DOI: 10.1007/s10967-008-1204-7 *Journal of Radioanalytical and Nuclear Chemistry, Vol. 278, No.3 (2008) 621–625*

Calibration of gamma-ray spectrometers coupled to Compton suppression and fast pneumatic systems for the k_0-standardized NAA method

H. M. Dung,[1]* M. C. Freitas,[1] S. Sarmento,[1] M. Blaauw,[2] D. Beasley[1]

[1] *Institute for Technology and Nuclear (ITN), E.N. 10, Ap. 21, 2686-953 Sacavém, Portugal*
[2] *Reactor Institute Delft, Mekelweg 15, 2629 JB Delft, The Netherlands*

(Received July 10, 2008)

A new Compton suppression system (CSS) for the gamma-ray spectrometer portion of the neutron activation analysis (NAA) was set up at the RPI/ITN. The pneumatic transfer system, SIPRA, for short-lived nuclides and cyclic irradiations was improved. A full calibration procedure of the CSS and SIPRA systems was performed. Two certified reference materials, NIST-SRM-1572 (Citrus Leaves) and NIST-SRM-1633a (Coal Fly Ash) were analyzed using the calibration factors. The CSS was instrumental in lowering the detection limits of Cr, Fe, Hg, Rb, Sr, Th and Zn by reducing background and/or spectral interference considerably. The analytical results were evaluated by comparison to the NIST certified values with deviations ranging from 2% to 8% for the above mentioned elements, except Zn ranging from 10% to 15% for biological and environmental samples, respectively.

Introduction

The instrumental neutron activation analysis (INAA) has two main stages that affect sensitivity and detection limits: the irradiation in a neutron field and the measurement on a gamma-ray spectrometer. A fast pneumatic transfer system is commonly used for the activation of samples that have short-lived nuclides and for transferring those samples to a gamma-ray spectrometer for measurement of their activities. A pneumatic system with the capability of cyclic irradiation and accumulative gamma-ray spectrum measurement is required for the INAA. The implementation of INAA on the pneumatic system by the k_0-standardization method (k_0-INAA) enhances the overall effectiveness of the system because of the inherent advantages of this method.[1,2]

An active Compton suppression system (CSS) was installed at the RPI/ITN in order to reduce the background of the gamma-ray spectrum for use with the INAA. In order to apply the k_0-INAA method, the gamma-ray spectrometers coupled to the pneumatic and the CSS systems require full calibration of energy, resolution and full-energy peak efficiency.[2] The calculation of the absolute efficiency of the gamma-ray spectrometer when coupled to the CSS in the anti-Compton mode is not an easy task because of the cascade coincidences in the level decay schemes of many of the radionuclides. The peak areas and, therefore, the absolute efficiencies are suppressed by the CSS with a certain probability that depends on the decay time of each level (on the order of ps or ns). The various coincidence types of β-γ, e-γ, X-γ, 511γ-γ, γ-γ angular correlations and the CSS itself all contribute to the complex coincidence summing effects. The CSS can simultaneously measure in two modes: with and without the anti-Compton enabled (two separate gamma-ray spectra acquired). In the spectrum with the anti-Compton enabled, the radionuclides without cascade coincidences in the level decay schemes (e.g., Cr, Hg, Zn) are identified and analyzed while the spectrum without anti-Compton enabled is processed for the radionuclides with cascade coincidences in the level decay schemes (e.g., Co, Eu, Se, etc.).

The purpose of this paper is to describe the calibration of the gamma-ray spectrometers connected to the pneumatic system and the CSS without the anti-Compton mode. Two certified reference materials, NIST-SRM-1572 (Citrus Leaves) and NIST-SRM-1633a (Coal Fly Ash) were analyzed using k_0-IAEA[3,4] software in order to check the applicability of the calibrated parameters.

Experimental

Description of CSS and SIPRA systems

The CSS for NAA in this work consists of a primary Canberra GX-3518 HPGe detector with a 35% relative efficiency and 1.8 keV resolution for the 1332.5 keV photopeak of ^{60}Co and a total of seven NaI(Tl) detectors. Six NaI(Tl) detectors are mounted on the surrounding annulus while another NaI(Tl) detector is used as a plug. All detectors are coupled to model Canberra 2026 spectroscopy amplifiers, Canberra Model 2040 coincidence analyzer and Canberra MultiPort-II (USB) operated by Genie-2000 software. The sample measurement positions are controlled by support rings, each ring with a height of 11 mm. The sample is mounted on a sample support of about 6–8 mm height depending on the type of sample vial. The sample support and rings are all put in a plastic cup fitting with

* E-mail: m.hodzung@itn.pt

the sample position space between plug and annulus NaI(Tl) detectors. The diagram of the CSS is shown in Fig. 1.

