Draft paper submitted to ICRM2001

Efficiency curve of the ionization chamber of the SIR

C. Michotte

Bureau International des Poids et Mesures (BIPM), Pavillon de Breteuil, F-92312 Sèvres Cedex, France

The efficiency of the SIR ionization chamber versus photon energy has been obtained by an iterative method starting from a few monoenergetic gamma-ray emitters. The relative standard uncertainty on the efficiency curve obtained is lower than 10^{-2} above 65 keV. An efficiency curve for β particles has been deduced in a similar manner from measurements of (quasi-) pure beta emitters. The possible uses of the efficiency curve to identify inconsistent SIR data or discrepant decay scheme data are discussed.

Introduction

The SIR (International Reference System for activity measurements of γ-ray emitting nuclides) was started in 1976 at the BIPM as a complement to the international comparisons of activity measurements organized by Section II of the CCRI (Comité Consultatif des Rayonnements Ionisants). The participating national metrology institutes (NMIs) submit SIR glass ampoules containing their standardized solutions of radionuclides to the BIPM where the current produced by these samples in a 4π-ionization well chamber (I.C.) is compared with the current obtained with a ^{226}Ra reference source. The chamber is of the Centronic 20th Century IG11 type, filled with N_2 at a pressure of 2 MPa and having steel walls of about 0.8 mm thickness in the well. A detailed description of the SIR is presented in Rytz (1978 and 1983). The simplicity and rapidity of the measurements as well as the long-term stability of the ionization chamber has allowed the comparison over 25 years of hundreds of radioactive solutions for a total of about 60 radionuclides.

Each SIR measurement may be considered as a measurement of the efficiency of the I.C. for a given radionuclide (Schrader, 1997; Reher, 1998). The construction of the efficiency curve, i.e. the plot of the efficiency versus γ-ray energy, is detailed in the first part of the present paper. This curve is required for the calculation of the efficiency of the I.C. for radionuclides not previously measured in the SIR. This is needed to evaluate the correction for a gamma-emitting impurity present in an ampoule when the efficiency for this impurity is not known experimentally (Michotte, 2000), or to give a point of comparison when a radionuclide is measured in the SIR for the first time.

Some of the radionuclides considered are β emitters. The consequent possible bremsstrahlung in the wall of the I.C. contributes to the measured ionization current and must be taken into account in establishing the efficiency curve. In consequence, a β-efficiency curve of the I.C. has been constructed using measurements of seven quasi-pure β emitters, as presented in part 2.

In the final parts of the paper, the experimental and calculated photon efficiencies are compared and discussed. A discussion considering the possible use of the γ-efficiency curve for the identification of outliers in the SIR data concludes that great care must be taken in adopting this approach.

Construction of the SIR photon-efficiency curve

An SIR measurement of a radioactive solution is expressed in terms of an equivalent activity A_e which corresponds to the activity of the solution that would produce the same ionization current as the SIR reference ^{226}Ra source. The inverse of the equivalent activity is proportional to the efficiency of the ionization chamber for this radionuclide. Therefore

$$\frac{1}{A_\mathrm{e}} = \sum_i e(E_i)\cdot P_i + \sum_j e_\mathrm{ß}(E_j)\cdot P_{\mathrm{ß},j} \qquad (1),$$

where $e(E_i)$ and $e_\beta(E_j)$ are the efficiencies of the SIR I.C. for each of the photons and β-particles emitted by the nuclide considered, with corresponding emission probabilities P_i and $P_{\beta,j}$. The beta efficiency is order of magnitudes lower than the photon efficiency. The second term in equation (1) is thus negligible for most radionuclides (see part 2).

