Status of the Monte Carlo library least-squares (MCLLS) approach for non-linear radiation analyzer problems

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ABSTRACT

The Center for Engineering Applications of Radioisotopes (CEAR) has been working for over a decade on the Monte Carlo library least-squares (MCLLS) approach for treating non-linear radiation analyzer problems including: (1) prompt gamma-ray neutron activation analysis (PGNAA) for bulk analysis, (2) energy-dispersive X-ray fluorescence (EDXRF) analyzers, and (3) carbon/oxygen tool analysis in oil well logging. This approach essentially consists of using Monte Carlo simulation to generate the libraries of all the elements to be analyzed plus any other required background libraries. These libraries are then used in the linear library least-squares (LLS) approach with unknown sample spectra to analyze for all elements in the sample. Iterations of this are used until the LLS values agree with the composition used to generate the libraries. The current status of the methods (and topics) necessary to implement the MCLLS approach is reported. This includes: (1) the Monte Carlo codes such as CEARXRF, CEARCPG, and CEARCO for forward generation of the necessary elemental library spectra for the LLS calculation for X-ray fluorescence, neutron capture prompt gamma-ray analyzers, and carbon/oxygen tools; (2) the correction of spectral pulse pile-up (PPU) distortion by Monte Carlo simulation with the code CEARIPPU; (3) generation of detector response functions (DRF) for detectors with linear and non-linear responses for Monte Carlo simulation of pulse-height spectra; and (4) the use of the differential operator (DO) technique to make the necessary iterations for non-linear responses practical. In addition to commonly analyzed single spectra, coincidence spectra or even two-dimensional (2-D) coincidence spectra can also be used in the MCLLS approach and may provide more accurate results.

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1. Introduction and background

The library least-squares (LLS) approach (Marshall and Zumberge, 1989) is a common mathematical approach to perform elemental analysis. The basic assumption of this approach is that any unknown sample spectrum can be taken as the sum of the products of the elemental amounts and the library spectrum of each element or component for every channel in a multi-channel analyzer spectrum. This sum is given by

$$y_i = \sum_{j=1}^{m} x_j a_{ij} + e_i, \quad i = 1, 2, 3, \ldots, n$$

where y_i is the total counts in channel i for the unknown sample spectrum, x_j the amount of component j in the unknown sample, m the number of elements or components in the sample, n the number of channels, a_{ij} are the counts in channel i for component j (the library spectrum of component j), and e_i the random error in channel i due to statistical fluctuations. A set of simultaneous linear equations can be generated from the above equation. The x_j, elemental amounts, can be obtained from the set of linear equations by minimizing the corresponding reduced Chi-square, χ_v^2, which is given by

$$\chi_v^2 = \sum_{i=1}^{n} \frac{e_i^2}{(n-m)\sigma_i^2}$$

where $(n-m)$ is the number of degrees of freedom and σ_i^2 is the square of the standard deviation or variance in each channel.

The LLS approach has the advantages of being able to use the entire spectrum (peaks and continuum) that is available and being capable of analyzing spectra with unresolved peaks. However, in applying the LLS approach, the elemental library spectra, a_{ij}, must be available. Two approaches are capable of providing elemental library spectra. One is by means of experimentally measuring the library spectrum for each element while the other is by simulation, which is the one used by the Monte Carlo library least-squares (MCLLS) approach. Since these library spectra in the non-linear case will vary from one sample to the other due to matrix interaction effects, sample density, sample moisture, etc.; each sample composition usually requires a separate experiment to obtain each library spectra for every element. Therefore, the

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experimental approach is very time-consuming and costly for the non-linear analyzer case.

The Center for Engineering Applications of Radioisotopes (CEAR) at North Carolina State University has been working for about 10 years on the Monte Carlo library least-squares approach for treating non-linear radiation analyzer problems including prompt gamma-ray neutron activation analysis (PGNAA) for bulk materials, energy-dispersive X-ray fluorescence (EDXRF) analysis, and carbon/oxygen (C/O) ratio analysis in oil well logging. This approach essentially consists of using Monte Carlo simulation to generate the libraries of all the elements to be analyzed plus any other required libraries. These libraries are then used in the linear LLS approach with unknown sample spectra to analyze for all elements in the sample. The basic procedure of the MCLLS approach is as follows: (1) assume a sample composition as close to the actual one as possible; (2) calculate by Monte Carlo simulation, the elemental library spectra for a sample with the assumed composition; (3) calculate by the linear LLS method, the elemental amounts from the experimental sample spectrum; and (4) compare the assumed to calculated elemental amounts and iterate steps 1 and 2 with a new assumed composition if amounts are so far apart that the linear assumption cannot be made. The detailed treatment of the MCLLS approach and its evaluation are given in articles by Shyu (1991), Shyu et al. (1993), Zhang and Gardner (2005), Han et al. (2005), Han and Gardner (2007), and Han et al. (2007).

The current status of the methods (and topics) necessary to implement the MCLLS approach are reported here. They include: (1) the Monte Carlo codes such as CEARXRF, CEARCO, and CEARCPG for generating the necessary elemental library spectra for the LLS calculation for X-ray fluorescence, carbon/oxygen tools, and neutron capture prompt gamma-ray analyzers; (2) the correction of spectral pulse pile-up (PPU) distortion by Monte Carlo simulation with the code CEARIPPU; (3) generation of detector response functions (DRF) for detectors with linear and non-linear responses for Monte Carlo simulation of pulse-height spectra; and (4) the use of the differential operator (DO) technique to make the necessary iterations for non-linear responses practical. In addition to commonly analyzed single spectra, coincidence spectra or even two-dimensional (2-D) coincidence spectra can also be used in LLS and may provide more accurate results under optimized conditions.

2. Monte Carlo codes CEARCPG, CEARCO, and CEARXRF

To implement MCLLS for the non-linear radiation analyzer problems, accurate forward Monte Carlo models are required to calculate elemental library spectra when the geometrical and sample composition variables are known. CEAR has been working on developing these forward models since about 1978.

