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Standardization of ^{59}Fe by 4π(PC)β–γ software coincidence system

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HIGHLIGHTS

- ^{59}Fe standardised by 4π(PC)β–γ software coincidence system.
- Thin windowed proportional counter employed along with both HPGe and NaI(Tl) detectors.
- Good agreement between 4π(PC)-HPGe and 4π(PC)-NaI(Tl) results.

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ABSTRACT

The procedure for the standardization of ^{59}Fe using a 4π(PC)β–γ software coincidence system is described. The standardization was performed with an experimental setup consisting of a thin window gas-flow proportional counter (PC) in 4π geometry coupled to a NaI(Tl) scintillator and to a HPGe detector. The data acquisition was carried out by means of a Software Coincidence System (SCS). The beta efficiency was changed by using Collodion films and aluminum foils as external absorbers.

1. Introduction

The procedure for the standardization of ^{59}Fe using a 4π(PC)β–γ software coincidence system is presented. The radioactive solution was part of an ampoule sent to the National Laboratory of Radiation Metrology (LNMRI), in Rio de Janeiro, by the Technical Committee on Ionizing Radiation of the BIPM for an international comparison.

Fe-59 decays with a half-life of 44.495 days by beta minus emission to the excited levels of ^{59}Co, followed mainly by 1099 keV and 1291 keV gamma transitions (Fig. 1). There are two main beta ray branches with maximum energies of 273.6 (45.21%) and 465.9 keV (53.33%), respectively. The first one is coincident with the 1291 keV gamma ray but only marginally with the 1099 keV gamma ray and the second beta ray is coincident only with the 1099 keV gamma ray (Bé et al., 2014).

The standardization was performed in a 4πβ–γ coincidence system consisting of a thin window gas-flow proportional counter (PC) in 4π geometry coupled to a NaI(Tl) scintillator and to a 20% relative efficiency HPGe detector. The data acquisition was carried out by means of the Software Coincidence System (SCS) developed at the Nuclear Metrology Laboratory (LMN) at the IPEN-CNEN/SP (De Toledo et al., 2008).

The activity was obtained by means of the extrapolation curve setting two gamma windows at 1099 keV and 1291 keV total absorption peaks, respectively. These measurements were carried out using the HPGe detector. The beta efficiency was changed by using Collodion films and aluminum foils as external absorbers on both sides of the sources. Measurements using the NaI(Tl) crystal for gamma detection were also performed, setting the gamma window at the total absorption peak of 1291 keV.

2. Experimental method

2.1. Source preparation

The ^{59}Fe sources were prepared by dropping known aliquots of the solution on a 20 μg cm^{-2} thick Collodion film. This film had been previously coated with a 10 μg cm^{-2} gold layer on both sides in order to render the film conductive. A seeding agent (CYASTAT SM) was used to improve the deposit uniformity, and the sources were dried under a nitrogen jet at 45 °C (Wyllie et al., 1970). The mass determination was performed by the pycnometer technique using a Mettler XP56 balance. A total of 12 sources were prepared with masses ranging from 12 to 45 mg.

Some of these sources were measured in the HPGe gamma ray spectrometer for impurity checks and no impurities were detected.

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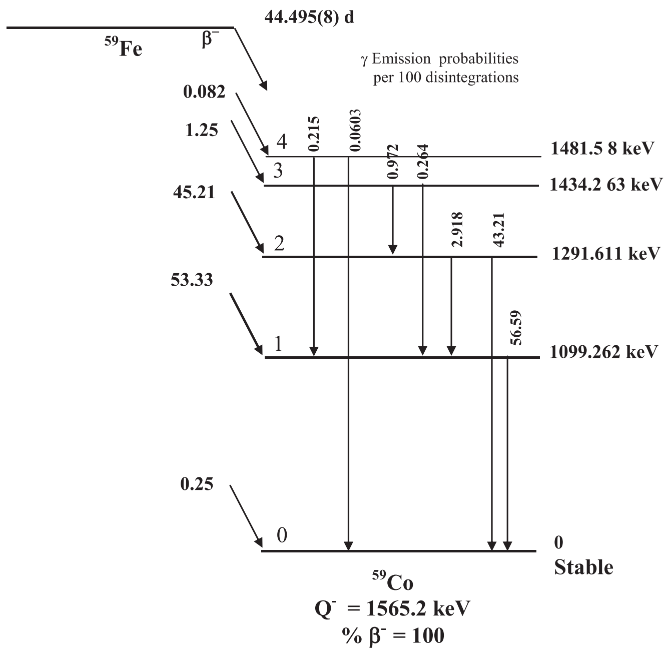

Fig. 1. Decay scheme of ^{59}Fe (Bé et al., 2014).

