Activity measurements of ^{55}Fe by two different methods

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Abstract: A calibrated germanium detector and CIEMAT/NIST liquid scintillation method were used in the standardization of solution of ^{55}Fe coming from a key-comparison BIPM. Commercial HISAFE'3 cocktails were used in source preparation for activity measurements in CIEMAT/NIST method. Measurements were performed in Liquid Scintillation Counter. In the germanium counting method standard point sources were prepared for obtaining atomic number versus efficiency curve of the detector in order to obtain the efficiency of 5.9 keV KX-ray of ^{55}Fe by interpolation. The activity concentrations obtained were 508.2 ± 3.56 and 510.0 ± 16.20 kBq/g, respectively for CIEMAT/NIST and germanium methods.

Keywords: ^{55}Fe; activity standardization; Liquid Scintillation counting; CIEMAT/NIST; Gamma spectrometry.

1. INTRODUCTION

^{55}Fe disintegrates by electron capture with emission of electrons and X-ray of low energy followed by the emission of a gamma ray of 125.95 keV with very low emission probability of 1.3 x 10^{-7} % [1].

It is a very important radionuclide in the calibration of low energy photon spectrometers, proportional and scintillating detectors due to its long half-life of 1001.1(21) d and emission of 5.9 keV characteristic X-ray. This work describes the results of the standardization of a ^{55}Fe solution by using a planar type germanium detector and the CIEMAT/NIST liquid scintillation counting methods [2, 3].

The measured solution originates from an international key-comparison organized by Bureau International des Poids et Mesures (BIPM) in 2005.

For germanium counting standard point sources of ^{51}Cr, ^{54}Mn, ^{57}Co and ^{65}Zn with standard combined uncertainties lower than 1 % were used for plotting the atomic number versus efficiency curve of the detector in order to get the efficiency of 5.9 keV energy of ^{55}Fe by interpolation. Correction for the attenuation in source support, air and beryllium window of the detector were determined for the energies of X-rays of the radionuclides used to obtain the efficiency curve.

In the CIEMAT/NIST method with ^{3}H as a tracer, a Liquid Scintillation Counter model Wallac 1414 was used and five samples made by commercial OPTIPHASE 'HISAFE' 3 scintillation cocktails with ^{55}Fe and $FeCl_3$ inactive carrier were prepared for measurements. Several commercial cocktails were tested for checking the stability of the sources in a period of 12 days, evaluated by means of the counts normalized to the value of the first measurement and the spectra obtained over the period. The results showed that only the samples in 'HISAFE' 3 with and without carrier showed to be stable.

2. MEASUREMENT BY PLANAR GERMANIUM DETECTOR

The planar germanium detector used was a Canberra model GL2020R with 400 eV FWHM energy resolution at 5.9 keV with Beryllium window of 0.5 mm thick. Standard point sources of ^{51}Cr, ^{54}Mn, ^{57}Co and ^{65}Zn, previously standardized by $4\pi\beta$(PC)-γ(NaI) coincidence counting method were used in the calibration of efficiency versus nuclide atomic number for the activity determination of ^{55}Fe solution. Standard uncertainties (k = 1) of these sources were 0.29, 0.41, 0.29 and 0.40 %, respectively for ^{51}Cr, ^{54}Mn, ^{57}Co and ^{65}Zn. The detection efficiency versus atomic number at 5 cm distance between the detector window and the source was obtained with these sources.

The uncertainties of counting the peaks in the KX-ray of each standard source were of the order of 0.1 to 0.3 % for each measurement. The output pulses were used to a multi-channel analyzer and the peak areas were analyzed by commercial Maestro software [4].

Each measurement was corrected for the attenuation effects of source support (polystyrene), air and Beryllium window of the detector. The data for calculating the attenuation effects were taken from NIST X-ray Mass Attenuation coefficient [5]. The detection efficiency at 5.90 keV ^{55}Fe K X-ray was evaluated by interpolating the efficiency versus atomic number curve obtained from the standard sources. The activity of ^{55}Fe sources were obtained by dividing the X-ray count rate by the interpolated efficiency and emission intensity of 5.9 keV X-ray of ^{55}Fe.