The method used to determine $e\,(E)$ is described by Rytz (1983) and by Schrader (2000). It consists of an iterative process starting with a few monoenergetic gamma-emitters for which the gamma efficiency can be determined directly. For multiple gamma emitters, the efficiency e_n for the gamma-ray making the largest contribution to the ionization current is deduced from $1/A_\mathrm{e}$ by subtracting the contributions of the other (n - 1) photons. In consequence, at each iteration k, a polynomial and/or exponential fit $e^k\,(E)$ to the efficiency values $e_n^{\,k}$ allows the interpolation and extrapolation at any energy value and thus, the calculation of a new efficiency $e_n^{\,k+1}$ for each radionuclide :

$$e_n^{\,k+1} = \frac{1/A_\mathrm{e} - \sum_{i=1}^{n-1} e^k(E_i)\cdot P_i - \sum_j e_\mathrm{ß}(E_j)\cdot P_{\mathrm{ß},j}}{P_n} \quad . \qquad (2)$$

For each radionuclide, the input data are the emission probabilities (from IAEA (1991), supplemented with the most recent Nuclear Data Sheet), the beta efficiencies (see part 2) and the median value A_e of the experimental equivalent activities available. Some SIR data were withdrawn from the evaluation of the median in order to limit the influence of any NMI which had participated several times at the SIR for the same radionuclides. The results of the CCRI international comparisons of activity measurements of ^{75}Se, ^{109}Cd, ^{133}Ba and ^{137}Cs solutions were included in the evaluation of A_e. The radionuclides considered for the determination of $e\,(E)$ verify the following conditions: radionuclides in liquid solution only (no gas for which the self-attenuation effect would be different); participation in the SIR of at least two NMIs; β-particles'contribution lower than 5×10^{-3} in relative terms, to limit the uncertainty arising from the β-efficiency curve (see part 2); β^+-branching ratio lower than 10 %. The latter condition is imposed because the emission points of annihilation γ-rays can be outside the ampoule, resulting therefore in a detection efficiency that differs from that of other γ-rays. This leads to a set of 40 radionuclides to determine $e\,(E)$.

Although an efficiency $e_n^{\,k}$ could be, in principle, evaluated for all the photons emitted by a given nuclide, only one energy E_n per nuclide is used in practice, in order to avoid correlations. As notified above, this γ-ray energy usually corresponds to the largest contribution to the ionization current. However, in some cases like ^{141}Ce, ^{153}Gd, ^{201}Tl and ^{133}Ba, a weaker photon emission has been selected in order to distribute the data more evenly over the whole energy range, especially at low energy.

To reduce the correlation between iteration k and $k+1$, all the γ-rays close[1] to E_n for a given nuclide were considered as a single photon with an energy E_m and a photon emission probability P_m :

$$E_m = \sum_k E_k \cdot P_k \Big/ \sum_k P_k \qquad\qquad P_m = \sum_k P_k \qquad (3)$$

This is the case for ^{60}Co, among many others, where eq. (2) strongly relates the efficiency at 1.33 MeV (e_2^{k+1}) to the efficiency at 1.17 MeV from the previous iteration:

$$e_2^{k+1} = \frac{1/A_\mathrm{e} - e^k(1.17\,\mathrm{MeV}) \cdot P_1}{P_2}$$

Considering a 'mean photon energy' as defined by (3), ^{60}Co corresponds to a mono-energetic photon emitter for which $$e(E_m) = \frac{1/A_\mathrm{e}}{P_m}$$

and the correlation problem is solved.

A correlation between the emission probabilities and the experimental activities may exist when an NMI calibrating an I.C. uses the emission probabilities determined from its own activity standards (Schrader, 2000). This is widely reduced in the present case because on the one hand recommended emission probabilities (which are weighted means of several measurements) have been used, and on the other hand, the input A_e values are medians of independent measurements from different NMIs.