2.2. 4πβ–γ coincidence system

A 4π proportional counter filled with P-10 gas and operated at 0.1 MPa was used for beta detection. The gamma ray detectors were: a 50.1 mm × 50.1 mm NaI(Tl) with 76 keV resolution at 1291 keV, positioned above the PC wall, and a HPGe coaxial crystal, model GMX-20195 ORTEC, with 56.8 mm length and 49.0 mm diameter with 2.6 keV of the resolution at 1291 keV, positioned below the PC wall.

The experimental system at the LMN in São Paulo includes a Software Coincidence Counting System (SCS) with three input channels, allowing two sets of coincidence measurements simultaneously: 4π(PC)-HPGe and 4π(PC)-NaI(Tl).

The SCS is based on a National Instruments PCI-6132 card capable of up to four independent analog inputs and interfaced by means of NI Lab VIEW 2010 Platform acquisition program (De Toledo et al., 2008; National Instruments, 2014). The ADCs coupled features a 14-bit maximum resolution, at a 2.5 MS/s rate. The signals from the amplifiers (572 Ortec amplifier, with shaping time = 2 μs) are sent to the interface connected by a special cable to the PCI to be processed by means of a LabView Version 8.5 acquisition program. Information on pulse height and time of occurrence were registered for one beta and two gamma channels. The nuclear pulses were digitized and recorded on disk.

The beta, gamma and coincidence counting are determined from recorded data. Some system parameters (such as delays and time fluctuations) can be set in the analysis program, providing a software adjusting capability. Thus, any changes in the system can be performed by proper parameter setting, in the analysis step, by means of the software coincidence code SCTAC version 6.0, developed at the LMN (Dias, 2012), which allows selection of several gamma windows for the coincidence measurements. Corrections for dead time and accidental coincidences were applied during the analysis, after the experiment has been completed.

2.3. Variation of beta efficiency

The beta efficiency variation was performed by applying external absorbers over and under the radioactive sources, in order to determine the activity of the ^{59}Fe solution by means of the efficiency extrapolation technique.

Two gamma ray windows were selected: one at 1099 keV and the other at 1291 keV full absorption energy peaks. The gamma window at 1099 keV is coincident with all beta branches, mainly with the beta ray branch with maximum energy of 465.9 keV (53.33%); the other window at 1291 keV is coincident mainly with the beta ray branch with maximum energy of 273.6 keV (45.21%).

Using external absorbers, the contributions from lower energy electrons to the beta efficiency are progressively excluded. The beta efficiency for the 465.9 keV beta branch is higher than for the 273.6 keV branch due to the difference in beta energy. For this reason, when the 1099 keV gamma window is selected, the measured beta efficiency is higher when compared to the true efficiency in the beta channel; as a result, the slope of the extrapolation curve is negative. This result is true because the gamma efficiency in the beta channel is low. On the other hand, when the 1291 keV gamma window is selected, the measured beta efficiency is lower; therefore the extrapolation curve slope becomes positive.

3. Results and discussion

Typical ^{59}Fe gamma ray spectra for NaI(Tl) and HPGe are presented in Figs. 2 and 3, respectively. The extrapolation curves obtained with 4π(PC)-HPGe and 4π(PC)-NaI(Tl) systems using external absorbers, are presented in Fig. 4. The fit of a straight line was performed by least-squares by means of code LINFIT (Dias, 1999) which incorporates covariance matrix methodology. The absorbers used to change the beta efficiency were made of Collodion films, previously coated with 20 μg/cm^2 gold layer, and aluminum foils.

The efficiency parameter was varied in the range from 46% to 82% for measurements with NaI(Tl) at the 1291 keV full energy absorption peak. The gamma ray window was selected between 1204 keV and 1394 keV (see Fig. 2).

The beta efficiency for the measurements with the HPGe detector varied in the range from 46% to 84% for the gamma window of 1291 keV total absorption energy peak, and in the range from 67% to 90% for the gamma of 1099 keV total absorption energy peak. The difference in the maximum beta efficiency is due to the contribution from beta branches of different maximum energies to each gamma ray transition.

The observed counting rates N_β and N_γ were corrected for background, dead time and decay in the usual way for measurements using the 1291 keV in gate gamma. Due to the off line

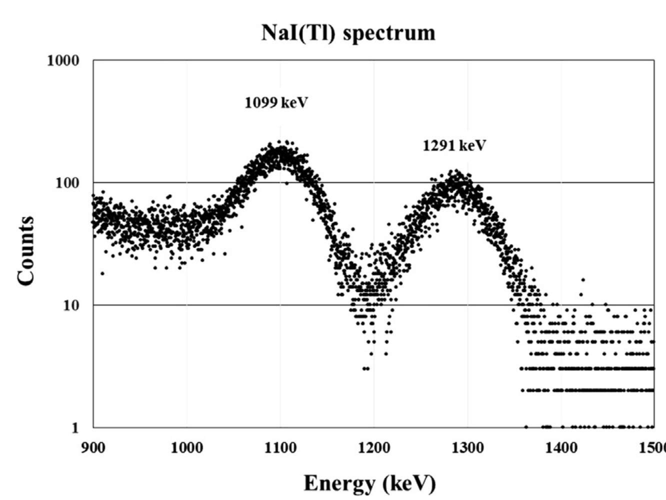

Fig. 2. A typical gamma ray spectrum obtained for ^{59}Fe using NaI(Tl) crystal from the 4πβ–γ system.

