Use of Monte Carlo Simulation Methods to Improve X-ray Detector Response Function

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Abstract

An accurate Si (Li) detector response function is needed for many applications such as X-ray spectroscopy and elemental analysis. A semi-empirical detector response function using Gaussian peak models and exponential tails was used to characterize the detector response functions. And the use of Monte Carlo method to simulate the detector response has been developed to predict both spectral shape characteristics and detector efficiency for any incident X-ray energy. The benchmark of the detector response function on Si (Li) detector was performed on elements Al, Si, Ti, V, Cr, Mn, Co, Ni, Cu, Zn, and Pb, which are excited by a 17.5 keV mono-energetic X-ray source using a micro-focused X-ray spectroscopy analyzer. The semi-empirical parameters are optimized by nonlinear regression with experimental spectra of pure-element samples. The model fitted results are presented and indicate good agreement with experimental data. The detector response functions are pre-calculated with high statistical precision. Consequently, it can be used in X-ray spectroscopy and elemental analysis with great accuracy.

Introduction

An accurate representation of Si (Li) detector response functions (DRFs) is needed in many applications [Statham 1976, McNells and Campbell 1975, Jorch and Campbell 1977, Gardner and Li 2006, Li and Gardner, 2008, Scholze and Procop 2009]. Monte Carlo simulation method has been used to generate the entire pulse-height spectrum and individual elemental spectra for the energy dispersive X-ray fluorescence (EDXRF) elemental analysis. While Monte Carlo models predict the photon spectra incidents on X-ray detectors, the detector response functions are used to generate the pulse-height spectra, which is a powerful variance reduction technique to speed up the Monte Carlo simulation process with enhanced statistical precision.

The detector response function is defined as the pulse height distribution for any incident mono-energetic X-ray, usually indicated by \( R (E', E) \), where \( E' \) is the pulse height energy and \( E \) is the incident \( \gamma \)-ray energy. Generally, semi-empirical DRF models are a set of probability distribution functions that are always larger than or equal to zero over the entire energy range and integrate over all \( E' \) to unity. Monte Carlo-based detector response functions are not only able to provide the spectral probability distribution, but also supply detection efficiency to generate pulse height spectrum accurately. The flux spectrum on the detector surface is calculated by X-ray Monte Carlo simulation software such as center for engineering applications of radioisotopes X-ray fluorescence (CEARXRF) code [Li 2008]. Surface flux spectra are then transformed into pulse-height spectrum when convolving with the optimized detector response functions. The
convolution of surface flux spectrum $\Phi(e)$ and DRFs is defined as the integral of the product of two functions after one is reversed and shifted. As such, pulse height spectrum is a particular kind of integral transform,

$$\text{PulseHeight} = (\Phi \ast \text{DRF})(E) = \int_{E_{\text{min}}}^{E_{\text{max}}} \Phi(e) \text{DRF}(E - e) \, de$$

It is essential to develop an accurate model for detector response functions for the convolution purpose. Several studies on developing the Si (Li) detector response function models have been reported. Yacout et al. (1986) used a semi-empirical response function model which consists of five major components:

1. Gaussian-shaped full-energy peak,
2. Gaussian-shaped Si escape peak,
3. flat continuum from zero to full energy,
4. a long-term exponential tail on the low-energy side of the full-energy peak, and
5. a short-term exponential tail on the low-energy side of the full-energy peak.

The model developed by Yacout is suitable for energy range from 5 to 20 keV. He et al. (1990) extended the model to useful energy range from 5 to 60 keV. These models treat detector response functions as probability distribution function. The model parameters are normalized constants such that:

$$\int R(E, E') \, dE' = 1$$

Consequently, this model considers the detector response distribution shape only and there is no detector efficiency information available. Also, the model is suitable for one incident X-ray line each time. In practice, there exist multiple characteristic X-ray energy lines for an element. For instance, the pair of $k\alpha$ and $k\beta$ lines is often detected together in the X-ray measurement system.

