First measurement of ^{153}Sm in the SIR

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In June 1998, the NIST sent to the International Reference System (SIR) a solution of ^{153}Sm standardized in a 4π ionization chamber. As this radionuclide had not previously been measured in the SIR, the resulting equivalent activity $A_{\mathrm{e,NIST}}$ is compared with the value calculated from the efficiency curve of the SIR. However, problems occurred owing to the presence of ^{154}Eu and ^{156}Eu impurities in the solution. The manner in which the final equivalent activity value for this solution of ^{153}Sm has been deduced is described in this report.

Calculated ^{153}Sm equivalent activity from the efficiency curve

The method of calculation of an equivalent activity from the efficiency curve of the SIR is described in Rytz, 1983. The uncertainty of the calculated equivalent activity $A_{\mathrm{e,calc}}$ was determined using three different decay schemes (NDS37, 1982; NDS83, 1998 and Bowles et al, 1998) for the γ-emission probabilities and two different efficiency curves (from Rytz and Müller, 1984 and Michotte, 1999a in which the differences between the two efficiency curves are described). Twenty gamma rays and three x-rays of the decay scheme of ^{153}Sm are included in the calculation. The results are summarized in the following table[1]:

Table 1: Comparison of $A_{\mathrm{e,calc}}$ for ^{153}Sm; results are given in kBq

^{153}Sm decay scheme	SIR efficiency curve	
	Michotte, 1999a	Rytz and Müller, 1984
NDS37, 1982	527 300 ± 5500	
NDS83, 1998	548 800 ± 6200	542 000
Bowles et al, 1998 (*)	570 500 ± 8000	

(*) The weak γ–ray emissions not given in this paper are completed using NDS83, 1998.

It is interesting to note that for radionuclides presenting a similar decay scheme, we have:

Table 2: Comparison between measured and calculated A_{e}

	Calculated A_{e} / kBq	Measured A_{e} (SIR mean) / kBq
^{153}Gd	371 000 ± 6000	366 100 ± 2000
^{155}Eu	470 200 ± 7400	478 400 ± 6800 (**)

(**) Only 1 entry in the SIR data base

These calculated values are in reasonable agreement with the measurements.

[1] All uncertainties given correspond to one standard uncertainty

SIR measurements of the NIST ^{153}Sm ampoule

The SIR measurement took place on 23 June 1998, the date of reference of the ^{153}Sm solution. It was repeated on 29 June. To determine the equivalent activity $A_{e,NIST}$ of the ampoule, a correction must be applied to account for the presence of impurities. It consists of dividing the measured ionization current, by the factor

$$C_i = 1 + \sum_k \frac{A_k}{A} \cdot \frac{A_e}{A_{e,k}} \cdot D_k$$

where A and A_k (A_e and $A_{e,k}$) are the activities (equivalent activities), at the reference date, of the main radionuclide and the impurity k respectively, and D_k is a decay correction (Michotte, 1999b). The half-lives used are (46.284 ± 0.004) h for ^{153}Sm, 3105 d for ^{154}Eu and 15.19 d for ^{156}Eu (uncertainties on the half-lives of impurities are neglected).

The ratios $R_k = A_k / A$ were given by the NIST on the SIR form for both impurities present in the solution:

^{154}Eu : R_{154} = (0.034 ± 0.002) % and ^{156}Eu : R_{156} = (0.126 ± 0.009) %.

The equivalent activities for ^{153}Sm and ^{156}Eu are not known experimentally and have to be determined from the efficiency curve of the SIR. The calculated results for ^{153}Sm are in Table 1. The equivalent activity $A_{e,156}$ of ^{156}Eu is estimated from the efficiency curve to be 15 330 ± 290 kBq, with the decay scheme parameters taken from NDS65,1992. Nuclides having a similar decay scheme are ^{152}Eu, ^{154}Eu and ^{166m}Ho for which the relative difference between the measured and calculated A_e values is smaller than 5×10^{-3} (Michotte, 1999a). In addition, ^{156}Eu emits beta particles with energies up to 2.45 MeV which contribute to the ionization current and should be taken into account. The relative correction to be applied is 1.8×10^{-3}, evaluated from the beta efficiency curve of the SIR ionization chamber measured using other beta emitters (Rytz and Müller, 1985 and Michotte, 1999c), giving :

$A_{e,calc}$(^{156}Eu) = 15 300 ± 290 kBq.