CEARPGA I was the first specific purpose Monte Carlo simulation code publicly reported to implement the MCLLS algorithm for PGNAA analyzers. This code was originally developed to generate the elemental library spectra needed to implement the MCLLS method. The code incorporated a variety of variance reduction techniques to improve calculation efficiency such as forcing all prompt gamma rays to be emitted after a neutron interaction (stratified sampling), using the expected value splitting technique to increase the score probability of each tracked gamma ray, using the correlated sampling method to deal with small variations of sample compositions, and using the detector response function to convert the incident gamma-ray spectra on the detector to pulse-height spectra. Based on CEARPGA I, Zhang (2003) developed the next generation code called CEARPGA II. The major improvement in that code was that

the big weight problem of code CEARCPG I was solved and library spectra of natural background and the neutron activation libraries of the NaI detector were added. The biggest limitation of the CEARPGA I and II codes are that they were originally designed to simulate the single library elemental spectra. The algorithm used to generate neutron-induced gamma rays limits its capability for simulating coincidence measurements. CEARCPG was recently developed for this purpose (Han et al., 2005). It is the first specific purpose Monte Carlo code for elemental analysis for using the coincidence technique. It can generate both the single library spectra and the coincidence spectra for all elements of interest at the same time.

The first Monte Carlo code to generate elemental library spectra for EDXRF analyzers was called NCSXRF (He et al., 1991), which was based on previous work by Gardner and Hawthorne (1975), Doster and Gardner (1982a, b) and Yacout et al. (1987). The CEARXRF code, an extension of the NCSXRF code, has been developed and continuously improved at CEAR (Ao et al., 1995; Guo et al., 2004a, b, c) through the implementation of more precise low-energy photon transport and XRF physics. The motivation for developing the CEARXRF code is to make it applicable to *in vivo* XRF system design. To use Monte Carlo simulation for the X-ray coincidence measurement, the CEARXRC code was developed from the CEARXRF code.

For carbon/oxygen tool analysis in oil well logging applications, the specific purpose Monte Carlo code CEARCO (Gardner et al., 2006) based on a modification of the CEARCPG code has been developed to generate elemental library spectra for all elements that yield gamma rays in the C/O tool.

3. Correction of pulse pile-up distortion

Pulse pile-up distortion is a common problem for pulse-height measurement at high counting rates. High counting rates are "a must" for many medical and industrial applications of radiation measurement to ensure good counting statistics in the shortest possible diagnosis or on-line measurement times. Modern advanced electronics may reduce "dead time" and increase the possible undistorted counting rate levels somewhat. However, this is usually with resolution and unknown variance penalties. In most cases, this distortion can be completely corrected by mathematical means without these penalties. CEAR has been working for some time on an off-line approach and more recently on an on-line approach.

The off-line approach developed at CEAR consists of first developing an accurate Monte Carlo code CEARPPU (Gardner and Lee, 1999; Guo et al., 2004b, c) to treat the forward calculation of the pile-up distorted spectrum from the known (or assumed) true spectrum. This is in the Public Domain-PSR-528 @ RSICC.ORNL (available from the Radiation Safety Information Computational Center (RSICC) at Oak Ridge National Laboratory, ORNL). Second, the Monte Carlo pile-up to true (MCPUT) approach (Guo et al., 2005) was introduced for correcting the pile-up distorted spectra to true spectra, by an iterative procedure. The CEARPPU code is fast enough (one case takes a minute or two) that it can be iterated to give the required true spectrum. Benchmark experiments are carried out for both codes with an Fe-55 source and a Si(Li) detector. Two measured spectra for low and high counting-rate measurement are compared in Fig. 1. This figure shows the true, double, triple, and quadruple and higher pile-up regions of a high counting-rate spectrum. The simulated high counting-rate measurement by CEARPPU is compared with the experimental one in Fig. 2 and shows excellent agreement. After implementing the MCPUT approach, the corrected high counting-rate spectrum fitted by the measured true spectrum is compared with the
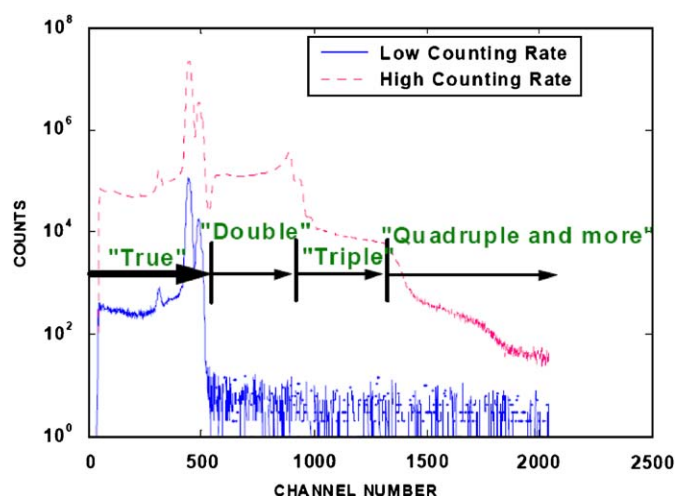
Fig. 1. Comparison of the Fe-55 spectra measured with low and high counting rates. Four regions are marked according to their major contribution of pulses. More accurately, double piled-up pulses contribute to both "True" and "Double" regions, and so on.
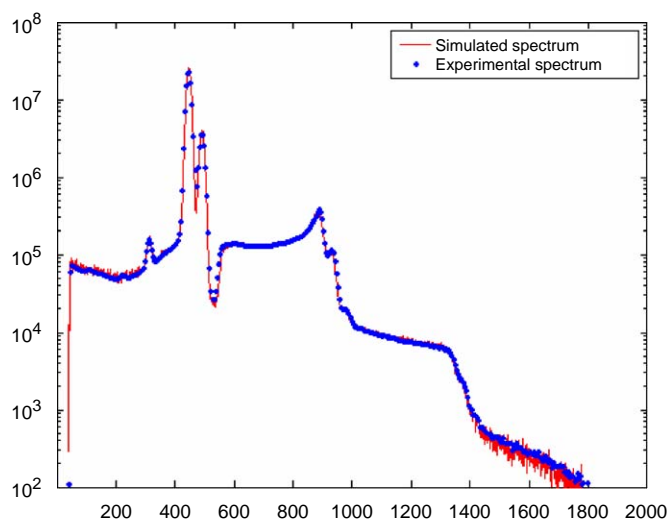