To overcome these limits, the authors improved the model to consider the cases of multiple incident X-rays and used Monte Carlo simulation method to calculate the detector efficiency. The updated model is given by:

$$R(E, E') = \sum_{i=1}^{n} \{ A_1(E_i) G(E_i, E', \sigma(E_i)) + A_2(E_i) G[E_i - 1.74, E', \sigma(E_i - 1.74)]\} \ldots$$

$$\ldots + \{ A_3(E_i) + A_4(E_i) \exp[\alpha(E_i)(E_i - E')] \} \times F[E_i, E', \sigma(E_i)] \}$$

Where $E_i$ is incident X-ray energy in keV for every incident energy (i) and $E'$ is the portion of the energy deposited in X-ray detector. $G(E_i, E', \sigma(E))$ is defined as a Gaussian function with peak standard resolution ($\sigma$), representing both the Gaussian-shape full-energy peak and Si escape peak. $F(E_i, E', \sigma(E))$ is given by an error function
(erfc), representing the flat continuum and exponential-tail terms, and \( \alpha(E_i) \) is the exponential exponent. The definitions of \( G \) and \( F \) are given by:

\[
G[E_i, E', \sigma(E_i)] = \exp\left[ -\frac{(E'-E_i)^2}{2\sigma^2} \right]
\]

\[
F[E_i, E', \sigma(E_i)] = \pi \times \text{erfc}\left[ -\frac{(E'-E_i)^2}{\sqrt{2}\sigma^2} \right]
\]

Where

\[
\text{erfc}(t) = \frac{2}{\sqrt{\pi}} \int_t^\infty \exp(-x^2) dx
\]

As demonstrated in the model, \( n \) is the number of incident X-ray energies. For example, two incident energies are commonly observed corresponding to K\( \alpha \) and K\( \beta \) peaks. For these cases, \( n=2 \). This model is also suitable for those cases when X-ray energy lines are larger than 2.

**Experiments and Detector Resolution**

The experimental data are obtained from a micro-focused XRF analyzer, which is an elemental analysis technique which uses doubly curved crystal (DCC) optics to enhance measurement intensities by capturing X-rays from a divergent source and redirecting them into an intense focused beam on the surface of the product [Chen et al. 2002, 2008]. Figure 1 shows the diagram of micro-focused XRF spectrometry used to perform XRF measurement. The DCC optic transforms the divergent polychromatic X-rays generated from an X-ray tube into a micro-focused monochromatic excitation source. This system has the benefits of low scattering background under the fluorescence peaks and greatly enhancing detection performance. This new analytical approach results in detection limits in the parts-per-billion (ppb) range for many elements of interest in a variety of commercial materials.
basis to optimize the parameters of semi-empirical detector response functions as discussed above.

The detector resolution (R) is an essential part of detector response functions and can be determined by investigating the Gaussian-shape peak width on measured spectra. The energy resolution for an X-ray detector is defined as:

\[ R = \frac{FWHM}{E_0} \] … …. 4)

where FWHM is defined as the full width at half maximum of the photopeak and \( E_0 \) is the photopeak energy. The model for a Gaussian-shape photopeak is described by:

\[ g(E) = \frac{1}{\sigma \sqrt{2\pi}} \exp \left[ -\frac{(E - E_0)^2}{2\sigma^2} \right] \] … …. 5)

where \( \sigma \) is the standard deviation of the photopeak. The relationship between FWHM and the standard deviation can be determined as:

\[ FWHM = 2.35\sigma \] … …. 6)

To get the relationship of detector photopeak standard deviation (\( \sigma \)) with respect to X-ray energy, a non-linear model is used:

\[ \sigma = \sqrt{a + b \cdot E} \] … …. 7)

where \( E \) is the X-ray energy in keV and \( \sigma \) is the peak standard deviation in keV. The parameters (a, b) are empirically determined by matching experimental spectral standard deviation (\( \sigma \)) at each photopeak energy (E). Table 1 shows the detector standard deviation (\( \sigma \)) variation with photopeak energy (E) for the Si (Li) X-ray detector. The regression results using equation 7) gives \( a=0.00177 \) and \( b=0.000455 \).

