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Elemental PGNAA analysis using gamma–gamma coincidence counting with the library least-squares approach

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Abstract

An accurate method for determining elemental analysis using gamma–gamma coincidence counting is presented. To demonstrate the feasibility of this method for PGNAA, a system of three radioisotopes (Na-24, Co-60 and Cs-134) that emit coincident gamma rays was used. Two HPGe detectors were connected to a system that allowed both singles and coincidences to be collected simultaneously. A known mixture of the three radioisotopes was used and data was deliberately collected at relatively high counting rates to determine the effect of pulse pile-up distortion. The results obtained, with the library least-squares analysis, of both the normal and coincidence counting are presented and compared to the known amounts. The coincidence results are shown to give much better accuracy. It appears that in addition to the expected advantage of reduced background, the coincidence approach is considerably more resistant to pulse pile-up distortion.

Keywords: PGNAA; Gamma–gamma coincidence

1. Introduction

One of the main advantages of gamma–gamma coincidence counting is minimization of background. For PGNAA, the sources of background include the gamma-rays from the naturally occurring radioisotopes (K-40, uranium and thorium), background generated by the neutron source including the source emitted gamma-rays and that generated by the source from the activation of surrounding materials and the detector itself. Hydrogen also (from sample and/or non-sample materials) causes problems in many typical cases of PGNAA. When activated, hydrogen de-excites by emitting a single prompt gamma-ray with an energy of 2.223 MeV. The magnitude of this peak from a normal sample spectrum is often one or two orders of magnitude larger than other elements of interest. This adds a source of complexity to the spectrum and causes sensitivity problems, particularly at energies less than or equal to 2.223 MeV. Other sources of complexity are summing and pulse pile-up effects.

Feasibility studies for PGNAA [3] were previously conducted and demonstrated convincingly the minimization of all these sources of background. In particular, the single commonly very intense hydrogen gamma-ray is eliminated, which opens up the low energy part of the prompt

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gamma-ray spectrum in bulk analysis applications. To illustrate these results further, Fig. 1 shows the single detector (HPGe) and coincidence spectra for cobalt-60. The spectra were taken in the NCSU PULSTAR reactor bay while the reactor was in operation. Because the reactor was operating at the time of the experiment, there was a high background level in the singles spectrum. Observing the coincidence spectrum, we notice that the natural background (as indicated by the 1.46 and 2.61 MeV gamma-rays) and the hydrogen spectrum are negligible. We also notice the elimination of the 2.5055 MeV cobalt sum peak.

The library least-squares (LLS) approach is very useful for inverse spectral problems such as determining radioisotope amounts from gamma-ray spectra and determining elemental amounts from X-ray or prompt gamma-ray spectra. While it requires somewhat more work than peak intensity determinations [1,4], it has the advantage [2] that it uses all of the spectral data that is available and, therefore, yields better accuracy.

An accurate method for determining elemental analysis is presented. The method combines the precision of the LLS approach and the advantages of gamma–gamma coincidence counting.

2. Instrumentation

Fig. 2 shows a block diagram of the coincidence system used. The first part of the system is based on Nuclear Instrument Module (NIM) instrumentation standards. Two pulses are extracted from the detector's preamplifier. The first pulse is passed to the amplifier that generates the linear pulse to be processed by the analog to digital converter (ADC).

The second pulse is used to generate the coincidence gate signal (discussed in the following), which plays a key role in coincidence measurements. The pulse is passed to a timing filter amplifier that generates a pulse having faster rise time and smaller width than that from linear amplifiers. The Constant Fraction Discriminator (CFD) processes the fast pulse and produces a logic signal when a constant fraction of the fast pulse peak amplitude is reached.