The final iteration is presented in Figure 1a. The plotted f factor was defined by Rytz (1983) as

$$f(E) = \frac{e(E)}{6 \times 10^{-8}\, E} \quad .$$

The order of the polynomial has been selected such that the reduced chi-square of the fit is minimized. The polynomial fit above 3 MeV cannot obviously be used to estimate the efficiency of the SIR chamber up to 4 MeV, as needed for radionuclides like ^{56}Co. In consequence, an exponential fit to the last 10 data points has been performed. The transition from the polynomial to the exponential occurs at the point where both fits are identical. The relative residuals presented in Figure 1b do not show any energy-dependent structure. The cut-off in efficiency occurs at around 30 keV.

The uncertainties shown on the graph are the result of the propagation in (2) of the uncertainties on the experimental A_e value, the emission probabilities and the uncertainty of the fitted curve from the previous iteration. The uncertainty budget for the ^{139}Ce case is given in Table 1 as an example.

[1] those "close" photons were selected arbitrarily, taking into account the curvature of the efficiency curve.

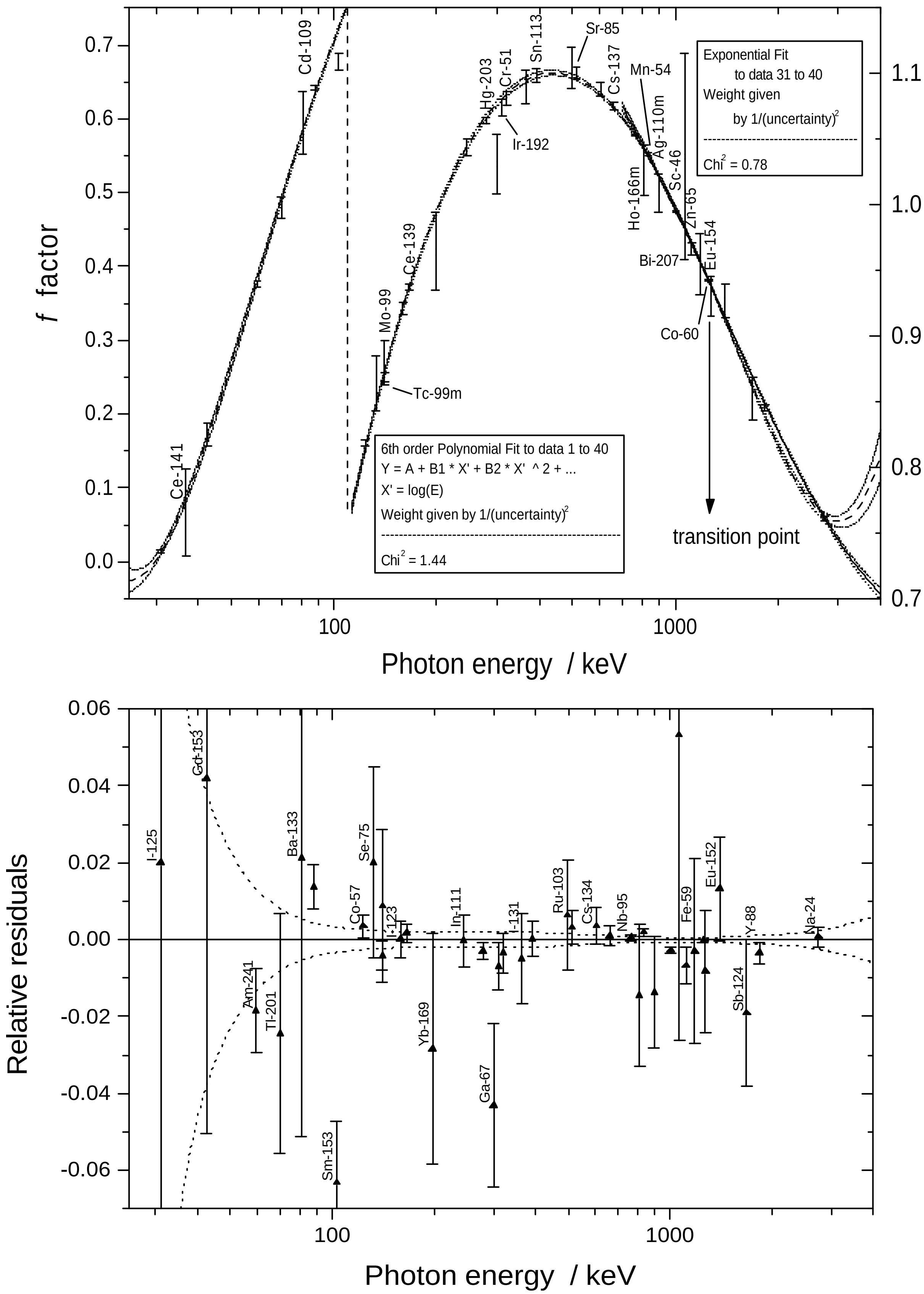

Fig. 1. SIR photon efficiency curve (*f* factor as defined in the text): polynomial (dashed line) and exponential (full line) fit to the e_n data and relative residuals. The dotted lines show the standard uncertainty of the fitted curve.

Table 1. Example of uncertainty budget.