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>( \sigma ) (keV)</th>
<th>FWHM (keV)</th>
<th>Resolution (R, %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.508</td>
<td>0.061</td>
<td>0.143</td>
<td>3.173</td>
</tr>
<tr>
<td>4.922</td>
<td>0.065</td>
<td>0.154</td>
<td>3.119</td>
</tr>
<tr>
<td>5.409</td>
<td>0.064</td>
<td>0.151</td>
<td>2.791</td>
</tr>
<tr>
<td>5.941</td>
<td>0.067</td>
<td>0.158</td>
<td>2.657</td>
</tr>
<tr>
<td>5.892</td>
<td>0.066</td>
<td>0.155</td>
<td>2.628</td>
</tr>
<tr>
<td>6.484</td>
<td>0.069</td>
<td>0.163</td>
<td>2.510</td>
</tr>
<tr>
<td>6.924</td>
<td>0.069</td>
<td>0.163</td>
<td>2.359</td>
</tr>
<tr>
<td>7.646</td>
<td>0.073</td>
<td>0.172</td>
<td>2.252</td>
</tr>
<tr>
<td>7.471</td>
<td>0.072</td>
<td>0.168</td>
<td>2.252</td>
</tr>
<tr>
<td>8.265</td>
<td>0.075</td>
<td>0.176</td>
<td>2.128</td>
</tr>
<tr>
<td>8.042</td>
<td>0.073</td>
<td>0.172</td>
<td>2.144</td>
</tr>
<tr>
<td>8.907</td>
<td>0.076</td>
<td>0.179</td>
<td>2.013</td>
</tr>
</tbody>
</table>
To obtain an accurate DRF, the X-ray source and pulse shaping time ought to be optimized to minimize the effects of pulse pile up (PPU) on the counting rate measurement. The dead-time is less than 1% in this experiment and thus the pulse pile up problem on this particular application is negligible. However, for high counting rate measurement, pulse pile up effects on X-ray spectral distortion cannot be neglected and should be corrected before further analysis. In order to correct the distortion of the spectrum, a CEAR Pulse Pile-Up program (CEARPSPU - forward PPU calculation code and CEARIPPU – the inverse PPU correction code) can be used to handle this problem and obtain the “true” spectrum [Gardner et al, 2004]. There are many other methods to correct pulse pile up distortion such as the real-time deconvolution of digitized waveforms [Guo et al, 2005] and pileup correction algorithms for very high count rate spectrometry [Bolic et al, 2010].

Spectral Regression

Nonlinear regression code (CURMOD) [Gardner et al, 2011] was used to determine both the linear (A₁, A₂, A₃, A₄, etc) and nonlinear parameters (α) of the response functions simultaneously. Initial guesses of the nonlinear parameters are provided before the fitting and the code calculated the optimized coefficients for the model to fit the experimental library spectra the best. The fitting reduced chi-square values for each element target are included in Table 2 and Figure 2 show the fitted model together with all of its components for Copper element and compared with the measured spectrum. They show that the improved semi-empirical DRF models have very good agreements with the experimental spectrum.

