Standardization of ^{55}Fe by tracing method

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Abstract

This work describes the procedure followed by the Laboratório de Metrologia Nuclear (LMN) for the standardization of ^{55}Fe by the tracing method. This technique was applied using two radionuclides, which decay by the electron capture process followed by a prompt gamma-ray, namely ^{51}Cr and ^{54}Mn, as tracers. The calibration was performed in a $4\pi\beta$–γ coincidence system. The efficiency was obtained by selecting a gamma-ray window set at the 320 keV total absorption peak for ^{51}Cr and at 834 keV for ^{54}Mn.
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1. Introduction

^{55}Fe decays with a half-life of 1101.1 days (Bé, 2004) by the electron capture process to the ground state of ^{55}Mn emitting X-rays with energy around 5 keV. This characteristic makes ^{55}Fe a suitable radionuclide standard for X-ray spectrometers. However, a standard source needs a primary method for its standardization, which is not an easy task, due to its decay scheme.

For this reason, ^{55}Fe was selected by the Comité Consultatif pour les Etalons de Mesures des Rayonnements Ionisants (CCEMRI) of the Bureau International des Poids et Mesures (BIPM) for an international comparison, to be standardized by several national metrological institutes.

The Laboratório de Metrologia Nuclear (LMN) at the IPEN-CNEN/SP, from São Paulo, participated in this comparison in collaboration with the Laboratório Nacional de Metrologia das Radiações Ionizantes (LNMRI), from Rio de Janeiro.

Independent results, using different techniques, were obtained by each of these laboratories and included in the comparison. This work describes the procedures followed by the LMN for the standardization of this radionuclide.

For the present calibration, the tracing technique was applied using, as a tracer, two radionuclides which decay by the electron capture process followed by prompt gamma-ray emission, namely ^{51}Cr and ^{54}Mn. The calibration was performed in a $4\pi\beta$–γ coincidence system. The efficiency was obtained by selecting a gamma-ray window set at 320 keV total absorption peak for ^{51}Cr and at 834 keV for ^{54}Mn.

Measurements with 1 and 2 aluminum (Al) foils, with a thickness each of 150 μg cm^{-2} on both sides of the mixed sources, were performed to extrapolate the counting efficiency to that for zero thickness of Al foil. The ratio between the results obtained from 1 and 2 Al foils was applied in order to obtain the result without the absorber.

The intrinsic efficiency for each X-ray energy was determined from the experimental efficiencies and from the X-ray emission probabilities per decay. An iterative procedure was developed, which gave rise to the effective track length in the 4π detector gas. This parameter, together with the gas absorption coefficients, yielded the efficiency for the ^{55}Fe.

2. Experimental method

2.1. Source preparation

Known aliquots of solutions of the tracers ^{51}Cr and ^{54}Mn, previously standardized in the $4\pi\beta$(PC)–γ coincidence

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system, were mixed with known aliquots of ^{55}Fe solution. The sources of ^{55}Fe + ^{51}Cr and the sources of ^{55}Fe + ^{54}Mn were prepared by dropping known aliquots of each radio-active mixed solution on a 20-µg cm^{-2}-thick collodion film. This film had been previously coated with a 10-µg cm^{-2}-thick gold layer in order to make the film conductive. A seeding agent (Ludox) was used to improve the uniformity of the deposit and the sources were dried in desiccators. The source masses and the solution aliquots were accurately determined by the pycnometer technique (Campion, 1975) in a Sartorius MC 21S balance.

2.2. Coincidence system

A conventional 4πX–γ coincidence system was used, consisting of a 4π proportional counter, operated at 0.1 MPa with a P-10 gas mixture (90% argon: 10% methane), coupled to a pair of 3″ × 3″ NaI(Tl) crystals. The events were registered by the method developed at LMN, which makes use of a time-to-amplitude converter (TAC) associated with a multi-channel analyzer (Koskinas et al., 2006). The gamma window was set by selecting the gamma-rays of the tracers, namely 320 keV for ^{55}Fe + ^{51}Cr sources and 834 keV for ^{55}Fe + ^{54}Mn.