Fig. 2. Spectrum comparisons between simulated high counting-rate spectrum and experimental spectrum.
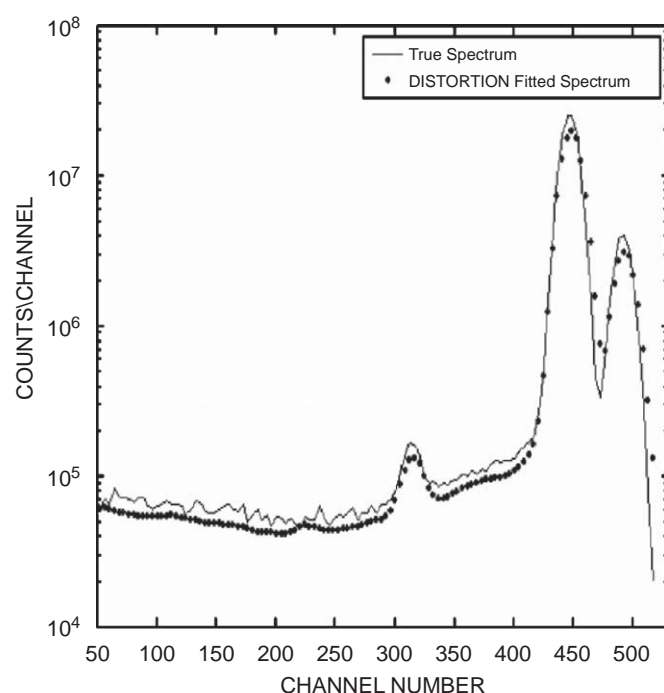

Fig. 3. Comparison of the Fe-55 measured true spectrum and the calculated true spectrum obtained by using the distorted high counting-rate measured spectrum.
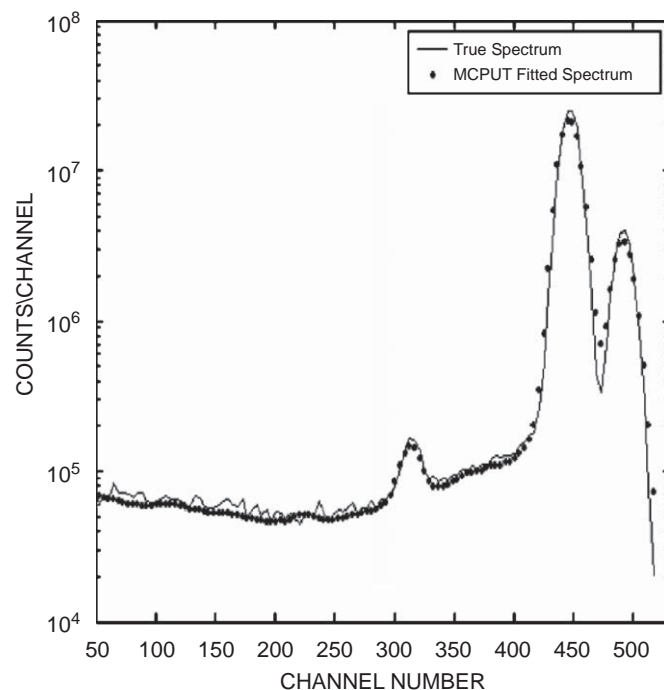

Fig. 4. Comparison of the measured true spectrum and the calculated true spectrum obtained by using the MCPUT corrected high counting-rate spectrum.

measured high counting-rate spectrum fitted with the measured true spectrum in Figs. 3 and 4. The reduced chi-square was reduced from 3.6 to 1.2 by using the MCPUT correction. The advantages for this off-line approach are: (1) for a constant or known form of the counting rate with time, it is very accurate; (2) if the range of the true pulse-height energy is known, it can even treat random true coincidences and iterations converge rapidly; and (3) under these specified conditions, the true spectrum is generated with original resolution and Poisson variance. The disadvantage is that the necessary conditions may not exist.

When the conditions necessary for using the off-line approach are not present, an on-line, real-time approach may be more appropriate. CEAR has been working on the use of a digitizer and a PC to replace the use of multi-channel analyzers (Guo et al., 2005). The preliminary studies have been promising. This approach consists of digitizing the signal at the preamplifier output without further pulse shaping. Then differentiation of this signal train and use of simple pulse models allows the generation of the true pulse-height spectrum. A Monte Carlo simulation code was devised to illustrate the effect of applying the proposed deconvo-

lution approach on radiation spectroscopy at high counting rates. An imaginary Cs-137 source was used to irradiate the NaI(Tl) scintillation detector at the variable counting rates: 1 k and 4 Mcps, respectively. The simulated spectra are shown in Fig. 5. It is obvious that the standard features in a gamma-ray spectrum are completely smeared at 4 Mcps. Fig. 6 shows the simulated spectra acquired when the real-time deconvolution approach was applied. The peak resolution at 4 Mcps is still maintained very close to the
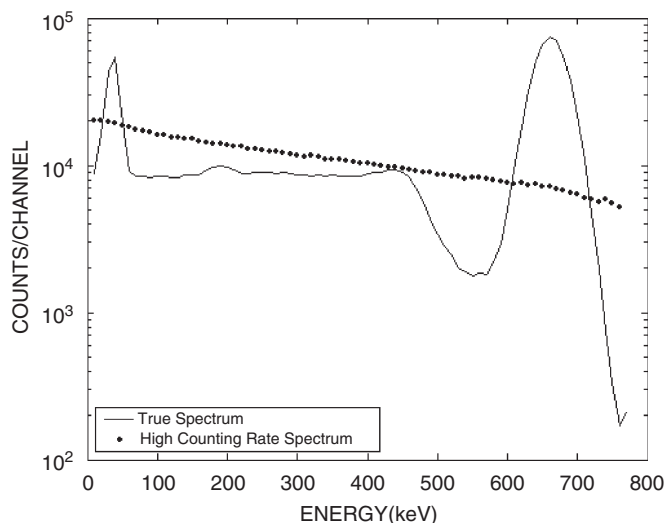
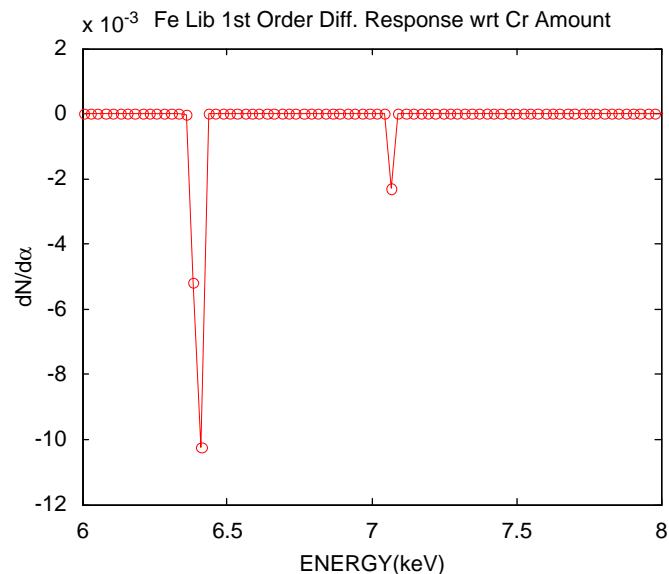**Fig. 5.** NaI(Tl) spectra for variable counting rates without application of the pulse pile-up deconvolution approach.
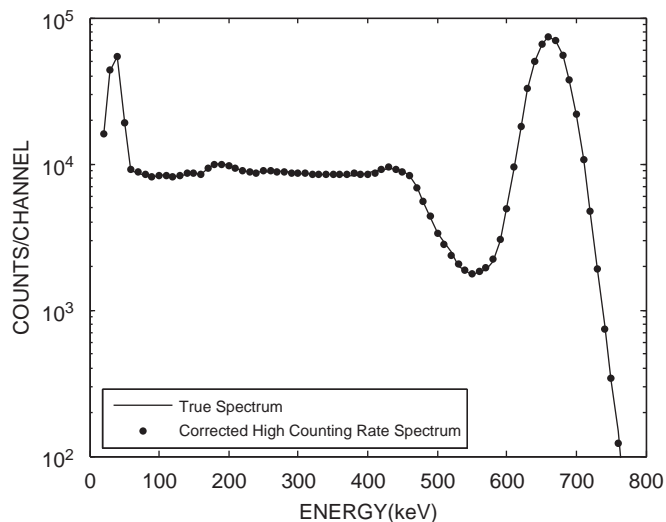