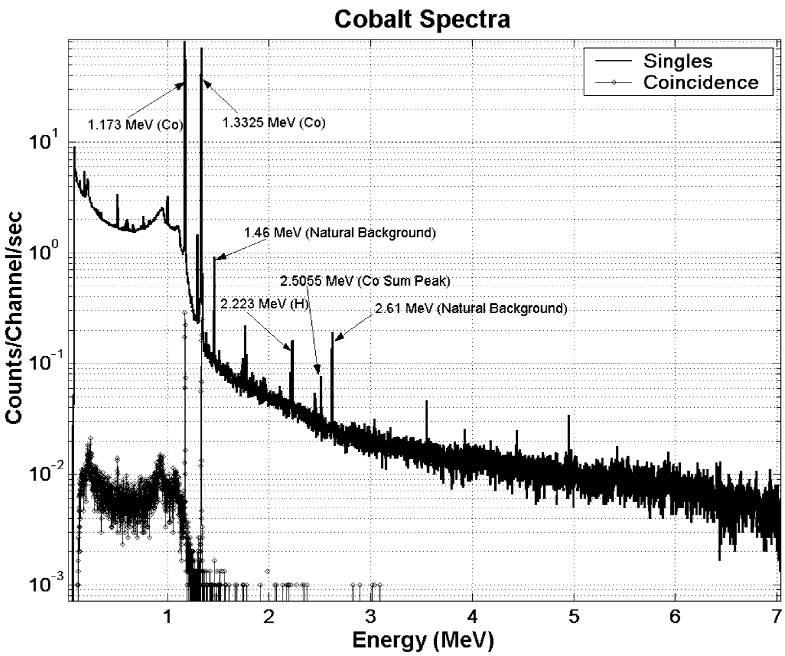

Fig. 1. Background reduction with coincidence counting.

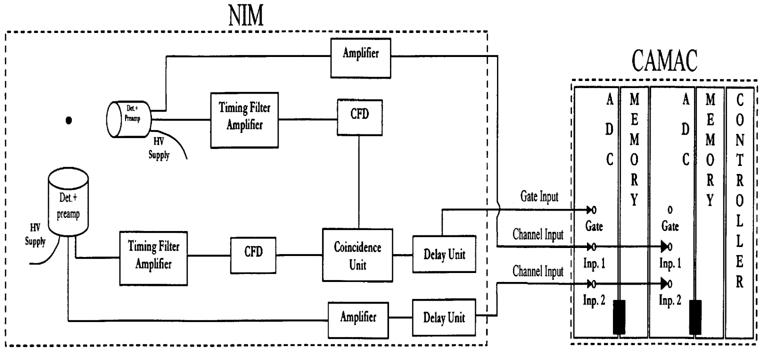

Fig. 2. Electronic setup for coincidence counting.

The coincidence unit processes the logic output of both CFD's. If they arrive within the preset resolving time of the unit, a logic signal is produced. In case of coincident events, this logic signal is produced much faster than the linear pulses by the amplifiers. A delay unit is therefore needed to make the logic signal coincide with the linear pulses. In addition, another delay unit may be connected to one of the amplifiers if it is set to a smaller shaping time than the other amplifier.

The second part of the system is based on Computer Automated Measurement and Control (CAMAC) instrumentation standards, which allowed us to collect both singles and coincidence data simultaneously. The linear pulse outputs from the NIM crate amplifiers are duplicated and passed to the ADC's in the CAMAC crate. Each ADC processes one set of linear pulses.

The logic signal from the coincidence unit in the NIM crate serves as the gate signal for one of the ADC's. Thus this ADC acquires coincidence data. The other ADC (without a gate signal) acquires the singles data.

Finally, the computer software used to manage the CAMAC modules and analyze the acquired data was KMAX NT.

3. Experimental approach and data

Two HPGe detectors with efficiencies of 59% and 72% were used for this experiment. The detectors were placed at 90° with respect to each other, with lead shielding in between to reduce the cross talk between them. An additional tungsten shield was placed around the 59% detector.

Three radioisotopes (Na-24, Co-60 and Cs-134) were made, in liquid solution form, using the NCSU PULSTAR reactor. Using the two HPGe detectors, the singles (for each detector) and coincidence spectra of each radioisotope were collected and later used as libraries. Then, a known mixture of the three radioisotopes was made and its singles and coincidence spectra were collected. The experiment was carried out at relatively high counting rates so that pulse pile up distortion of the spectra was a significant problem. The samples were placed 20 cm from each detector and the collection duration for each spectrum was 2 h. The resolving time of the coincidence unit was set at 80 ns.

Figs. 3 and 4 show the single detector and coincidence spectra collected for the 59% detector, respectively.

4. Elemental library least-squares analysis

Using the libraries obtained for Co-60, Cs-134 and Na-24, the LLS analysis was performed to determine the amount of activity of each radioisotope in the mixture. A FORTRAN program, CURLLS, was used for this purpose.

The single detector and coincidence mixture spectra and their corresponding LLS fitting obtained by the program are shown in Figs. 5 and 6, respectively. A summary of the activity results is given in Table 1.