The number of detected events in the proportional counter is given by

$$N_{(X,A)} = N_{0Fe}\varepsilon_{(X,A)Fe} + N_{0T}(\varepsilon_{(X,A)_T} + C(1 - \varepsilon_{(X,A)_T})) \quad (1)$$

The number of gamma-detected events in the scintillator is given by

$$N_\gamma = N_{0T}\varepsilon_\gamma \quad (2)$$

and the number of coincident events is given by

$$N_c = N_{0T}\varepsilon_\gamma\varepsilon_{(X,A)} \quad (3)$$

and

$$N_c/N_\gamma = \varepsilon_{(X,A)_T} \quad (4)$$

resulting in

$$\frac{N_{(X,A)}N_\gamma}{N_c} - N_{0T}\left(1 + C\frac{(1 - \varepsilon_{(X,A)_T})}{\varepsilon_{(X,A)_T}}\right) = N_{0Fe}f\varepsilon, \quad (5)$$

where *Fe* and *T* indices stand for ^{55}Fe and tracer, respectively; N_{0Fe} is the ^{55}Fe disintegration rate; N_{0T} is the tracer disintegration rate; $N_{(X,A)}$ is the combined counting rate due to the Auger electrons and X-rays of the ^{55}Fe and of the tracer; N_γ is the gamma counting rate of the tracer; N_c is the coincidence counting rate; $\varepsilon_{(X,A)_T}$ is the effective detection efficiency for the combination of Auger electrons and X-rays of the tracer; ε_γ is the gamma detection efficiency of the tracer in the gamma detector; C is the tracer disintegration scheme constant, obtained in previous measurements by extrapolation technique.

The correction factor for ^{55}Fe efficiency is given by

$$f_\varepsilon = \varepsilon_{(X,A)_{Fe}}/\varepsilon_{(X,A)_T}. \quad (6)$$

Suitable corrections for mass, background, decay, dead time and resolution time were included in the count-rates.

Usually, when the tracing method is applied, the extrapolation technique is used and the disintegration rate is obtained when the inefficiency parameter goes to zero (Baerg et al., 1964; Dias and Koskinas, 2003). Nevertheless, due to the variation of the absorption and attenuation coefficient, of each X-ray energy from different nuclides, this extrapolation was not possible. To solve this problem, measurements were carried out using one and two Al absorbers on both sides of the mixed sources with approximately 150 µg cm^{-2} thickness, which is thick enough to absorb all the Auger electrons, in order to extrapolate the efficiency to zero Al foil thickness.

The ^{55}Fe X-ray energy is different with respect to the tracers; therefore, the efficiencies obtained with one and two layers of Al foils for the tracers, ^{51}Cr ($Z = 23$) and ^{54}Mn ($Z = 24$), are also different from ^{55}Fe efficiency. To determine ^{55}Fe efficiency, it was assumed that the counting efficiency for each nuclide is linearly dependent on the Z-number. This may be expressed as $\varepsilon_{(Z=25)} - \varepsilon_{(Z=24)} = \varepsilon_{(Z=24)} - \varepsilon_{(Z=23)}$. This can be rearranged to give the relationship

$$\varepsilon_{(Z=25)} = 2\varepsilon_{(Z=24)} - \varepsilon_{(Z=23)} \ . \quad (7)$$

Using this linear relationship, it is possible to determine the ^{55}Fe efficiency for one and two Al foils and calculate the correction factor f_ε (Eq. (6)) to correct the activity obtained with the tracers for the ^{55}Fe efficiencies.

Using these corrected activities for one and two absorbers, the activity extrapolated to zero absorber was

Table 1
Efficiencies and experimental activity values of ^{55}Fe obtained with ^{51}Cr as a tracer with one and two Al absorbers

