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▣ 論 文 ▣

Application of Pulse Pile-Up Correction Spectrum to the Library Least-Squares Method

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펄스 중첩 보정 스펙트럼의 라이브러리 최소자승법에의 이용

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Abstract - The Monte Carlo simulation code CEARPPU has been developed and updated to provide pulse pile-up correction spectra for high counting rate cases. For neutron activation analysis, CEARPPU correction spectra were used in library least-squares method to give better isotopic activity results than the convention library least-squares fitting with uncorrected spectra.
Key words : Pulse Pile-Up, Library Least-Squares Method, Monte Carlo Method

요약 - 개선된 몬테칼로 코드 CEARPPU는 고계수율 상황에서의 펄스 중첩 보정 스펙트럼을 제공한다. 중성자 방사화 분석을 위하여 CEARPPU를 이용하여 보정된 스펙트럼으로 라이브러리 최소자승 동위원소 방사능 분석을 수행하여 보정하지 않은 스펙트럼을 이용하는 방법보다 우수한 결과를 얻었다.
중심어 : 펄스 중첩, 라이브러리 최소자승법, 몬테 칼로법

Introduction

There are a number of reasons for using the highest possible radiation detection counting rates in radiation measurement applications. In many applications[1-3] one desires maximum precision in minimum measurement time, which inevitably introduces high counting rates and pulse pile-up distortions. When a radiation measurement is taken at a high counting rate, pulses entering the multi-channel analyzer in the system are easy to overlap with the neighboring pulses and the resulting piled-up pulses introduce distortions in the measured spectrum. In Figure 1, overlapped pulses are presented to illustrate pulse pile-up effects. Presently, one of the factors controlling the highest possible radiation detection counting rate is the pulse pile-up characteristics of the detection system of interest. While it is possible to reduce pulse pile up with hardware based pulse pile-up rejections, one can never completely eliminate it by this approach. In addition, the use of hardware based pulse pile-up rejection may introduce another distortion to the measured spectrum. Therefore, it is desirable to use software based pulse pile-up models which can, in principle, correct for any amount of pulse pile up. The present paper describes a pulse pile-up modeling approach based on Monte Carlo simulation. In the Monte Carlo code CEARPPU[4] two types of

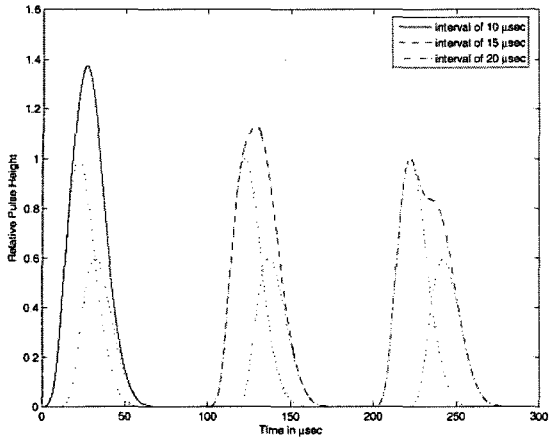

Fig. 1. Pulse Distortions from Pile-Up.

calculations became available; 'forward calculation' which gives a pulse piled-up spectrum from a true spectrum and a true counting rate and 'inversion calculation' which gives a true or corrected spectrum from a measured spectrum and a measured counting rate.

Previous work[5-7] on pulse pile-up modeling consisted of an exact deterministic model for double pulse pile up. Every possible double pulse pile-up event was considered which resulted in n^2calculations for a true spectrum of n channels. In principle, this approach could be extended to triple and more piled-up pulses, but that would involve n^m calculations where m is the number of pulses in the piled-up pulses. An extensive study of pulse pile up - including the use of Monte Carlo simulation is presented in Ref. 8. However, their Monte Carlo simulation was primarily for benchmarking other results and may not provide a practical simulation tool

Materials and Methods

1. Forward Simulation Modeling

The measurement system which is to be simulated usually consists of a semiconductor detector, a pre-amplifier, an amplifier and an MCA. Liquid nitrogen cooled Si(Li) and Ge detectors are common choices in X-Ray Fluorescence (XRF) and Prompt Gamma Neutron Activation Analysis (PGNAA) applications because of their excellent energy resolution and reasonable detection efficiency. Usually a pre-amplifier is attached to the detector and the amplifier produces semi-Gaussian shaped pulses to be processed and digitized by an analog-to-digital converter (ADC) which is a major functioning part of an MCA. Only the first local peak in a sequence of pulses which are piled up is detected by the MCA as the resulting pulse height of the pulse train. The rest of the pulse train is ignored until the baseline voltage is restored. It is assumed that all the components in the measurement system work perfectly, independent of experimental counting rate changes.