The sources were prepared by dropping quantitative masses of radioactive solution onto the center of a polystyrene disk of 25.4 mm diameter and 0.05 mm thick. A drop of TWEEN 20 was diluted by 1000 times and added to each source and dried in a desiccator with silica gel. After drying, the sources were covered by the same polystyrene film and placed at 5 cm from the detector for measurements.

Figure 1 shows the efficiency versus atomic number curve obtained with ^{51}Cr, ^{54}Mn, ^{57}Co and ^{65}Zn and sources placed at 5 cm from de detector window. Table 1 gives the relevant data of mass attenuation corrections to correct count rates and obtain the efficiency versus atomic number curve.

The measured activity obtained was 509.95 ± 16.20 (3.2 %) kBq/g at reference date of 1 December 1st, 2005, 00:00 UTC. The uncertainty budget is presented in table 2.

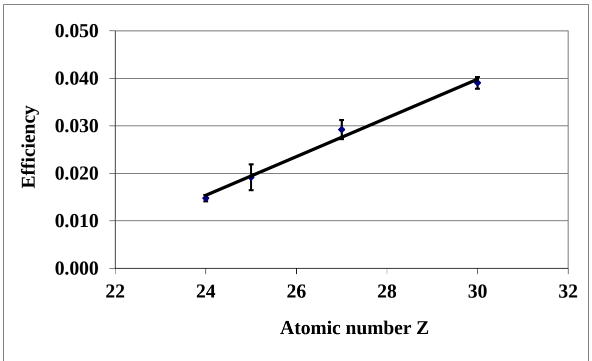


Figure 1 Efficiency versus atomic number curve.

Table 1 Mass attenuation correction for X-ray of ^{51}Cr, ^{54}Mn, ^{57}Co and ^{65}Zn used to obtain the detection efficiency of 5.9 keV X-ray of ^{55}Fe.

Nuclide (Z)	Mass attenuation correction		
	Polystyrene	Air	Beryllium
^{51}Cr (24)	0.9109	0.7852	0.6688
^{54}Mn (25)	0.9282	0.8244	0.7267
^{57}Co (27)	0.9550	0.8863	0.8183
^{24}Zn (30)	0.9044	0.9441	0.9044
^{55}Fe (26)	0.9466	0.8659	0.7879

Nuclide (Z)	Energy KX-ray (keV)	Efficiency	Standard uncertainty
^{51}Cr (24)	4.95	0.014773	0.000338
^{54}Mn (25)	5.40	0.019174	0.001366
^{57}Co (27)	6.40	0.029212	0.000999
^{65}Zn (30)	8.04	0.039044	0.000611
^{55}Fe (26)	5.90	0.023077	0.000641

Table 2. Uncertainty budget (% of activity concentration) evaluated in the determination of activity concentration of ^{55}Fe by germanium method.

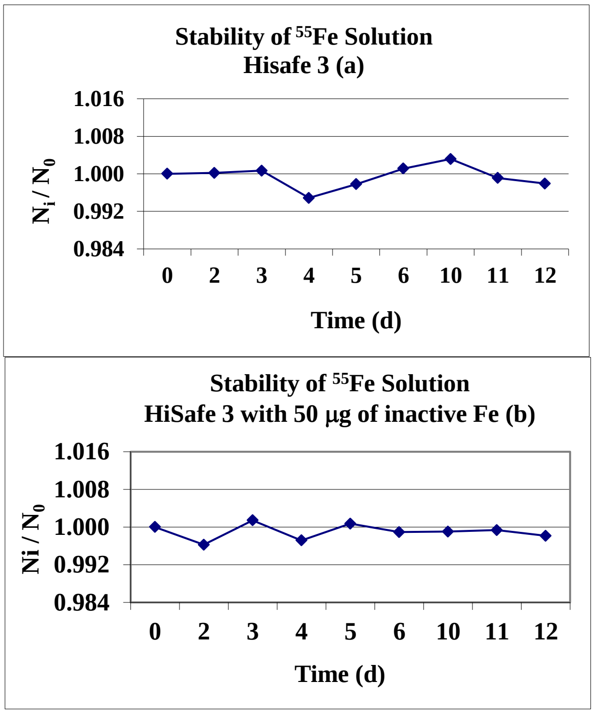
Component due to	Relative standard uncertainty
Counting statistics	0.30
Decay data	1.63
Detection efficiency	2.8
Weighing	0.05
Pile-up	0.01
Half-life	0.006
Combined standard uncertainty*	3.2