Quantity	Relative stand. uncert.	Contribution to the uncertainty of the f factor for ^{139}Ce (E_n = 165.9 keV) f = 0.9379
efficiency for 34 keV x-rays	8.9×10^{-2}	2.0×10^{-3}
x-rays emission probability	3×10^{-2}	2.3×10^{-4}
γ-ray emission probability	7.5×10^{-4}	7.0×10^{-4}
Equivalent activity	9.0×10^{-4}	8.6×10^{-4}
		Quadr. sum: 2.3×10^{-3}

Construction of the SIR b-efficiency curve

The method is similar to that used for the photon efficiency curve e (E). The radionuclides selected are (quasi-)pure β emitters (first-forbidden transitions, except ^{32}P) and the small photon contribution to the ionization current is evaluated from e (E) and subtracted :

$$e_{ß,n}^{\;k+1} = \frac{1/A_e - \sum_{j=1}^{n-1} e_ß^{\;k}(E_{ß,j}) \cdot P_{ß,j} - \sum_i e(E_i) \cdot P_i}{P_{ß,n}} \quad , \qquad (4)$$

where $E_{β,j}$ is the mean energy of each β spectrum. There are only 7 radionuclides available, with SIR measurements much less precise than for γ emitters. The efficiency curve appears to be exponential as shown in Figure 2. The ^{106}Ru datum point is not used in the fit as it does not seem to lie on the same line as the other points, but much higher. This could be explained by the presence of very high-energy beta particles emitted by ^{106}Rh (end point of the beta spectrum 3.54 MeV) which, in addition to the production of bremsstrahlung, are likely to reach the sensitive volume of the chamber. The ^{85}Kr datum point is shown for information but is not used in the fit as the self-absorption effect in a gas will differ from that in a liquid solution.