The Monte Carlo simulation approach is a 'forward' one that is based on assuming that a true counting rate and spectrum are available. Let N_t be the true total number of counts in a given counting time t_m and $h(E)$ be the true probability distribution function (pdf) of pulse-height energy E. To simulate the pulse piled-up total counts in time t_m and pulse-height distribution one simply samples N_t pulse events by first choosing a leading pulse size from the $h(E)$, then choosing an interval between this pulse and the next from the interval distribution, then choosing the first piled-up pulse from the $h(E)$ pdf, etc., until the interval between pulses is larger than the pulse width of a sample pulse. When this occurs, the pulse train being generated is terminated. The first local peak in each pulse train is numerically determined and scored in the pulse piled-up spectrum.

The interval distribution required is given by:

$$f(\Delta t) = \lambda \exp(-\lambda \Delta t) \quad (1)$$

where Δt is time interval and λ is the true counting rate N_t/t_m. Time intervals are chosen randomly by using the cumulative distribution function (cdf) obtained by integrating the pdf from 0 to Δt. This cdf is:

$$F(\Delta t) = \int_0^{\Delta t} f(x)dx = 1 - \exp(-\lambda \Delta t) \tag{2}$$

It is set equal to the cdf of the uniform random number to obtain t from the uniform random number R.

$$F(\Delta t) = R \tag{3}$$

$$\Delta t = -(1/\lambda)\ln(1-R) \tag{4}$$

As the input true spectrum is essentially an unnormalized pdf of pulse-height energy in tabular form, the pdf $h(E)$ and cdf $H(E)$ can be easily constructed from it. A random pulse-height energy is chosen by setting this cdf equal to the cdf of the uniform random number R and solving for E.

$$H(E) = \int_{x=0}^{E} h(x)dx = R \tag{5}$$

$$E = H^{-1}(R) \tag{6}$$

Where H^{-1} is the inversion function of H.

In simulation, the actual shape of the amplifier pulses should be used for the best result. Figure 1 shows the pulse shape used in the present case which comes from the ORTEC amplifier model 572[9] and the pulse trains formed by double pulse pile up. Usually only the observed counting rate, m, is known. In this case the true counting rate associated with the measurement can be obtained by numerical inversion from the paralyzable model[10] for a given pulse width which is assumed to be constant and independent of the pulse height.

$$m = \lambda \exp(-\lambda \tau) \tag{7}$$

A forward simulation result is presented in Figure 2 with measured spectra which were taken with a Si(Li) detector and an ^{55}Fe source. Amplifier shaping time of 2 μsec was used. Figure 2 shows excellent agreement with the measurements and it is seen that simulation results are accurate and correct at least through the triple pulse pile-up region. The larger difference in the region above 18 keV is thought to come mainly from increased background contributions. Also it is noticed that when the same number of pulses are simulated as are present in the experiment the same degree of random fluctuations as in the measurement can be obtained from simulation.

From Monte Carlo simulation one can extract important information by modifying the bookkeeping routines. Figure 3 shows various

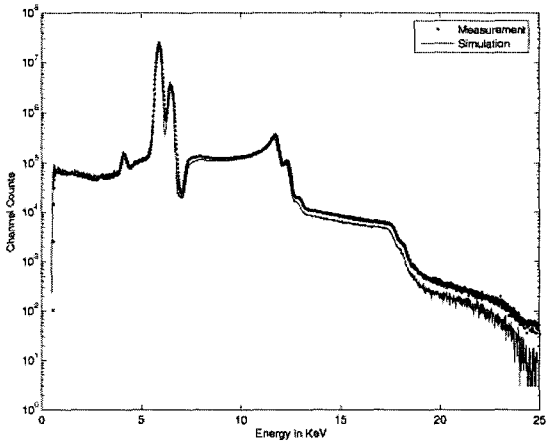

Fig. 2. CEARPPU Forward Calculation Result. (Source: ^{55}Fe, Detector: Si(Li), Counting Rate: 18167cps, Shaping Time: 2 μsec)