Fig. 6. NaI(Tl) spectra for variable counting rates with the pulse pile-up deconvolution approach applied.

Fig. 7. Simulated XRF spectra for alloy samples of SS316 and SS304.

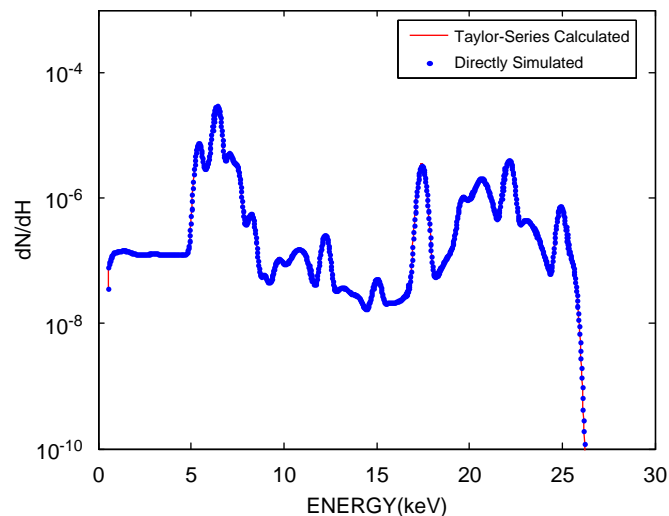**Fig. 8.** Simulated differential response for Fe.

Fig. 9. Spectrum comparisons between directly simulated SS304 XRF spectrum and using SS316 as initial guess values and DO calculated SS304 XRF spectrum.

one at the low counting rate of 1 kcps. The overall spectral shape was maintained and the chi-square test for these two spectra reported a reduced chi-square value of 1.6 (Figs. 7–9).

4. Detector response functions

The DRF is used to predict pulse-height spectra in the MCLLS approach for the inverse elemental analysis of PGNAA and EDXRF. Our use of the DRF consists of first simulating the detector-incident X-ray spectral flux by the CEARXRF code, or prompt gamma-ray spectral flux by the CEARCPG code, then convoluting them with a detector response function to transform it into detected pulse-height spectra. The advantages of using this DRF approach are: (1) on the order of one-half of the calculations per history can be omitted by the use of a DRF since the transport of particles inside the detector are not necessary, (2) use of the DRF has a natural smoothing effect which reduces the number of histories necessary for the desired accuracy by a factor of about 100 (Metwally et al., 2004a), and (3) use of the DRF yields better
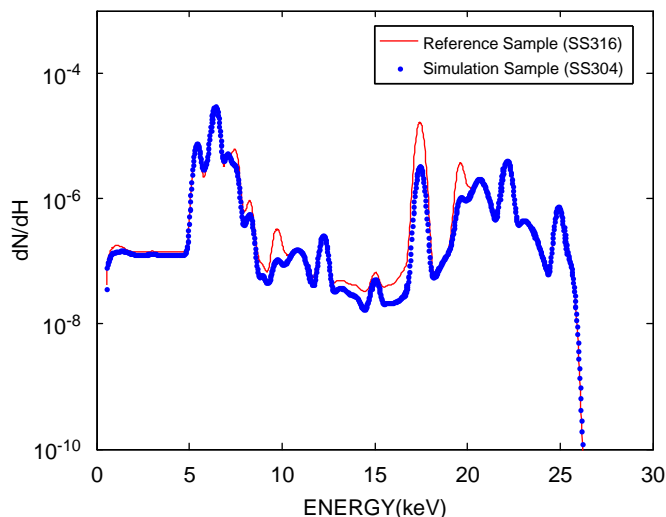
accuracy in spectral simulations because it is more accurate than calculations of particle transport with existing physics inside the detector. Present cross sections are for amorphous materials and scintillation and semi-conductor detectors are single crystals.