The pneumatic transfer system, SIPRA that is designed for NAA of short-lived nuclides and cyclic irradiations, was recently improved. The SIPRA is specifically designed to rapidly transfer samples to and from an irradiation terminal for use in the NAA application. The measurement devices of the SIPRA system consist of a Canberra GC-2018 detector and an ORTEC DSPEC Jr 2.0 operated by GammaVision software for acquisition and spectral analysis while the irradiation is controlled by locally developed software. The "Head" above the detector can be moved up or down (farther away or closer to the detector, respectively) and is set by manual controls before performing the irradiation and counting.

Investigation of the gamma-ray spectrometers

A test of the CSS was made using a ^{137}Cs source in the suppressed and unsuppressed modes – meaning with and without the NaI(Tl) guard detectors engaged. Two measurement positions were chosen on the detector. One position was directly on top of the end cap and the second position was at 10 cm from the detector end cap. Several mathematical comparisons were made between the two spectra. These include peak-to-compton plateau ratio, peak-to-Compton edge ratio and peak-to-total ratio. The Compton plateau is set to a range of 358–382 keV, the Compton edge is a range of 475–581 keV, the 661.7 keV peak is calculated from 654 to 669 keV and the total is from 0 to 8191 channel.

The results of the comparison of suppressed to unsuppressed ^{137}Cs spectra show that at the measurement position on the top of the detector the continuum of the spectra is reduced for the peak-to-Compton plateau by a factor of 6.29, the peak-to-Compton edge by a factor of 3.27 and the peak-to-total by a factor of 1.91. At the measurement position of 10 cm from the detector end cap the continuum of the spectra is reduced for the peak-to-Compton plateau by a factor of 3.43, the peak-to-Compton edge by a factor of 1.92 and the peak-to-total by a factor of 1.87. Figure 2 shows the ^{137}Cs spectra on detector top measured with (lower) and without (upper) Compton suppression.

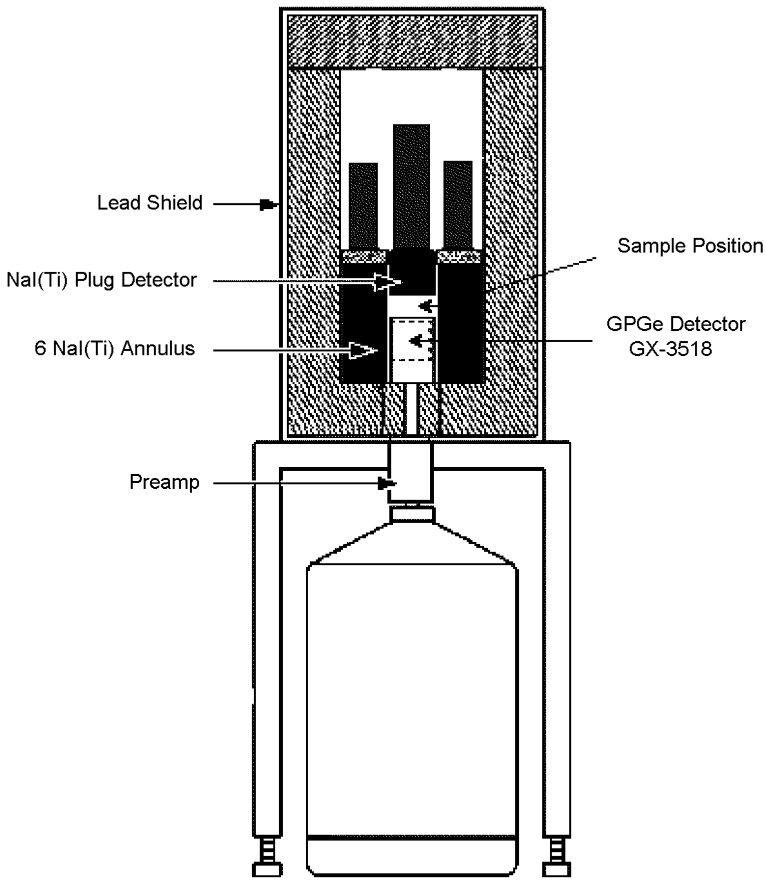

Fig. 1. The diagram of RPI/ITN Compton suppression system (CSS)