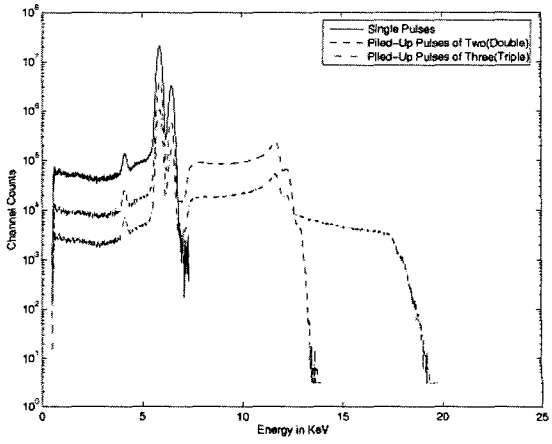

Fig. 3. Components in a Piled-Up Spectrum.

components of the pulse piled-up response for the case in Figure 2. Spectral information is gathered and grouped according to the number of pulses in a pulse train to give the components. Spectral response from single pulse trains has exactly the same shape as the input true spectrum. All the higher components also have lower pulse pile-up contributions in them. The shapes of these components look reasonable and might be used in inversion calculations, which is finding the true spectrum from a pulse pile-up distorted spectrum

2. Inversion Calculation

With the spectral results of the forward calculation, which is calculating distorted spectrum from true spectrum, one can notice that any measured spectrum is made up with spectral components (h_1, h_2 and so on; h_1 means the spectral response of single pulse only cases) as shown in Figure 3.

$$h_M(E) = h_1(E) + h_2(E) + h_3(E) + h_4(E) + \ldots \quad (8)$$

Since h_1 should have the same shape as the true spectrum, which is h_T , one can get simply get the true spectrum by subtracting higher order spectral terms from the measured (or distorted) spectrum.

$$h_T(E) = ch_1(E) = c(h_M(E) - h_2(E) - h_3(E) - h_4(E) - \ldots) \quad (9)$$

In equation (9) c is simply a constant.

In CEARPPU the following iterative strategy is adopted to obtain the true spectrum out of a measured spectrum.

[step1] Adopt the measured spectrum as a true spectrum.
(Or if a spectrum closer to the true is available, use the spectrum.)

[step2] Use the adopted true spectrum to calculate higher order spectral terms.

[step3] Obtain better spectrum (closer to the true) by subtracting higher order spectral terms[step2] from the measured spectrum.

[step4] Check convergence.

[step5] Return to [step1] with new adopted true spectrum.

This inversion approach have been applied for high counting rate measurements for Si(Li) detectors in the previous research, and in this paper it is applied for Ge detector responses for Neutron Activation Analysis.

Results

In Library Least-Squares method (LLS) unknown sample spectrum is expressed with linear combination of element spectra, which are called elemental libraries. The elemental fraction or elemental activity is proportion to the coefficients of linear combination, and these coefficients are obtained from nominal least square fitting using the libraries as basis functions.

For Neutron Activation Analysis (NAA) three element library measurements were taken for ^{24}Na, ^{60}Co and ^{134}Cs at North Carolina State Univ. NAA facility and these measured elemental spectra along with their CEARPPU-corrected ones are presented in Figures 4 through 6. Figure 7 contains the measured spectrum for unknown sample (a mixture of ^{24}Na, ^{60}Co and ^{134}Cs) and its CEARPPU-corrected spectrum. Linear combination coefficients of the elemental libraries were obtained through LLS fitting to the spectrum of unknown sample. In this case the coefficients represent elemental activities. The fitting result using CEARPPU- corrected spectra is shown in Figure 8 and the two spectra agreed closely with each other over wide range of energy. In Figure 9 the result with uncorrected spectra is presented. The resulting isotopic activities from