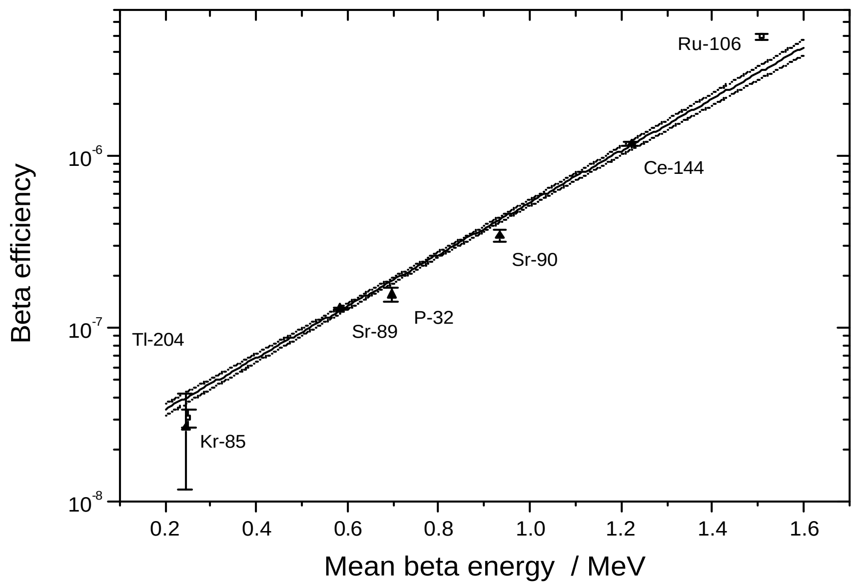

Fig. 2. SIR beta efficiency curve: exponential fit to the black data points. The dotted lines show the standard uncertainty of the fitted curve.

The contribution of the beta-particles to the ionization current in the SIR I.C. can be non-negligible in comparison with the photon contribution when the latter is small owing to the low energy of the emitted photons and/or low emission probability. Such radionuclides are listed in Table 2.

Table 2. Relative contribution of β particles to the ionization current in the SIR I.C.

Radionuclide	E_{b} **/MeV**	A_{e} ´ $\sum e_{ß} \cdot P_{ß}$
^{140}Ba	0.49	0.001 0
^{131}I	0.19	0.001 2
^{124}Sb	0.92	0.001 2
^{47}Ca [a]	0.24	0.001 4
^{47}Sc	0.20	0.001 8
^{156}Eu [a]	0.97	0.002 0
^{18}F [b]	0.63	0.002 3
^{99}Mo	0.44	0.004 2
^{68}Ge [b]	0.84	0.004 3
^{153}Sm	0.26	0.004 8
^{56}Mn	1.2	0.006 9
^{86}Rb [a]	0.71	0.032
^{186}Re [b]	0.36	0.057
^{170}Tm [a]	0.32	0.31

a : impurities b : possible SIR canditates

Discussion

Using (1), a consistency check was performed by calculating the 40 input equivalent activities from the efficiency curves e (E) and e_{β} (E). The results are given in Table 3. A two-sigma discrepancy between the experimental and calculated equivalent activities (A_{e} and $A_{\mathrm{e,c}}$, respectively) is observed only for ^{153}Sm (see below). The calculation of $A_{\mathrm{e,c}}$ for ^{125}I and ^{139}Ce is very sensitive to the low-energy part of e (E). The agreement noted for both nuclides in Table 3 gives confidence in the efficiency curve obtained. Considering the case of ^{124}Sb, the same conclusion applies for the high-energy part of the curve. The $A_{\mathrm{e,c}}$ value for ^{153}Sm is also sensitive to the low-energy part of e (E). However, fluctuations within acceptable limits in the curve near the cut-off energy could not resolve the large discrepancy noted above for this nuclide.

In Figure 3 the present photon efficiency curve is compared with the previous evaluation by Rytz and Müller (1984), where the curve was drawn by hand. The agreement is surprisingly good in view of the large changes which have occurred since 1984: new evaluations of emission probabilities and the number of SIR measurements have increased by a factor of 2. This emphasizes the stability and accuracy of the photon efficiency curve of the SIR.

Table 3. Ratio (*R*) of calculated and measured equivalent activities for the 40 radionuclides used in the determination of *e* (*E*).