3. MEASUREMENT BY THE LIQUID SCINTILLATION COUNTER

The Liquid Scintillation Counter Wallac 1414 was used in the CIEMAT/NIST counting method with ^{3}H as tracer and efficiency calculation CN2003 code [6].

Samples made by commercial scintillation cocktails Ultima Gold, Instagel Plus and Optiphase 'Hisafe'3, manufactured by Perkin Elmer were measured by a period of 12 days to stability checking of the samples. The measurement counts were normalized to the first measurement and the results are presented in the Fig. 2.

The results showed that only the samples prepared in Optiphase 'Hisafe' 3 with and without carrier showed to be stable. The spectra obtained from this cocktail in the initial data and after 12 d are presented in the Fig. 2. Five samples were prepared with this cocktail and the activity concentration obtained was 508.17 ± 3.56 (0.70 %) kBq/g at reference date. The uncertainty budget is presented in Table 2.



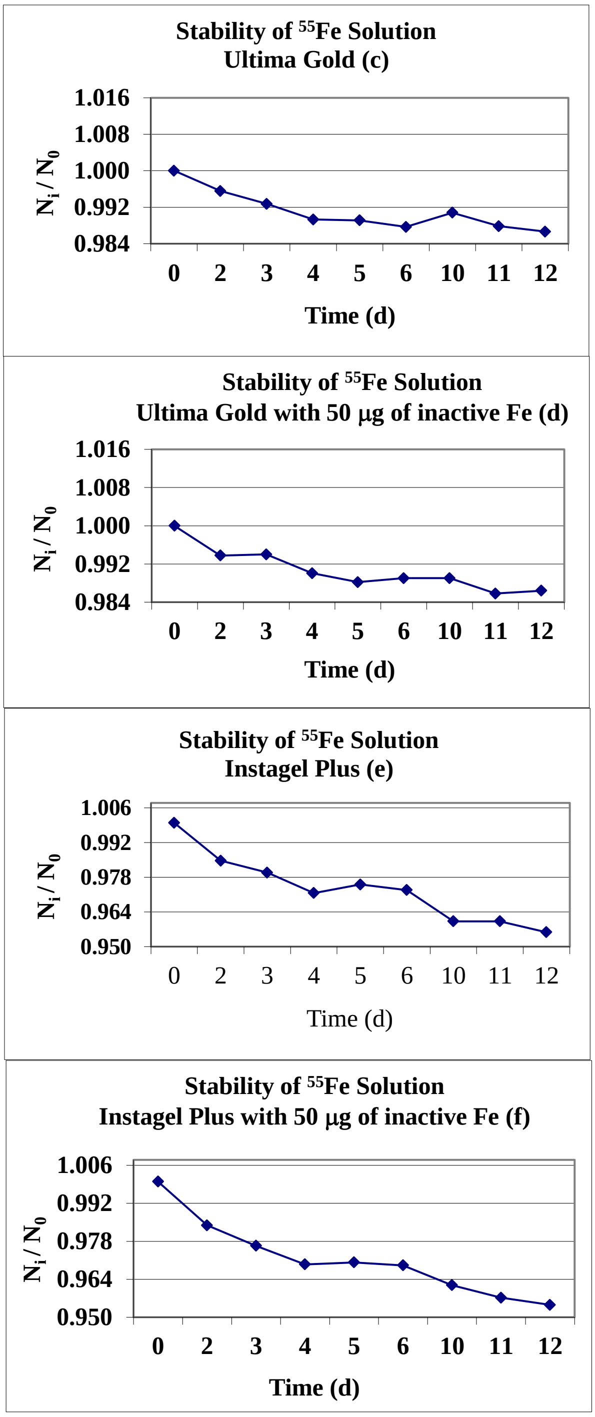


Figure 2 (a) to (f). Test of source stability checking for samples prepared by several cocktails.

Table 3. Uncertainty budget (% of activity concentration) evaluated in the determination of activity concentration of ^{55}Fe by germanium method.

Component due to	Relative standard uncertainty
Counting statistics	0.03
Decay data	0.30
Weighing	0.01
Half-life	0.006
Quench determination of ^{3}H	0.363
^{3}H activity standardization	0.307
^{3}H counting	0.07
PMT efficiency lost	0.33
Quenching ^{55}Fe	0.20
PMT asymmetry	0.10
Ionization quenching	0.10
Combined standard uncertainty*	0.70

4. DISCUSSION

The two major contributions to the total uncertainty in the germanium method are the determination of the efficiency of the 5.9 keV KX-ray of ^{55}Fe (2.8 %) by interpolation of the efficiency curve and the atomic decay parameters of the radionuclides found in literature used to obtain this curve (1.63 %). Standard sources with smaller uncertainties should be used to