Presently, there are no general purpose codes like MCNP that can accurately simulate gamma-ray and X-ray DRF's for either scintillation or semi-conductor detectors by using the normal physics of photon and electron transport (Sood and Garden, 2004; Gardner and Sood, 2004). The major difficulties for scintillation detectors, such as the NaI detector, includes energy resolutions, flat continua, and the non-linear scintillation efficiency. For semi-conductor detectors, this also includes exponential tails on the low-energy side of the full-energy peaks. CEAR has been working on these problems for more than 20 years. A semi-empirical model of the DRF for a Si(Li) detector (He et al., 1990) and a low-energy photon germanium detector (Jin et al., 1986; Lee et al., 1987; Gardner et al., 1986) are currently available and used in the CEARXRF simulation. A specific purpose Monte Carlo simulation
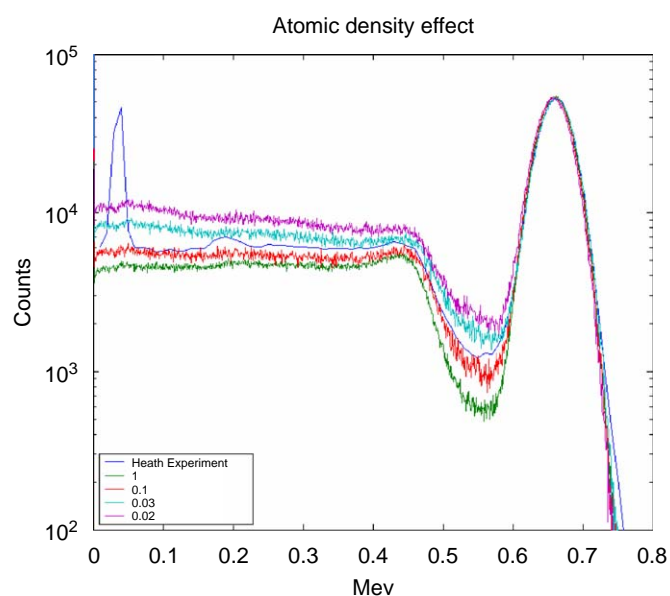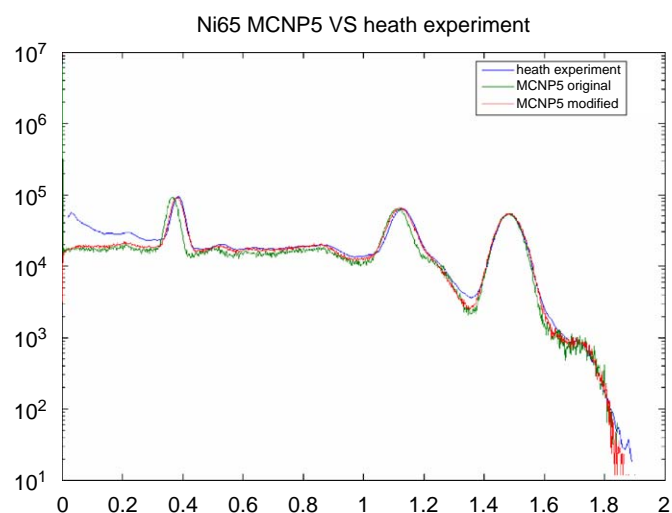

Fig. 12. A comparison of original and electron non-linear efficiency modified MCNP5 simulations with Heath NaI experimental spectra for Ni-65.

code named g03 (Gardner and Sood, 2004; Sood, 2000) was developed to provide DRF's for NaI(Tl) scintillation detectors, which are used in the CEARCPG codes.

The general purpose code MCNP5 is now being modified to include the capability of simulating DRF's for scintillation detectors. The main changes are focused on (1) adjusting the electron densities of detector materials to allow more electrons to escape from the detector surfaces to make changes in the predicted flat continua and (2) implementing non-linear energy transfer from electron energy to effective energy, which eventually induces scintillation. Some preliminary results are shown in Figs. 10–12.

5. Differential operators

The MCLLS approach suffered from the disadvantage that the Monte Carlo simulation takes a long calculation time, so that the iteration steps were not practical. To overcome this problem, DO method was combined with MCLLS.

The DO method is discussed in the literature (Hall, 1982; Rief, 1984; Rief, 1994) and it is a very powerful tool for measurement sensitivity studies and system optimization. The Monte Carlo-differential operator approach was implemented in CEARXRF for X-ray fluorescence (Guo, 2003; Guo and Gardner, 2004) to simulate differential responses of both sample and elemental library spectra for variations of elemental concentrations. By using the Taylor series expansion, these differential responses can be used for spectra adjustment according to possible weight fraction differences between initial guess values used for Monte Carlo simulation and true values of the unknown sample. This is potentially a very accurate approach for taking account the non-linear EDXRF response due to inter-elemental absorption-enhancement effects. The implementation of DO in CEARCPG for neutron capture prompt gamma-ray analyzers is still ongoing.

Monte Carlo simulations are conducted to illustrate the effect of using DO. Two stainless steel samples (SS316 and SS304) were irradiated with Cd-109 and the fluorescence X-rays are detected with a Si(Li) detector. The simulated spectra are shown in Fig. 7. It is obvious that the differences are caused by different elemental weight fractions. After using DO combined with CEARXRF, the differential elemental responses are calculated. One example of the Fe differential response is given in Fig. 7. By using alloy sample SS316 as the initial guess values, together with the previously

Fig. 10. Simulated Cs-137 NaI detector spectra for various pseudo detector densities to simulate the effect of electron transport losses from the detector surfaces.
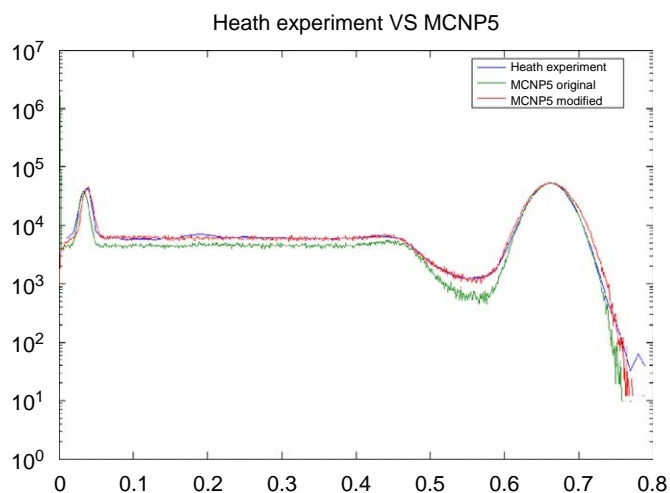

Fig. 11. A comparison of original and flat continuum modified MCNP5 simulations with Heath NaI experimental spectra for Cs-137.

calculated differential elemental responses, the SS304's spectra, which is shown in Fig. 8, is calculated based on the weight fraction differences between SS316 and SS304 after several iterations.

6. MCLLS when using coincidence spectra

The coincidence technique has been applied to various fields to improve measurement sensitivity and to better understand the physical processes involved. The fundamental idea of the coincidence technique is to detect multiple events that are correlated in time of emission, which has the effect of minimizing the background. Compared with single spectra, coincidence spectra are much cleaner and sharper. With coincidence spectra replacing single spectra in implementing MCLLS, more accurate elemental analysis should be obtained. The feasibility study of using coincidence spectra in MCLLS has been done by Metwally et al. (2004a, b). 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. The coincidence results shows a much better accuracy and more resistant to pulse pile-up distortion than single results. The reduced chi-square is minimized from 43.3 to 3.6 by using coincidence spectra in MCLLS to replace single spectra. Inspired with the successful trial, CEAR extended the idea to PGNAA for bulk analysis, EDXRF, and C/O tool applications.