Nuclide	*R* - 1	st. uncertainty	Nuclide	*R* - 1	st. uncertainty
^{24}Na	0.0005	0.0022	^{123}I	-0.0003	0.0044
^{46}Sc	0.0001	0.0011	^{124}Sb	-0.0077	0.0078
^{51}Cr	-0.0031	0.0055	^{125}I	-0.0407	0.1200
^{54}Mn	0.0018	0.0012	^{131}I	-0.0042	0.0095
^{57}Co	0.0037	0.0037	^{133}Ba	0.0000	0.0032
^{59}Fe	-0.0013	0.0233	^{134}Cs	0.0017	0.0019
^{60}Co	-0.0001	0.0006	^{137}Cs	0.0011	0.0028
^{65}Zn	-0.0064	0.0048	^{139}Ce	0.0003	0.0024
^{67}Ga	-0.0160	0.0080	^{141}Ce	-0.0026	0.0058
^{75}Se	0.0039	0.0054	^{152}Eu	0.0036	0.0037
^{85}Sr	0.0029	0.0046	^{153}Gd	0.0056	0.0182
^{88}Y	-0.0022	0.0019	^{153}Sm	-0.0491	0.0119
^{95}Nb	0.0009	0.0012	^{154}Eu	-0.0029	0.0057
^{99}Mo	0.0037	0.0085	^{166}Ho^{m}	-0.0054	0.0067
^{99}Tc^{m}	-0.0042	0.0044	^{169}Yb	-0.0094	0.0098
^{103}Ru	0.0059	0.0132	^{192}Ir	-0.0038	0.0033
^{109}Cd	0.0135	0.0073	^{201}Tl	-0.0115	0.0150
^{110}Ag^{m}	-0.0049	0.0055	^{203}Hg	-0.0029	0.0029
^{111}I	-0.0003	0.0042	^{207}Bi	0.0273	0.0406
^{113}Sn	0.0006	0.0050	^{241}Am	-0.0189	0.0171

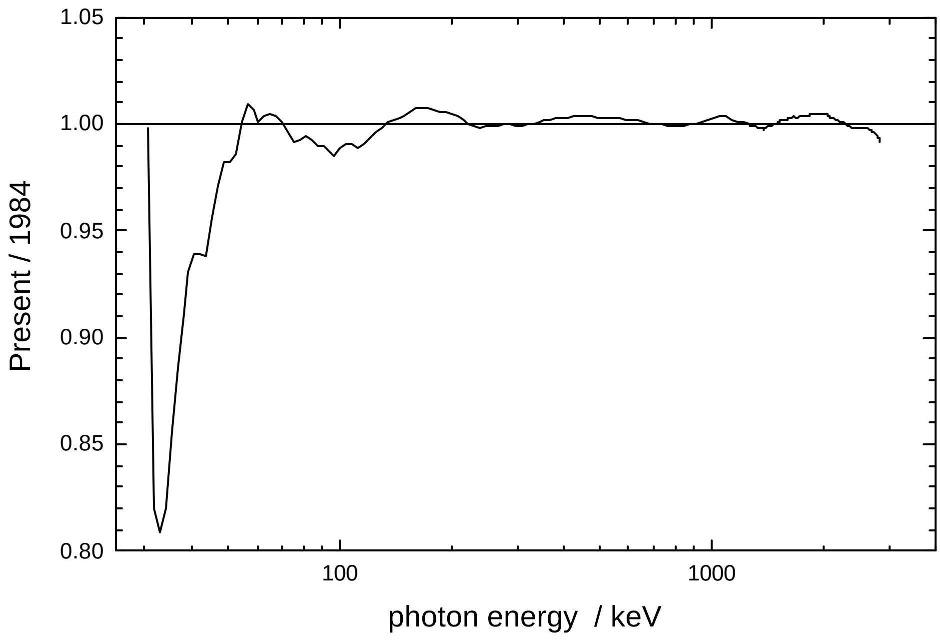

Fig. 3. Ratio of the present photon efficiency curve and the Rytz and Müller (1984) curve.

Applications

The SIR efficiency curve is required for the calculation of the equivalent activity to give a point of comparison when a radionuclide is measured in the SIR for the first time. This was the case for ^{47}Sc, ^{56}Mn, ^{140}Ba, ^{155}Eu, ^{177}Lu, ^{182}Ta, ^{195}Au, ^{203}Pb, ^{228}Th, ^{237}Np and ^{243}Am, for which the SIR measurement is in agreement with $A_{e,c}$ within twice the standard uncertainties (except the last three nuclides for which more investigations are needed[2]).