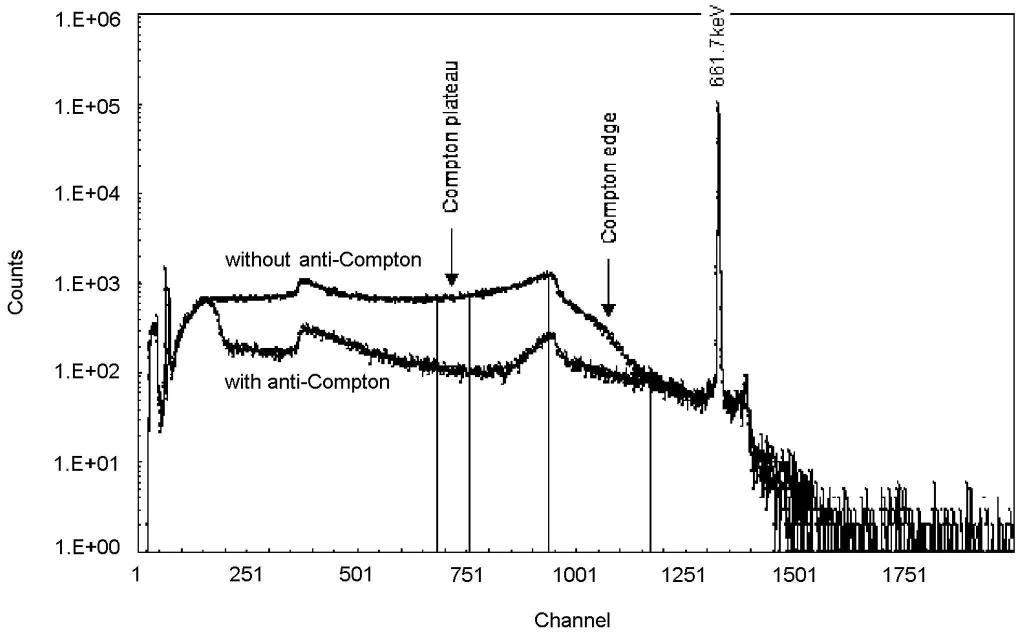

Fig. 2. The Cs-137 spectra on detector top measured with (lower) and without (upper) Compton suppression

The counting positions on the SIPRA system are changed by moving the "Head" down or up on the detector with distances limited to 36 mm below and 300 mm above. The measurements of specially activated samples containing several radionuclides (^{141}Ce, ^{166}Ho, ^{85}Sr and ^{170}Tm) covering the appropriate gamma-ray energy regions were performed at positions inside and outside of the "Head." The ratios of the net peak areas at the different gamma-ray energies inside and outside of the "Head" were fitted to a second order curve in order to derive the appropriate values at the position inside the "Head", since the standard sources could only be measured outside of the "Head." The fitted curve of the outside to inside ratio of the "Head" counting data obtained in this work for different gamma-ray energies at 150 mm from detector end cap is shown as follow:

$$y = 1.26 - 1\cdot 10^{-5} x - 1\cdot 10^{-8} x^2$$

with y is "Head" outside to inside ratio and x is energy.

Calibration of gamma-ray spectrometers

A series of measurements were carried out on CSS (without anti-Compton mode) and SIPRA systems in the following order: E-calib, background and point sources of ^{137}Cs and ^{152}Eu at reference positions of 105 mm and 150 mm from detector end cap for CSS and SIPRA, respectively, in which E-calib denotes the mixing of two sources ^{60}Co and ^{137}Cs for purposes of the energy and peak shape calibration.