Source	Number of Al absorbers	$Nc/N\gamma$	Uncorrected 55Fe activity, $N_{0Fe}f_\varepsilon$ (Eq. (5)) (kBq g^{-1})
1	1	0.1112 (6)	591.8 (30)
	2	0.1009 (6)	610.0 (40)
2	1	0.1123 (7)	592.1 (41)
	2	0.0973 (7)	630.4 (46)
3	1	0.1135 (8)	598.6 (42)
	2	0.1038 (7)	604.1 (45)
4	1	0.1100 (9)	592.3 (48)
	2	0.0931 (8)	610.6 (53)
5	1	0.1138 (6)	591.9 (33)
	2	0.0957 (7)	618.4 (43)
6	1	0.1002 (13)	633.0 (80)
	2	0.0862 (12)	658.3 (91)
7	1	0.1104 (12)	601.5 (64)
	2	0.0927 (11)	632.3 (73)
8	1	0.1116 (8)	580.1 (41)
	2	0.0995 (10)	601.5 (63)

Uncertainties are expressed as standard uncertainties ($k = 1$).

obtained by using the following ratio:

$$\left(\frac{N_{0Fe}^2(1)}{N_{0Fe}(2)}\right)_i = N_{0Fe}(0) \qquad (8)$$

with $i = {}^{51}Cr, {}^{54}Mn$, where, using Eq. (5), $N_{0Fe}(1)_i$ corresponds to one Al absorber ^{55}Fe activity corrected by the $f_\varepsilon(1)_i$. $N_{0Fe}(2)_i$ corresponds to two Al absorber ^{55}Fe activity corrected by the $f_\varepsilon(2)_i$. $N_{0Fe}(0)_i$ corresponds to the ^{55}Fe activity extrapolated to zero absorber.

To check the results obtained with this concept, the theoretical ^{51}Cr, ^{54}Mn and ^{55}Fe efficiencies for each X-ray energy were calculated by means of the code MCNP. In the theoretical calculation the X-ray emission probabilities per decay and the intrinsic efficiency of the PC counter by the MCNP code were considered. The correction factor f_ε from these efficiencies was determined and applied to the experimental values to determine the ^{55}Fe activity.

3. Results and discussion

Tables 1 and 2 present the individual experimental efficiencies obtained with ^{51}Cr and ^{54}Mn tracers with one and two Al absorbers, respectively. Table 3 shows the average efficiencies of ^{51}Cr and ^{54}Mn with one and two Al absorbers obtained from the data shown in Table 1. It also shows the value of ^{55}Fe efficiency obtained using Eq. (7), and the correction factor f_ε as described by Eq. (6), for one and two Al absorbers for ^{51}Cr ($f_{\varepsilon Cr}$) and ^{54}Mn ($f_{\varepsilon Mn}$), respectively.

Table 4 presents the averages, from Tables 1 and 2, of the activities for one and two Al foils and the values of these activities after the application of the correction factor f_ε. The final value of $N_{0Fe}(0)$ for zero absorbers for each tracer is determined using Eq. (8).

The average activity of ^{55}Fe solution obtained by the Z-number relationship using the two tracers ^{51}Cr and ^{54}Mn gave a result of $(513 \pm 11)\,kBq\,g^{-1}$.

Table 5 shows the X-ray emission probabilities per decay, the intrinsic efficiencies (obtained by means of the code MCNP), the partial and total efficiencies, and the correction factor that was applied to the experimental data. This correction factor has been obtained by the ratio between the MCNP theoretical efficiencies calculated for ^{55}Fe and tracer, respectively, as shown in Table 5. The uncertainty in this correction factor was estimated by running the code in different 4π detector conditions.

It can be observed from the MCNP results that the absolute uncertainties in the calculated efficiencies are expected to be much larger due to the simplifications in the detector geometry adopted for the present calculation. The ^{55}Fe corrected activity corresponds to the ratio between the uncorrected ^{55}Fe activity for zero absorber (from Table 4) and this correction factor. The average ^{55}Fe final activity using this procedure was $(539 \pm 15)\,kBq\,g^{-1}$.

Table 6 gives the individual uncertainty components involved in the standardization of ^{55}Fe. The dominant uncertainty is related to the Z-number extrapolation procedure and related to the decay data used in the theoretical calculation.