reduce these uncertainties. Much lower uncertainty (0.70 %) was found in the CIEMAT/NIST method, so the value of this method was adopted as a reference value for comparison with the results of the BIPM key-comparison. However, the activities of ^{55}Fe obtained by planar germanium and liquid scintillation methods agreed well within the evaluated uncertainties.

The values of activity concentration by germanium and liquid scintillation methods agree also reasonably well with the published reported value (without outliers) of the international key-comparison, which was 516.8 ± 2.9 kBq/g [7], as depicted in figure 3. The standardization of ^{55}Fe is not trivial due to its decay scheme (it decays mainly by electron capture with nearly 100 % emission probability to the ^{55}Mn ground state), emitting a very low energy characteristic X-ray (5.9 keV). This has been shown in the international comparison between the results reported by participants where the spread of the results relating to the arithmetic mean ranged between -1.7 % and 3.7 % [7].

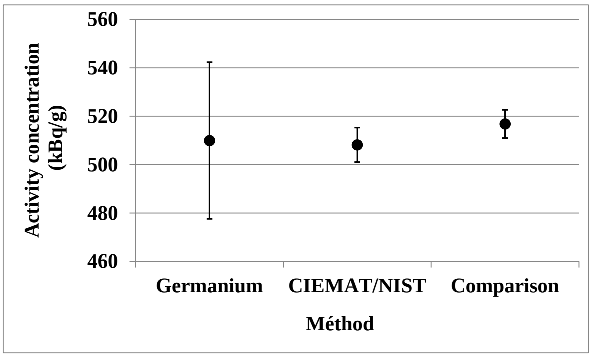


Figure 3 Activity concentration of ^{55}Fe solution. Uncertainty bars refer to expanded k = 2.

5. CONCLUSIONS

Despite large uncertainty the indirect measurement method using a planar germanium detector was satisfactory for activity determination of ^{55}Fe with results comparable to the liquid scintillation method. Of the two methods the most simple with less work is the liquid scintillation counting using CIEMAT/NIST method which gave uncertainty compatible with the needs of the end users as laboratories of metrology and medical application. Most cocktails tested proved to be unstable over time requiring more research in finding a suitable scintillation cocktail for ^{55}Fe radioactive solution. The results obtained in LNMRI presented good agreement with BIPM key-comparison value (k = 2).

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