6.1. PGNAA

PGNAA is generally suffers from low signal-to-noise (S/N) ratio. The major background sources include the gamma rays from the naturally occurring radioisotopes (K-40, uranium, and thorium), background from the neutron source including the source-emitted gamma-rays and that generated by neutron activations of surrounding materials, and the detector itself. Normally, hydrogen causes problems in many typical cases of PGNAA. When hydrogen is activated, a single prompt gamma ray is emitted with energy of 2.223 Mev. The intensity of this peak is often one or two orders of magnitude higher than those from other elements of interest. This complicates the spectrum, particularly at energies less than or equal to 2.223 Mev.

For most elements, prompt gamma rays are emitted in coincidence. The study of coincidence counting approach for PGNAA applications at CEAR was started by Gardner (2000), and continued by (Metwally, 2002). The typical experimental setup is shown in Fig. 13. A Cf-252 neutron source was placed in the middle of a lead cube to attenuate the source gamma rays. The

cube was placed over a layer of wax to thermalize the fast neutrons before them interacting with the sample box containing sample. Finally, two detectors were mounted on wooden table side-by-side. The detectors were covered by lithium sheets to absorb neutrons, and the sides were wrapped with lead to reduce cross talk. Recently, the CEARCPG code was developed to generate both single and coincidence spectra for PGNAA applications. Several cases are simulated and benchmarked by experiments. Results of pure sulfur sample by using this experimental setup are shown in Fig. 14. The peaks which appear in the measured spectra are tabulated in Table 1. Comparing the coincidence spectrum and single spectrum, improvements are noticed and can be summarized as follows: (1) complete elimination of the hydrogen 2.223 Mev peak #3, the potassium 2.61 Mev peak #4, and the NaI 6.826 Mev peak #11, and (2) the sulfur peaks are more obvious. The two-dimensional coincidence experimental spectrum is shown in Fig. 15 and compared with simulated results. The Q-value diagonal projection is applied and shown in Fig. 16 and compared with simulated results. The energy window is the Q-value of $^{32}S(n,\gamma)^{33}S$ reaction, which is 8.641 Mev. It can be seen that the Q-value diagonal projection spectrum has much better resolution and signal-to-noise ratio compared to ordinary single spectrum.

The investigation of replacing single spectra with coincidence spectra in MCLLS for PGNAA has been conducted at CEAR. Some preliminary results have been reported by Han (2005). Both total coincidence spectra and diagonal projection coincidence spectra are utilized. The CEARCPG code is also capable of generating elemental diagonal projection spectra. Since the diagonal projec-
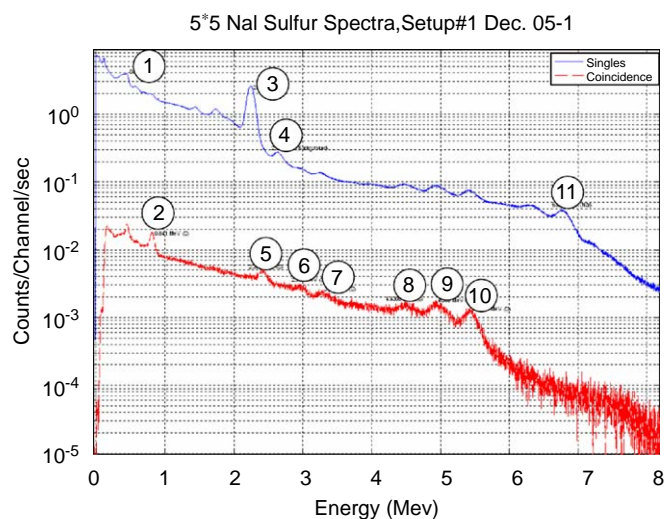

Fig. 14. Measured single and coincidence spectrum for sulfur sample.

Table 1
Peak energies of pure sulfur sample shown in Fig. 14.

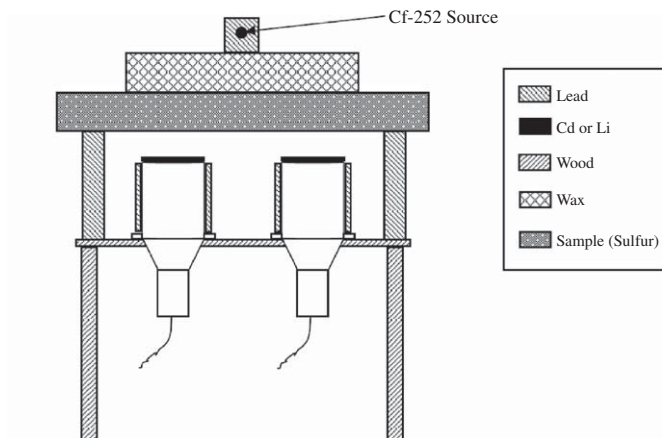| Peak	Energy (MeV)	Source
1	0.511	Pair production
2	0.841	Sulfur
3	2.223	Hydrogen
4	2.61	Potassium-40
5	2.379	Sulfur
6	2.931	Sulfur
7	3.22	Sulfur
8	4.4308	Sulfur
9	4.869	Sulfur
10	5.4205	Sulfur
11	6.826	NaI

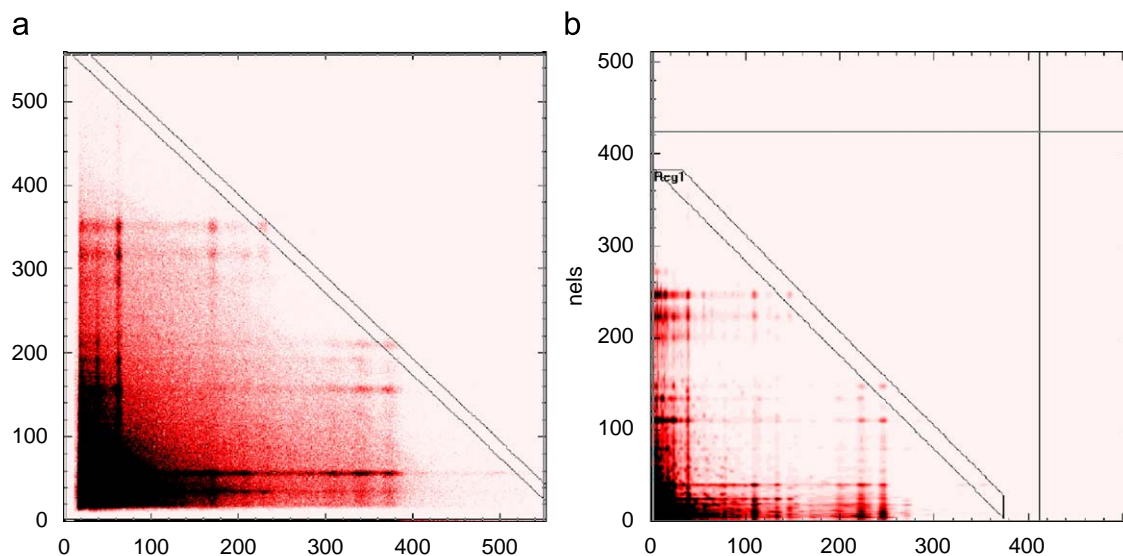**Fig. 13.** Experimental arrangement for the coincidence measurement of sulfur.