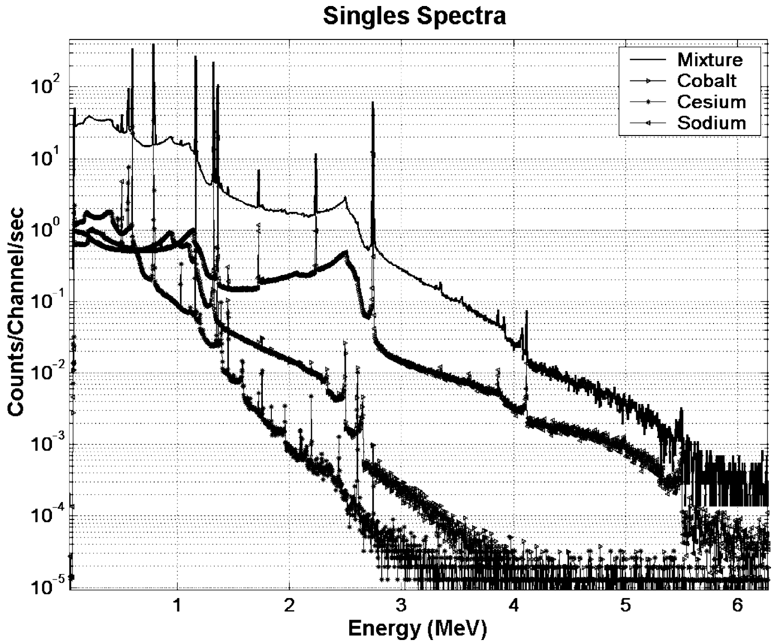

Fig. 3. Single detector spectra of the mixture and libraries.

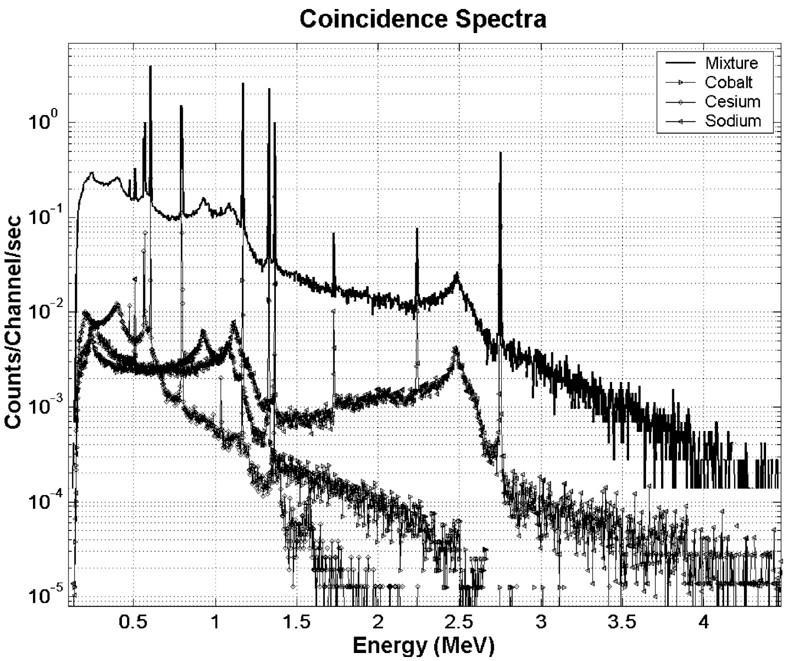

Fig. 4. Coincidence spectra of the mixture and libraries.