The results of the SIR measurements are shown in Table 3a with about 1.7 % discrepancy observed between the measurements carried out on 23 and 29 June 1998. An evaluation of the impurity content of the solution, made on 1 July using the BIPM Ge(Li) spectrometer (Michotte, 1999d) and the ^{153}Sm decay scheme in NDS83, 1998, gives different values for the ratios R_k, particularly for ^{156}Eu:

R_{154} = (0.037 ± 0.001) % and R_{156} = (0.093 ± 0.003) %.

The resulting equivalent activities $A_{e,GeLi}$ show a trend in the opposite direction and are also presented in Table 3a :

Table 3a: Results of SIR measurements of ^{153}Sm (given in kBq)

	First SIR measurement 23 June 1998	Second SIR measurement 29 June 1998
Input data for the calculation of C_i	$A_{e,153}$ = **548 800** kBq / $A_{e,154}$ = 13 750 kBq / $A_{e,156}$ = 15 300 kBq	
$A_{e,NIST}$ (use of NIST R_k values)	579 600 ± 4 800	589 700 ± 13 400
$A_{e,GeLi}$ (use of BIPM R_k values)	573 800 ± 4 400	562 300 ± 6 000

At this stage, the four results in Table 3a may be recalculated using the SIR value of the NIST (579 600 kBq) in the calculation of the correction for impurities C_i. This does not significantly change the results of the first SIR measurement, but increases both values of C_i calculated for the second measurement. In consequence, the consistency is improved when using the BIPM R_k values, while the discrepancy is enlarged when using the NIST R_k values (see Table 3b). However, this could be due to inaccurate $A_{e,153}$ and $A_{e,156}$ input values rather than to the R_k values.

Table 3b: Results of SIR measurements of ^{153}Sm (given in kBq)

	First SIR measurement 23 June 1998	Second SIR measurement 29 June 1998
Input data for the calculation of C_i	$A_{e,153}$ = **579 600** kBq / $A_{e,154}$ = 13 750 kBq / $A_{e,156}$ = 15 300 kBq	
$A_{e,NIST}$ (use of NIST R_k values)	581 400 ± 4 800	599 100 ± 14 300
$A_{e,GeLi}$ (use of BIPM R_k values)	575 200 ± 4 400	570 200 ± 6 300

Other analysis of the SIR measurements

In considering an alternative analysis, the quantity $A_{e,154}$ and the decay constants of ^{153}Sm, ^{154}Eu and ^{156}Eu are not brought into question. The constraint imposed is that the SIR results must stay constant with time:

$$M(t_1)\cdot\left[1+R_{154}\cdot\frac{A_{e,153}}{A_{e,154}}\cdot D_{154}(t_1)+R_{156}\cdot\frac{A_{e,153}}{A_{e,156}}\cdot D_{156}(t_1)\right]$$

$$=M(t_2)\cdot\left[1+R_{154}\cdot\frac{A_{e,153}}{A_{e,154}}\cdot D_{154}(t_2)+R_{156}\cdot\frac{A_{e,153}}{A_{e,156}}\cdot D_{156}(t_2)\right]\ ,$$

where, $M(t_1)$ and $M(t_2)$ are the two SIR measurements without any correction for impurity. It is then possible to deduce a value for the product $P = R_{156}$ x $A_{e,153}$ / $A_{e,156}$ which slightly depends on the R_{154} and $A_{e,153}$ values taken for the correction for the ^{154}Eu contribution. The SIR result, which is now the same at both dates, is then obtained. The calculations are presented in the following table using five input values of $A_{e,153}$ from Tables 1 and 3b.