Fig. 15. The coincidence spectra of a pure sulfur sample: (a) the experimental coincidence spectrum and (b) the calculated coincidence spectrum.
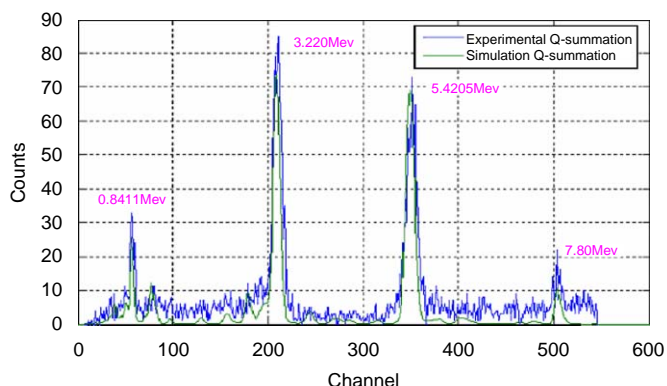

Fig. 16. Comparison between the experimental spectrum and the simulated spectrum for the Q-value diagonal summing.

Fig. 17. Schematics for CXRF experimental geometry.

tion spectra has better characteristic over the total coincidence spectra, using the diagonal projection spectra is a better way to improve the accuracy. Different diagonal energy windows may be applied depending on the element of interest. For gamma-rays measurement, most of interferences concentrate in the low-energy region. The high-energy window projection can get a good resolution spectrum and have fewer peaks. It already has been proved in the sulfur and mercury experiment.

6.2. EDXRF

The main interfering background for the *in vivo* EDXRF measurement of lead in bone was found to be from the back-scattered Compton and Raleigh source photons, which also limits the improvement of sensitivity. Based on the physics of atomic transition, certain fractions of the characteristic K-series X-rays and L-series X-rays are in true coincidence.

Coincidence X-ray fluorescence (CXRF) study at CEAR was conducted by Guo (2003). Two experiments were made for CXRF measurement. For both experiments, one rectangular piece of pure lead of 2 mm thickness was used as the sample. Cadmium-109 was used as the activation source, which was collimated in the direction of sample. One of the experiments used two X-ray NaI(Tl) detectors (25 mm radius × 1 mm thickness), Fig. 17-a, and the other one substituted one detector by a low-energy Ge (LEGe) detector (25 mm radius × 15 mm thickness), Fig. 17-b. The measured single and coincidence spectra for the two NaI(Tl) detectors and for one NaI(Tl) and one LEGe detector is shown in Fig. 18-a and -b, respectively. It is clear that the scattered source photons (both Cd-109 88 keV Gamma rays and Ag X-rays, 22.2 and 24.9 keV) are minimized in the coincidence spectra. The Pb K and L X-ray intensities are enhanced relative to the continuum. The 2-D histograms were sorted for both measurements (Fig. 19-a and -b). The X- and Y-axis in Fig. 19 represent the energy information of each detector, respectively. The intensity of each point in the figure represents the measured intensity of the coincidence event with energy deposition in the two detectors as the corresponding values on the X and Y axes.

A Monte Carlo simulation code named CEARXRC has been developed to generate both single and coincidence spectra. The geometry in Fig. 17b was simulated to benchmark the Monte Carlo simulation code. This diagonal summing coincidence spectrum is compared to the Monte Carlo simulated spectrum in Fig. 20 and shows good agreement. By using these simulated coincidence spectra in MCLLS for lead amount analysis, a factor of 10 (Guo, 2003) is improved in the sensitivity compared to the analytical results by single spectra.