The calibration was performed by the k_0-IAEA software with the following steps: (1) creation of a sample series containing the above mentioned spectra, and input of the source's data; (2) calibration for energy and peak shape using the E-calib spectrum and their storage; (3) interpretation of the background spectrum and its storage; (4) interpretation of the ^{137}Cs spectrum to determine the curves of peak-to-total, single and double escapes,[5,6] storage of these results; (5) interpretation of the ^{152}Eu spectrum to compute full energy peak efficiencies, storage of the fitted efficiency curves. The obtained efficiency curves were stored in the "permanent database" as full energy peak efficiencies at reference positions. When processing real samples, the reference efficiencies are recalled from the "permanent database" and the actual efficiencies for the sample are calculated with the appropriate corrections.

Figure 3 shows the fitted reference efficiency curves (a) CSS (without anti-Compton) and (b) SIPRA (inside "Head") measured at 105 mm and 150 mm from the detector end cap, respectively.

Analysis of certified reference materials

Two certified reference materials Citrus Leaves NIST-SRM-1572 and Coal Fly Ash NIST-SRM-1633a were prepared with about 150 mg of sample and placed inside high purity polyethylene vials. Each sample was submitted to a short irradiation time of 30 seconds through SIPRA (Cell 26 in RPI's core), while about 200 mg of sample was submitted to a long irradiation time of 5 hours in Cell 55. In the case of short-lived nuclides, the irradiation and measurement procedures were performed with a decay time of 15 seconds and counting time of 300 seconds. The measurement of long-lived nuclides was performed on the CSS both with and without anti-Compton modes, with a decay time of about 3 weeks and a counting time of 10 hours. A comparison of the analytical results between the two CSS modes has been reported by several other authors,[7,8] therefore, in this study the emphasis is placed on the exploration of the applicability of the k_0-NAA method using CSS (k_0-CSNAA).

Results and discussion

The investigated CSS and SIPRA systems present the suitable characteristics for the application of the k_0-NAA method.

The ratios of experimental to certified values, R(E/C), for some elements as determined by the k_0-CSNAA method of the two SRMs are displayed in Table 1.

In the case of NIST-SRM-1572, the deviation between the experimental and certified values is about 2% for Fe, 5% for Cr and Rb, 7% for Sr, 8% for Hg and 15% for Zn. For NIST-SRM-1633a, the deviation between the experimental and certified values is about 3% for Cr and Rb, 4% for Sr, 5% for Fe, 8% for Th and 10% for Zn.

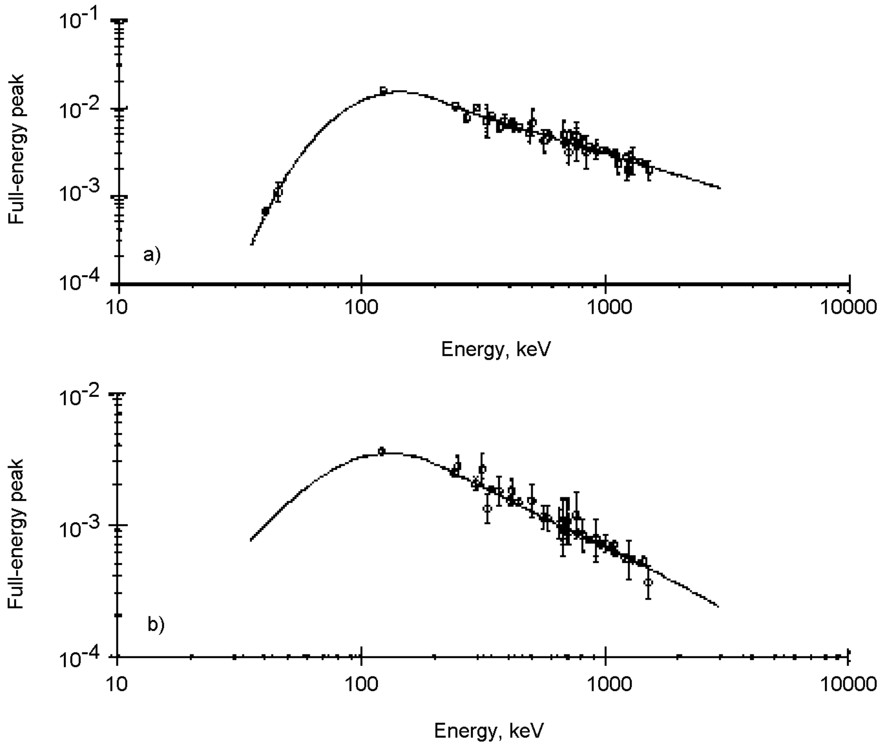

Fig. 3. The fitted reference efficiency curves (a) at 105 mm from detector end cap for CSS (without anti-Compton) and (b) at 150 mm from detector end cap for SIPRA (inside "Head")