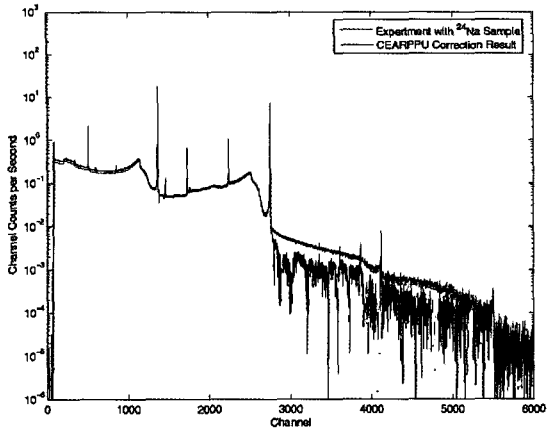

Fig. 4. Measured and Corrected Spectra for a ^{24}Na Sample at NCSU NAA Facility.

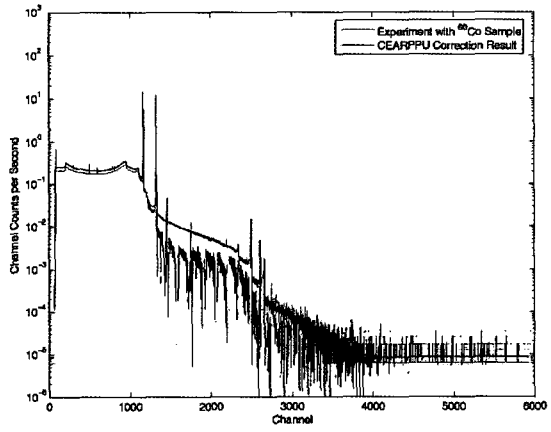

Fig. 5. Measured and Corrected Spectra for a ^{60}Co Sample at NCSU NAA Facility.

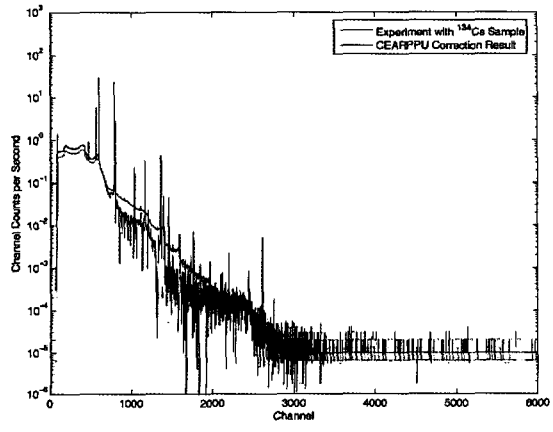

Fig. 6. Measured and Corrected Spectra for a ^{134}Cs Sample at NCSU NAA Facility.

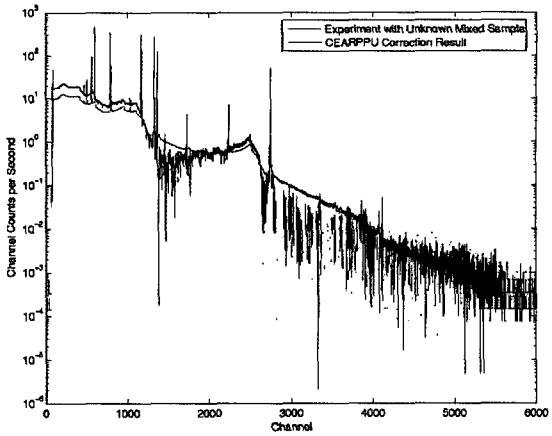

Fig. 7. Measured and Corrected Spectra for Unknown Mixed Sample at NCSU NAA Facility.