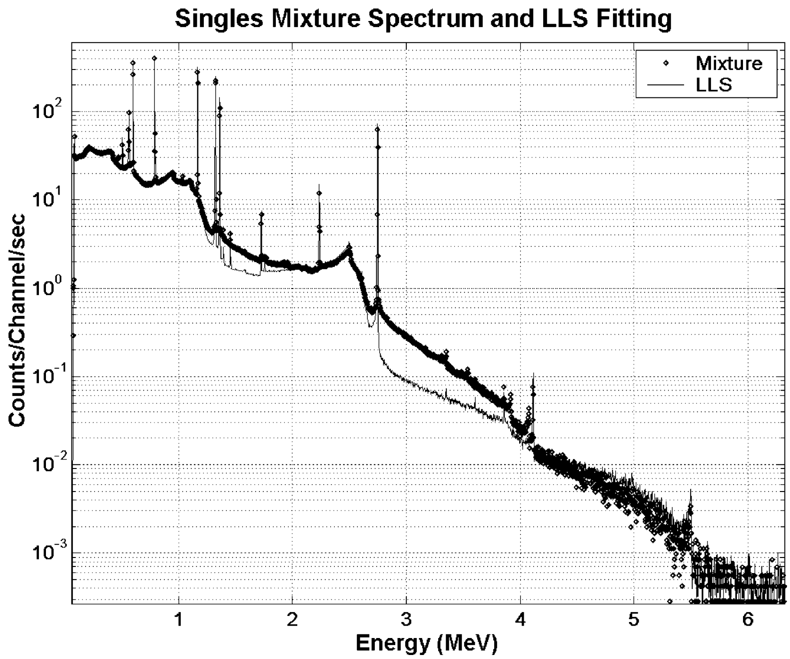

Fig. 5. Singles mixture spectrum and LLS fitting.

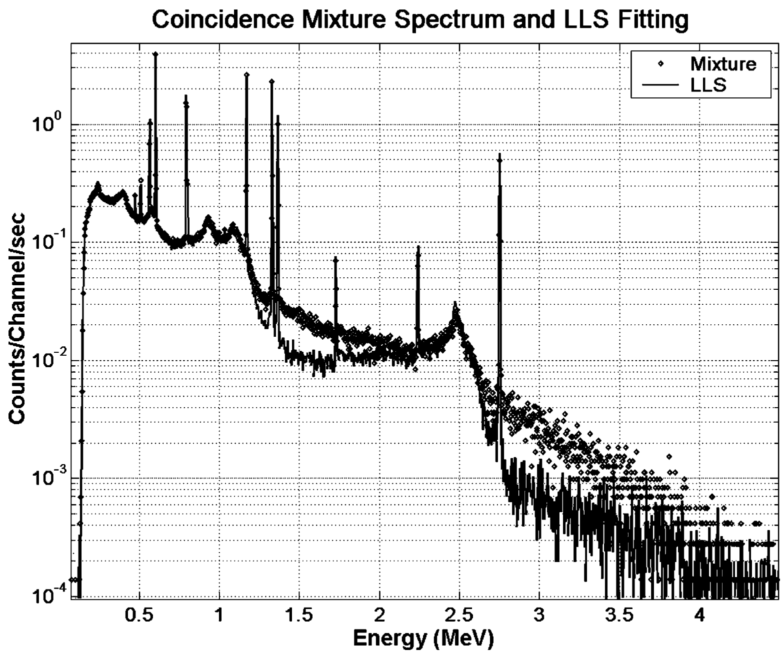

Fig. 6. Coincidence mixture spectrum and LLS fitting.

Table 1
Results of LLS analysis

	Activities (μCi)		
	Singles	Coincidence	Actual
Co-60	18.3498 ± 0.021	21.6415 ± 0.075	22.035
Cs-134	11.2869 ± 0.019	15.0725 ± 0.049	15.644
Na-24	5.86855 ± 0.0038	7.1469 ± 0.033	7.157
Reduced χ^2	43.2627	3.62929	

5. Discussion and conclusions

Table 1 shows that the results obtained by applying the LLS method to the coincidence spectrum are much closer to the actual values than those from the singles. This is attributed to background elimination in the coincidence spectrum as well as to the reduction of pulse pile-up effects. As a consequence, the reduced χ^2 value from the coincidence fitting is much closer to unity than that from the singles.

It appears that in addition to the expected advantage of coincidence counting of reducing the background, one also has the additional advantage that the coincidence approach is considerably more resistant to pulse pile-up effects.

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References

[1] P.P. Ember, T. Belgya, G.L. Molnar, Appl. Radiat. Isot. 56 (2001) 535.
[2] R.P. Gardner, A. Sood, Y.Y. Wang, L. Liu, P. Guo, R.J. Gerhke, Appl. Radiat. Isot. 48 (1997) 1331.
[3] R.P. Gardner, C.W. Mayo, E.S. El-Sayyed, W.A. Metwally, Y. Zheng, M. Poezart, Appl. Radiat. Isot. 53 (2000) 515.
[4] M. Vobecky, J. Jakubek, C. Granja Bustamante, J. Komicek, J. Pluhar, L. Pospisil, L. Rubacek, Anal. Chim. Acta 386 (1999) 181.