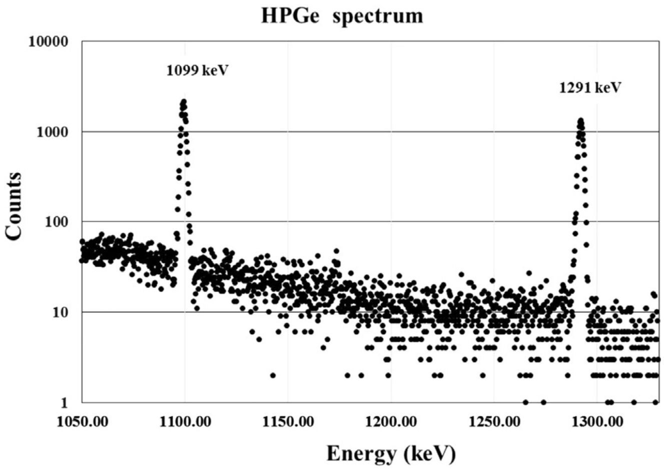

Fig. 3. A typical gamma ray spectrum obtained for ^{59}Fe using HPGe detector from the 4πβ–γ system.

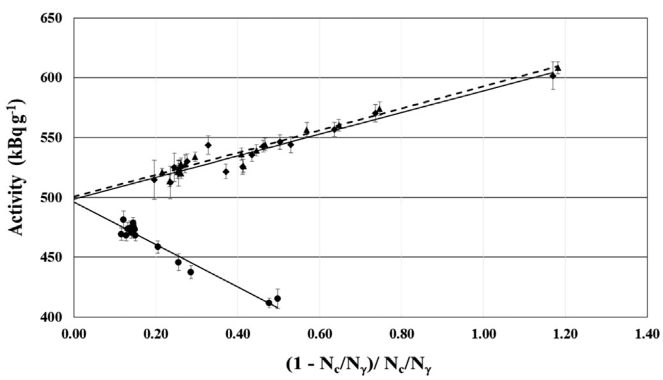

Fig. 4. Extrapolation curves for 1099 keV and 1291 keV gamma windows, obtained with the two gamma ray detectors, using external absorbers. The black dots are experimental points from 1099 keV, the black diamonds are from 1291 keV, both obtained with HPGe detector. The triangles are from 1291 keV obtained with NaI (Tl). The fitting curves are shown by the continuous line from HPGe and the dashed line is from NaI(Tl).

Table 1
Typical partial uncertainties in the activity using the external absorbers method, in percent (k=1).

Component	Uncertainty (%)		
	HPGe 1099 keV	HPGe 1291 keV	NaI(Tl) 1291 keV
Counting statistics in N_β, $N_c/N\gamma$ and gamma background were included in the fitting procedure	–	–	–
Weighing	0.10	0.10	0.10
Dead time/software	0.10	0.10	0.10
Decay correction	0.27	0.27	0.27
Resolving time /software	0.05	0.05	0.05
Extrapolation of efficiency curve/ least square fit error	0.50	1.04	0.41
Combined uncertainty	0.73	1.17	0.67

analysis, in the correction for gamma background, for the measurements at 1099 keV gamma ray, it was included the contribution of Compton events from the 1291 keV gamma ray transition. The coincidence rate was corrected for dead time and accidental coincidences using the Cox-Isham formalism (Cox and Isham, 1977) adapted by Smith (Smith, 1978).

The values obtained for the activity were 496.1(36) kBq g^{-1} for the 1099 keV gamma ray window using the HPGe detector, and 498.5(58) kBq g^{-1} and 500.5(34) kBq g^{-1} for the 1291 keV gamma ray window, using HPGe and NaI(Tl) detctors, respectively. The uncertainty budget associated with the experimental activity is shown in Table 1. The main uncertainties involved were: fitting procedure; counting statistics, from N_β and $N_c/N\gamma$ counting and gamma background, which were considered uncorrelated, and the uncertainties in weighing, dead time, decay correction and resolving time, considered correlated.

4. Conclusion

The standardization of a ^{59}Fe solution was successful applying the SCS installed at the LMN by using external absorbers. A good agreement was observed comparing two different gamma windows, obtained by using HPGe and NaI(Tl) detectors.

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