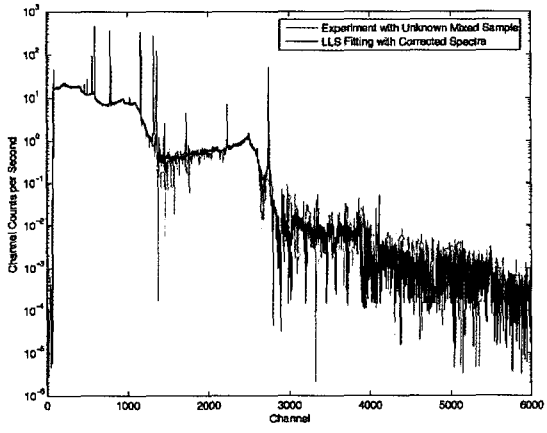

Fig. 8. Library Least-Squares Fitting Result with Corrected Spectra.

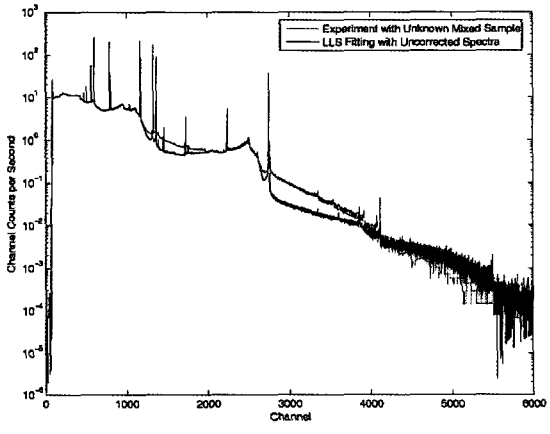

Fig. 9. Library Least-Squares Fitting Result with Uncorrected Spectra.

Table 1. Library Least-Squares Fitting Results with Corrected and Uncorrected Spectra.

Isotopes	LLS Fitting Result with Uncorrected Spectra [μCi]	LLS Fitting Result with Corrected Spectra [μCi]	Actual(True) Isotopic Activity [μCi]
^{60}Co	18.27	22.31	22.04
^{134}Cs	11.37	15.19	15.64
^{24}Na	5.81	6.83	7.16

two methods are listed in Table 1.

As presented in Table 1 and Figure 9 the conventional fitting (with uncorrected spectra) result using the distorted measurement spectra deviates maximum 19% (^{24}Na activity) from the actual value and the disagreement of the two spectra shown in the Figure 9 is considered to be the main reason. The spectral disagreement came from different distortions in each spectrum and hiders taking full advantage of all spectral information. Each spectrum has different regions of distortion and as a result the fitting does not work for the whole range of the spectrum and one can notice distinct difference between the fitting result and the measured spectrum.

With the help of CEARPPU, pulse pile-up corrections were performed onto the four measured spectra and these corrected spectra were used in getting isotope activity information through Library Least-Squares Fitting. As presented in Table 1 and Figure 8 the maximum error for the new fitting result with corrected spectra is 5% (^{24}Na activity) and the reason for the lowered error is shown in the Figure. The measured spectra and the resulting linear sum of libraries (model-fit) match very closely through out the whole energy range, which essentially means LLS can take advantage of all the data in the spectrum.

Discussion and Conclusions

Based on the underlying physics of the radiation spectroscopy measurements, a generalized Monte Carlo forward approach has been proposed and recently updated with inversion calculation routine. The inversion approach proposed by this article iteratively employs this Monte Carlo forward approach to correct the distorted spectrum to the true spectrum. From the applications to Neutron Activation Analysis spectra it is shown that Library Least-Squares fitting result can improve greatly by taking full use of all data in the spectrum.

With the help of this pulse pile-up model and a detector response function, a spectrum calculated by a radiation transport/simulation code like MCNP or CEARXRF can be converted to a simulated pulse-height spectrum which has a convolved Gaussian spread and pulse pile up. It can then be compared directly with an actual measured spectrum to give a reduced chi-square value that can be used directly in a suitable iterative analysis method. Or with the use of the inversion calculation, one may get the true spectrum from a measured spectrum without using pulse pile-up rejectors.

Use of this code with a suitable Monte Carlo radiation transport code and a detector response function allows the simulation of actual observed pulse-height spectra for analyzer systems taken at high counting rates with a large amount of spectral distortion. This will enable one to perform elemental analyses by an iterative technique using the actual uncorrected experimental spectra directly. Use of such high counting rate data will minimize *in vivo* measurement errors due to body movement and allow the highest possible accuracy for on-line process measurements.

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