted lines correspond to the values to be registered.

Discussion and conclusion

The present case demonstrates the importance of measuring short-lived radionuclides in the SIR as rapidly as possible after preparation. This should reduce the influence of any possible long-lived impurity.

The SIR measurements of ^{177}Lu confirm that the PTB half-life value $T_{1/2}$ = (6.646 ± 0.005) d is an improvement on the earlier value given in the literature (NDS **68**, 747 (1993)).

The slightly decreasing values evident in the NIST results may be linked to the NDS **68**, 747 (1993) decay scheme used to calculate the equivalent activity of ^{177}Lu^m. More recent decay data are needed for this metastable state. An SIR measurement of pure ^{177}Lu^m would also provide a better equivalent activity value.

The final results show a 2-sigma discrepancy between the PTB and NIST results which merits investigation, especially as both laboratories based their standardization at least in part on the same method (CIEMAT/NIST). The low-energy photon spectrum emitted by ^{177}Lu was considered in terms of the effect of self-absorption in the ampoule, which is a function of the density of the solution. The ^{177}Lu solution was prepared by different methods at the NIST and at the PTB. However, the HCl concentration is at most 0.1 mol/L and the carrier concentration 20 µg/g at maximum. The density of both solutions remains very close to 1.000 g/cm^3. In consequence, no significant effect on the response of the I.C. is expected, as demonstrated by experimental observations made at the NIST and the BIPM (CCRI(II) report, 1999).

The equivalent activity calculated from the efficiency curve of the SIR I.C. is closer to the PTB result than to the NIST result. However, the $A_{e,calc}$ depends on the γ-emission probability values which were actually derived at the PTB. In consequence, the PTB SIR result and $A_{e,calc}$ are highly correlated.

The apparent discrepancy between the NIST and the PTB corresponds to 1.4 % relative difference which may be acceptable for such a short-lived and not commonly measured radionuclide.

References

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