Table 4: Results for A_e of ^{153}Sm with the constancy constraint

$A_{e,153}$ input value / kBq	Calculations using the NIST R_k values			Calculations using the BIPM R_k values		
	P	$A_{e,NIST}$ / kBq	$A_{e,156}$ / kBq	P	$A_{e,GeLi}$ / kBq	$A_{e,156}$ / kBq
527 300	0.0412	577 100	16 100	0.0396	576 900	12 400
548 800	0.0404	577 000	17 100	0.0388	576 700	13 200
570 500	0.0397	576 900	18 100	0.0380	576 600	14 000
575 200	0.0395	576 800	18 300	0.0378	576 600	14 200
581 400	0.0393	576 800	18 600	0.0376	576 500	14 400

An important conclusion is that the SIR result is now almost independent of the ratios R_k and of the value taken for $A_{e,153}$ to calculate the correction for impurities. The highlighted line in Table 4 is the most coherent one as the input $A_{e,153}$ value is closest to the output SIR result. This $A_{e,153}$ input value is that obtained using the BIPM R_k values (Table 3b). The corresponding SIR result is in agreement with $A_{e,calc}$ (Table 1) estimated from the new SIR efficiency curve using the ^{153}Sm partial decay scheme presented in Bowles et al., 1998.

In addition, the equivalent activity of ^{156}Eu is determined from P, $A_{e,153}$ and R_{156} and can be compared with $A_{e,calc}(^{156}Eu)$ given above: neither NIST nor BIPM R_k values give $A_{e,156}$ compatible with the expected range. An intermediate R_{156} value seems to be necessary.

This second analysis of the ^{153}Sm SIR data gives indications coherent with the measurements of this ampoule made at the NIST. Indeed increasing values were also observed in the NIST ionization chamber and the product (R_{156} x response of the NIST chamber for ^{156}Eu) had to be reduced by a factor of 0.825 to obtain a measurement constant with time. The corresponding factor for the SIR data is 0.836 (using the NIST R_k values). In consequence, it is likely that the cause of the increase observed in both NIST and SIR measurements is the NIST R_{156} value. It seems reasonable for the NIST to correct the R_{156} value by a factor of 0.830 ± 0.005, giving R_{156} = (0.105 ± 0.008) %. From this, $A_{e,156}$ may be determined as in Table 4, giving 15 300 kBq, in excellent agreement with $A_{e,calc}(^{156}Eu)$.

Conclusion

The final SIR results calculated as usual are the following:

input data for the calculation of C_i: A_e of ^{153}Sm: 576 800 kBq
^{154}Eu: 13 750 kBq
^{156}Eu: 15 300 kBq
R_{154} = (0.034 ± 0.002) % and R_{156} = (0.105 ± 0.008) %.

$A_{e,NIST}$ (^{153}Sm) = (576 900 ± 4 700) kBq measured on the 23 June 98
(577 200 ± 12 500) kBq ” 29 June 98.

References

Bowles, N.E. et al., 1998. *Standardisation of ^{153}Sm*. Appl. Rad. Isot. **49**, pp 1345-1347.

Michotte, C., 1999a. *Update of the efficiency curve of the SIR*. Rapport BIPM, in preparation.

Michotte, C., 1999b. *Influence of radioactive impurities on SIR measurements.* ICRM99 communication, Prague, accepted for publication in Appl. Rad. Isot.

Michotte, C., 1999c. *Beta efficiency curve of the SIR ionization chamber*. Rapport BIPM-99/8.

Michotte, C., 1999d. *Efficiency calibration of the Ge(Li) detector of the BIPM for SIR-type ampoules*. Rapport BIPM-99/3.

NDS37, 1982: M.A. Lee in Nuclear Data Sheets **37,** p. 487.

NDS83, 1998: R.G. Helmer in Nuclear Data Sheets **83**, p. 285.

Rytz, A., 1983. *The international reference system for activity measurements of γ-ray emitting nuclides.* Appl. Rad. Isot. **34**, pp 1047-1056.

Rytz, A. and Müller, J.W., 1984. *Système international de référence pour la mesure d'activité d'émetteurs de rayons γ (SIR).* Procès-verbaux de la 73e session du CIPM, **52**, ed. BIPM, Sèvres, France, pp 76-77.
Rytz, A. and Müller, J.W., 1985. *Système international de référence pour la mesure d'activité d'émetteurs de rayons γ (SIR).* Procès-verbaux de la 74e session du CIPM, **53**, ed. BIPM, Sèvres, France, pp 100-101.