Table 1. The analytical result of some elements in NIST-SRM-1572 and NIST-SRM-1633a determined by the k_0-CSNAA method

Element	NIST–SRM–1572				NIST–SRM–1633a			
	Average	STD	Certified	*R*(E/C)	Average	STD	Certified	*R*(E/C)
Cr	0.76	0.19	0.80	0.95	189	15	196	0.97
Fe	88	17	90	0.98	89000	4000	94000	0.95
Hg	0.09	0.01	0.08	1.08	–	–	–	–
Rb	5.1	1.6	4.84	1.05	127	19	131	0.97
Sr	107	20	100	1.07	795	7	830	0.96
Th	–	–	–	–	22.8	0.9	24.7	0.92
Zn	33.3	5.6	29.0	1.15	242	15	220	1.10

Average: Average experimental results.
R(E/C): Ratio of experimental to certified values.

The 320 keV energy of ^{51}Cr is free of cascade coincidence in its decay scheme. This energy peak is nearly obscured by the Compton background. By using the CSS the Compton background is reduced significantly in the measured gamma-ray spectrum thereby making this element suitable for INAA.

Similarly to ^{51}Cr described above, the 279 keV energy of ^{203}Hg is also free of cascade coincidence in its decay scheme and is also obscured by the Compton background. Moreover, in the normal INAA mode, the overlap of the 279.2 keV of ^{203}Hg with the 279.6 keV of ^{75}Se creates a spectral interference that affects the Hg detection limit. However, this interference is overcome considerably by using CSS in INAA because the 279.6 keV peak of ^{75}Se is suppressed primarily due to the cascade coincidence with other energy levels in the decay schemes. The interference of ^{75}Se to ^{203}Hg is according to Se/Hg ratio in the sample, i.e., the Se/Hg ratio in NIST-1572 and NIST-1633a are of 0.31 and 64, respectively. So, the interference for the first case was found to be insignificant, but the latter one presented an obvious interference for which the detection limit of Hg is improved considerably when the CSS is applied.

The 1115.5 keV of ^{65}Zn is free of cascade coincidence in its decay scheme. Although the 1115.5 keV is not obscured by the Compton background, its energy line has interferences from the 1112.1 keV of ^{152}Eu and the 1120.5 keV of ^{46}Sc. These spectral interferences are mostly suppressed in INAA using CSS because the 1112.1 keV of ^{152}Eu and the 1120.5 keV of ^{46}Sc are cascade coincidence in their decay schemes (121.8 keV and 889.3 keV, respectively).

Conclusions

The absolute efficiency calibration of gamma-ray spectrometers coupled to CSS and SIPRA systems enables the user to carry out the k_0-standardized NAA (k_0-NAA) for samples measured on these systems. The application of the k_0-NAA on the CSS (k_0-CSNAA) has sucessfully been performed in this work. Some radionuclides are free of cascade coincidence and can be interpreted using gamma-ray spectra acquired with Compton suppression enabled, while the other radionuclides that are cascade coincidence are to be interpreted as in traditional INAA. The CSS improves the detection limits of Cr, Fe, Hg, Rb, Sr, Th and Zn by reducing background and/or spectral interference considerably. In order to avoid the irradiation of a lot of reference materials, the k_0-NAA used on SIPRA is a significant solution at the ITN. A cadmium tube can be introduced before irradiation in the irradiation channel connected with the SIPRA in order to apply epithermal neutrons (ENAA). Therefore, the combination of the three options: ENAA, fast pneumatic transfer and CSS permits the user to enhance the capability of NAA by not only lowering the detection limits but also by extending the scope of the elements. The combinated INAA techniques at RPI/ITN will be presented in a near future.

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The research carried out in this report has been supported by the Portuguese Foundation for Science and Technology (FCT) through the REEQ/1075/FIS/2005 Project, the Post-doc SFRH/BPD/26345/2006 Programme for one of authors (H.M.D.) and the Gulbenkian Foundation. Thanks are due to M. EBIHARA (Tokyo Metropolitan University, Japan) and J. LASSATER (Ortec, USA) for their help in many ways.

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