The SIR efficiency curve $e\,(E)$ can also be used to identify radionuclides that might present problems in their decay scheme or standardization methods. Indeed, when the efficiency e_n for a given radionuclide is not in agreement with $e\,(E)$ it should mainly[3] arise from inaccuracies in the emission probability or A_e values. In the case of ^{153}Sm, when the Bowles (1998) emission probabilities are used instead of the Nuclear Data Sheets 1998 evaluation, the residual shown in Figure 1b is reduced to (-0.013 ± 0.020) which is then consistent with zero. Another example is ^{67}Ga, as the emission probability for the γ-ray at 93.3 keV has been measured recently (Simpson, 2000) giving a value much more precise than in the Nuclear Data Sheets 1991 evaluation. Again, the new residual (-0.024 ± 0.018) is closer to zero, suggesting that the whole decay scheme of ^{67}Ga should be reviewed.

The SIR efficiency curve may also help in the interpretation of the SIR results, as shown for ^{124}Sb in Table 4. A first look at the data leads to the conclusion that the second laboratory has a biased result. After comparison with $A_{e,c}$ (using the 1997 Nuclear Data Sheet evaluation of the emission probabilities), it may be concluded that the results are scattered around the exact value and that the uncertainties quoted by the first two laboratories may be underestimated.

Table 4: SIR measurements of ^{124}Sb

Participant, date of measurement	A_e / kBq	u(A_e) / kBq
Lab. 1, 1992	9539	23
Lab. 2, 1995	9365	47
Lab. 3, 1998	9590	110
Median	9539	71
Calculation with $e\,(E)$	9466	23

A direct possible application of the SIR efficiency curve could be to use the $A_{e,c}$ as reference values and, eventually, designate as outliers the experimental A_e values which lie apart from this reference. This has to be carried out very carefully, first because any $A_{e,c}$ value depends of course on the emission probabilities used in the calculation. Second, because in an energy range where $e\,(E)$ is not strongly constrained by many precise data points, there may be a large correlation between a given input A_e value and the fitted curve, consequently resulting in a correlation with $A_{e,c}$. This is obvious for ^{24}Na which completely determines $e\,(E)$ at high energy: the calculated $A_{e,c}$ from $e\,(E)$ will inevitably be in excellent agreement with the median A_e of the ^{24}Na SIR data whatever its accuracy. Another example is ^{241}Am as illustrated in Figure 4. The ^{241}Am datum point e_n has been re-evaluated by replacing the A_e input value

[2] The discrepancy for ^{237}Np is resolved when using emission probabilities from Woods (2001).

[3] It may also be related to possible high-energy β particles (> 2 MeV) like for ^{106}Ru.

by A_e', a value deliberately chosen in disagreement with the SIR data. The low-energy part of the fitted *e* (*E*) is shifted accordingly and the new calculated value $A_{e,c}$' for ^{241}Am is found to be in agreement with the discrepant A_e' input data. This shows that for ^{241}Am, the comparison of $A_{e,c}$ with the SIR experimental data is meaningless due to their correlation.