6.3. C/O

The traditional C/O tool has always had relatively low detection efficiency, poor resolution, and small signal-to-noise ratio. The reasons for this are primarily that the detector diameters are limited in size by the necessary small tool diameters. To offset this problem, CEAR proposes the use of the entire collected spectrum by using the Monte Carlo library least-squares approach, which involves the generation of complete elemental libraries by Monte Carlo simulation.
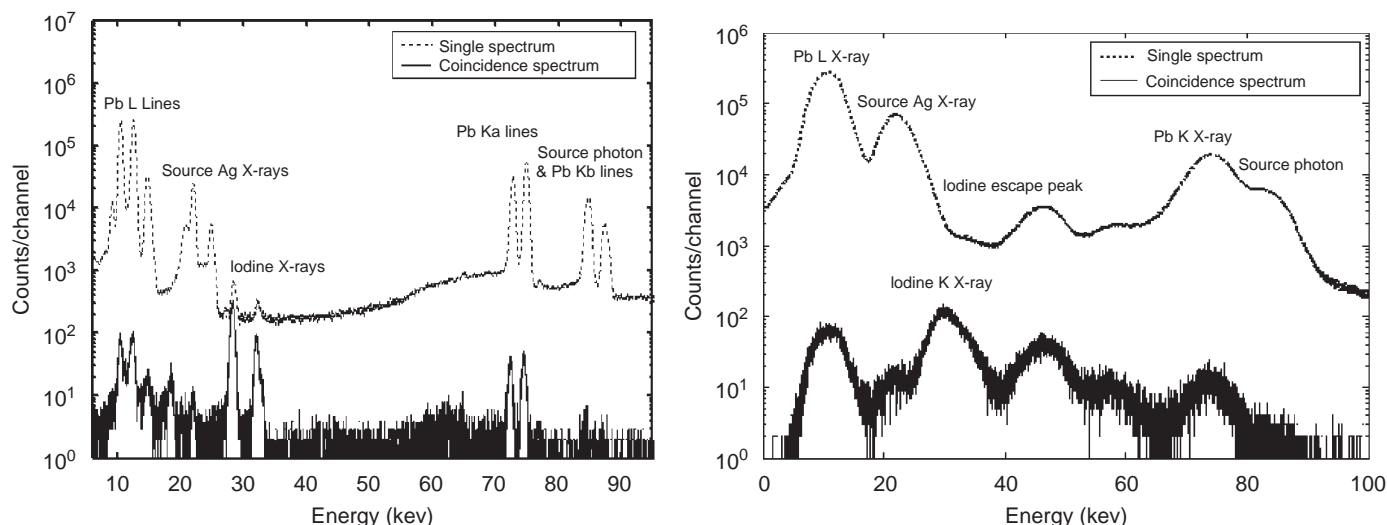
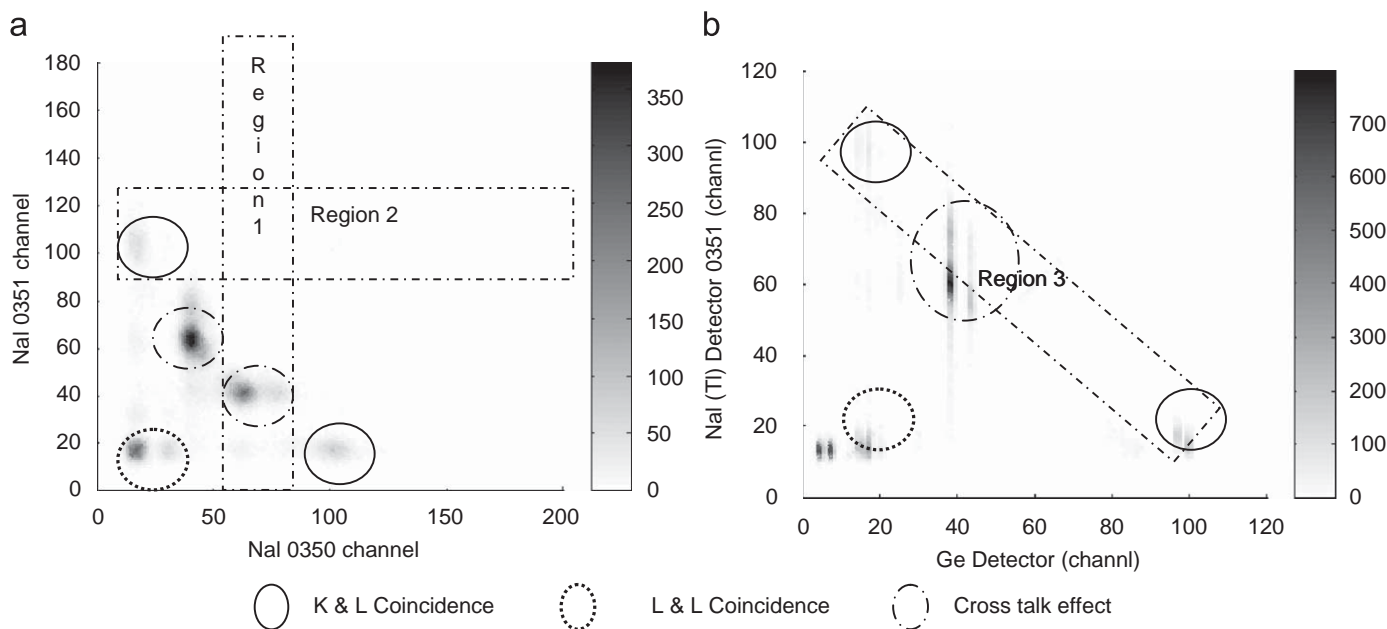**Fig. 18.** Experimental singles and coincidence spectra comparison of CXRF.

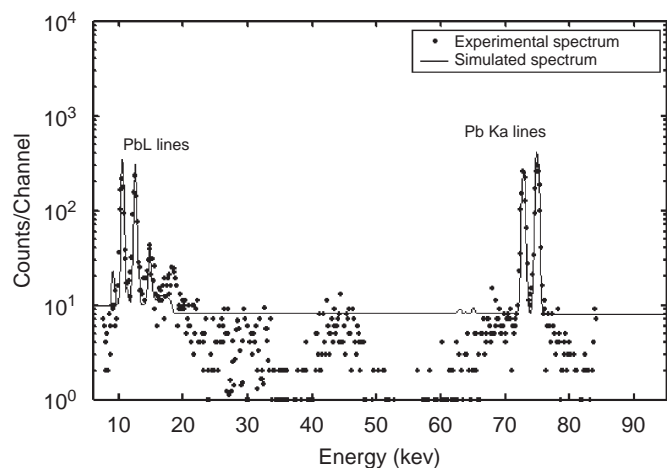**Fig. 19.** 2-D histograms for CXRF measurement.

Fig. 20. Comparison of simulated and experimental spectrum.

The specific purpose Monte Carlo code CEARCO has been developed and used for this purpose. The elemental library spectra for all elements that yield gamma rays in the C/O tool are generated by first using the Monte Carlo code CEARCO to generate the elemental library spectra incident on the detector. These library spectra are then used with unknown mixture C/O spectra to calculate the amounts of C, O, and other elemental amounts present by the LLS approach. It is estimated that this approach produces results with uncertainties between two and three times smaller than the use of the window approach that uses only peak intensities with the available data.

7. Conclusions

Results so far indicate the MCLLS approach is accurate for both EDXRF and PGNAA. The CEARXRF and CEARPGA codes and a DRF for the appropriate detectors provide all that is needed for the

inverse problem. The GUI that has been developed and differential operators added to CEARXRF makes the approach practical for EDXRF. We are doing the same for PGNAA. Now, we need to develop the approach for all commercial analyzers—including those with X-ray machines and secondary fluoresces for EDXRF. For Routine EDXRF Sample Analysis the Advantages of this Approach are:

Use of CEARPPU makes all the data available with known Poisson statistics. Use of MCLLS corrects for all matrix effects including tertiary and beyond. It will be easy to include other refinements (such as electron transport) as necessary. Use of LLS avoids all problems with intensity measurement and gives statistical estimates of results automatically. Coincidence counting for both EDXRF and PGNAA may provide a significant increase in signal-to-noise (S/N) ratio by essentially eliminating the background (noise). Since sensitivity and accuracy usually depend on S2/N, removing almost all of the noise does not guarantee an improved device. The signal must not be reduced less than some critical amount.

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