Table 2
Efficiencies and apparent activity values of ^{55}Fe obtained with ^{54}Mn as a tracer with one and two Al absorbers

Source	Number of Al absorbers	N_c/N_γ	Uncorrected ^{55}Fe activity, $N_{0Fe}f_\varepsilon$ (Eq. (5)) (kBq g^{-1})
1	1	0.1308 (3)	541.0 (17)
	2	0.1141 (3)	555.7 (18)
2	1	0.1236 (3)	542.6 (18)
	2	0.1141 (3)	556.5 (18)
3	1	0.1190 (3)	552.7 (14)
	2	0.1126 (3)	567.8 (18)
4	1	0.1130 (3)	558.6 (17)
	2	0.1152 (3)	570.2 (17)
5	1	0.1133 (3)	553.1 (13)
	2	0.0995 (3)	575.0 (14)
6	1	0.1269 (3)	563.1 (11)
	2	0.1123 (3)	570.7 (13)
7	1	0.1247 (3)	550.1 (15)
	2	0.1114 (3)	564.9 (17)

Uncertainties are expressed as standard uncertainties ($k = 1$).

Table 3
Extrapolation of the average efficiencies with one and two Al absorbers as a function of atomic number Z

Tracer	Number of Al absorbers	Z	Average N_c/N_γ	^{55}Fe efficiency (Eq. (7))	f_ε (Eq. (6))
^{51}Cr	1	23	0.1104 (15)		1.203 (30)
^{54}Mn	1	24	0.1216 (26)		1.092 (20)
	1	25		0.1328 (53)	
^{51}Cr	2	23	0.0962 (20)		1.316 (24)
^{54}Mn	2	24	0.1113 (20)		1.136 (25)
	2	25		0.1265 (51)	

Uncertainties are expressed as standard uncertainties ($k = 1$).

Table 4
Average activity of ^{55}Fe obtained with the two tracers, for one and two Al absorbers and corrected for the factor f_ε

Tracer	Number of Al absorbers	Average uncorrected ^{55}Fe activity (kBq g^{-1})	f_ε	^{55}Fe N_0 (Eqs. (5) and (8)) (kBq g^{-1})	Average activity (kBq g^{-1})
	0	575.6 (97)		513 (11)	
^{51}Cr	1	597.7 (55)	1.203 (30)	497 (11)	
^{51}Cr	2	620.7 (67)	1.316 (24)	472 (8)	
					513 (11)
	0	537.8 (50)		512 (9)	
^{54}Mn	1	551.6 (36)	1.092 (20)	505 (9)	
^{54}Mn	2	565.8 (28)	1.136 (25)	498 (10)	

Uncertainties are expressed as standard uncertainties ($k = 1$).

Table 5
Efficiencies of ^{55}Fe for zero absorber calculated by the code MCNP, and corrected ^{55}Fe activities calculated from MCNP correction factors

Nuclide	X-ray energy	MCNP efficiency (A)	X-ray intensity (B)	MCNP efficiency/decay (A × B)	Correction factor (MCNP sum ^{55}Fe/ sum tracer)	Corrected ^{55}Fe activity (kBq g^{-1})
^{51}Cr	4.95	0.4447	0.202	0.0898		
	5.43	0.4117	0.024	0.0099		
			Sum	0.0997	1.054 (25)	546 (16)
^{54}Mn	5.40	0.4140	0.225	0.0932		
	5.98	0.3700	0.030	0.0111		
			Sum	0.1043	1.008 (25)	534 (14)
^{55}Fe	5.90	0.3769	0.250	0.0942		
	6.50	0.3297	0.033	0.0109		
			Sum	0.1051		
					Average	539 (15)

Table 6
Typical uncertainties components of the activity concentration

Components	Standard uncertainty ($k = 1$) (%)	
	^{51}Cr	^{54}Mn
Tracer		
Weighing	0.10	0.10
Dead time	<0.1	<0.1
Background	0.25	0.03
Tracer activity	0.19	0.12
Decay scheme/ Z-extrapolation (includes random counting uncertainties)	2.0	2.0
Half-life	<0.1	<0.1
Combined uncertainty	2.03	2.01

From the preliminary, unpublished results of the international comparison, the activity obtained using Z-number extrapolation agrees well with the average activity from other laboratories. However, the activity obtained by means of the theoretical efficiency calculated with the MCNP code is higher than this average value. This shows that the assumptions applied in the simulation have to be revised.

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