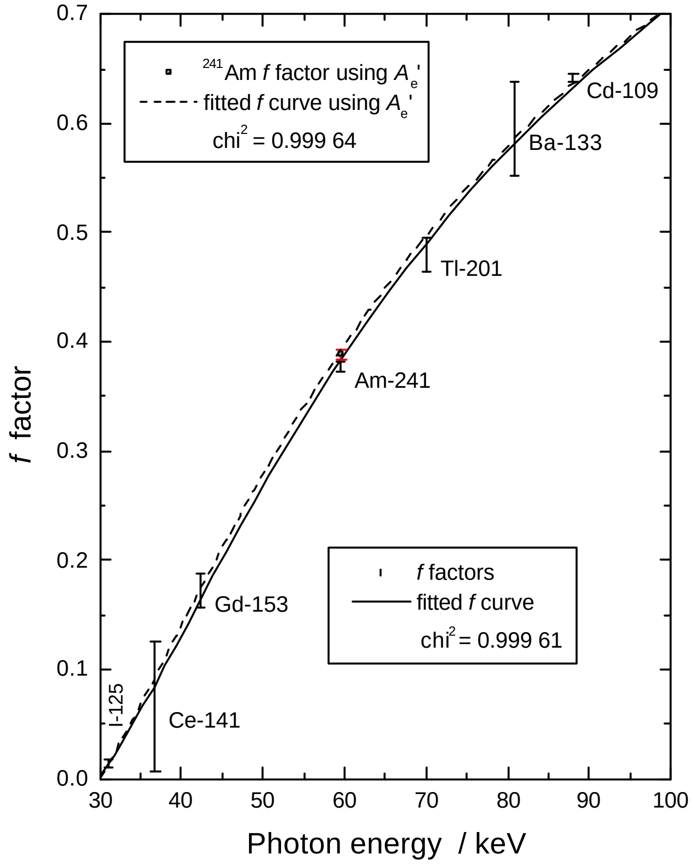

Fig. 4. Low-energy part of the SIR photon efficiency curve: influence of ^{241}Am data point on the fitted curve (see text).

Conclusion

The SIR photon efficiency curve has been revised and a relative standard uncertainty lower than 10^{-2} was reached above 65 keV. The contribution to the ionization current of the possible β emission has been taken into account. This updated curve will be used to evaluate the correction for any gamma-emitting impurity present in an SIR solution when the efficiency for this impurity is not known experimentally, or to give a point of comparison when a radionuclide is measured in the SIR for the first time. The utility of the SIR efficiency curve for the interpretation of the SIR data or to point out problems in the decay scheme data of ^{67}Ga, ^{153}Sm and ^{237}Np for example, has been shown. The low-energy part of the curve could be improved in the future by making measurements for ^{125}Te^m, ^{129}I, ^{131}Cs, ^{178}W and ^{210}Pb. Monte Carlo simulations (Gostely, 2000) could also help to provide a better understanding of the SIR efficiency for γ-rays above 2 MeV, annihilation γ-rays, high-energy β-particles and gaseous radionuclides.

Acknowledgements-The author wishes to thank C. Colas for its technical assistance and the safekeeping of the SIR archives, G. Ratel for his experience in SIR calculations and M.-N. Amiot-Péron, M. Cox, J. Keightley, B.R.S. Simpson and W. Wöger for many useful discussions.

References

Bowles N.E.et al., 1998. Standardization of ^{153}Sm. Appl. Rad. Isot. 49 (9-11), 1345-1347.

IAEA, 1991. TECDOC-619, Vienna.

Michotte, C., 2000. Influence of radioactive impurities on SIR measurements. Appl. Rad. Isot. 52 (3), 319-323.

Reher, D.F.G. , Woods, M.J., Simpson, B.R.S., Ratel, G., 1998. Portability of the calibration of SIR of BIPM to other ionization chambers for radioactivity measurements. Appl. Rad. Isot. 49 (9-11), 1417-1419.

Rytz, A., 1978. International coherence of activity measurements. Environment International 1, 15-18.

Rytz, A., 1983. The international reference system for activity measurements of γ-ray emitting nuclides. Appl. Rad. Isot. 34 (8), 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 73^{e} session du CIPM, 52, ed. BIPM, Sèvres, France, pp. 76-77.

Schrader, H., 1997. Activity Measurements with Ionization Chambers. Monographie BIPM-4, Sèvres, France, pp. 40-47 and pp. 62-63.

Schrader, H., 2000. Calibration and consistency of results of an ionization-chamber secondary standard measuring system for activity. Appl. Rad. Isot. 52 (3), 325-334.

Simpson, B.R.S. and Ntsoane, T.P., 2000. Decay scheme of ^{67}Ga. Appl. Rad. Isot. 52 (3), 551-556.

Woods, M. et al., 2001. Standardization and decay data of ^{237}Np. Submitted to the ICRM2001 conference in Braunschweig.