Table 2 Reduced chi-square values for detector response functions regression with experiments

<table>
<thead>
<tr>
<th>Element</th>
<th>Reduced chi-Square values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al</td>
<td>6.59</td>
</tr>
<tr>
<td>Si</td>
<td>5.73</td>
</tr>
<tr>
<td>Ti</td>
<td>6.24</td>
</tr>
<tr>
<td>V</td>
<td>6.35</td>
</tr>
<tr>
<td>Cr</td>
<td>6.1</td>
</tr>
<tr>
<td>Mn</td>
<td>5.77</td>
</tr>
<tr>
<td>Co</td>
<td>4.56</td>
</tr>
<tr>
<td>Ni</td>
<td>4.48</td>
</tr>
<tr>
<td>Cu</td>
<td>3.56</td>
</tr>
<tr>
<td>Zn</td>
<td>3.42</td>
</tr>
<tr>
<td>Pb</td>
<td>9.75</td>
</tr>
</tbody>
</table>
As shown in figure 2, both Kα and Kβ peaks of Copper elements and their associated Si escape peaks are demonstrated. Full-energy peak 1 is corresponding to Copper Kα energy (8.043keV) and the Si escape peak energy is 6.313keV. And full-energy peak 2 is Kβ energy (8.897keV) and the Si escape peak is equal to 7.167keV. It is noted that the difference between full-energy peak and Si escape peak is fixed (1.73keV). The figure also shows the flat continuum from 0 keV up to the full-peak energy. Both the long-term and short-term exponential tails are located at the low-energy side of the full-energy peaks.

The linear and non-linear parameters are obtained from regression of the experimental spectra for eleven pure-elements, corresponding to a pair of X-ray energies (Kα and Kβ) for every element. These parameters are dependent on X-ray energy and fitted with a set of non-linear models with respect to the variable incident energy. After these models are available, the parameters can be obtained for an input energy and the DRF can be generated to predict detector response to any incident X-ray energy within the applicable range.

The fitted result for the normalized Gaussian-shape full-energy peak amplitude A₁ (E) is given by:

\[ A₁(E) = -4.745 \times \exp(-0.364 \times E) + 55.771 \times \exp(-1.604 \times E^{0.202}) \]

where E is given in keV. The fitted function is shown in Figure 3.
The fitting result for the normalized Gaussian-shape Si-escape peak amplitude $A_2(E)$ is given by:

$$A_2(E) = 2.087 \times E^{-2.684}$$

where $E$ is in keV. As the X-ray energy increases to 17 keV or above, no escape peak is observed. Therefore there is no need to consider Si escape when incident energy is larger than 17 keV. The fitted curve is shown in Figure 4.

Similar to $A_1(E)$, a two-component fit for the normalized flat-continuum amplitude is given by:

$$A_3(E) = 3.815 \times \exp(-2.214 \times E) + 0.00157 \times \exp(-3.34 \times E^{0.0635})$$

and the exponential-term amplitude was fitted to the function given by:

$$A_4(E) = 0.0138 \times \exp(-0.00172 \times E^{4.363}) + 4.384 \times 10^{-5} \times \exp(1.44 \times E^{0.528})$$

where $E$ is in keV. The fitted curves for flat-continuum amplitude and exponential-term amplitude are shown in Figure 5. The exponential-term exponent $\alpha(E)$ was fitted to the function:

$$\alpha(E) = 0.51 \times E^{-0.749}$$

where $E$ is in keV and the fitted curve is shown in Figure 6.
Figure 4 Fitted Si-escape peak amplitude and standard deviation with respect to incident energy.

Figure 5 Fitted flat continuum amplitude and exponential tail amplitude with incident energy.

Figure 6 Fitted $\alpha$ (E) coefficient with incident energy.
Monte Carlo Simulation

The semi-empirical detector response model considers only the major energy lines (Kα and Kβ). In reality, the X-ray analyzer system also records the interactions of photons with surrounding contacts which will produce spectral variations as demonstrated by the multiple scattered energy lines (surface flux in Figure 8). The Monte Carlo-based gamma-ray detector response function (GAMDRF) code [Li et al, 2011] can take full photon interaction and relaxation physics into account and will predict the accurate element targets detector responses plus the surrounding material effects on the elemental library spectra. Besides, the Monte Carlo code also calculates the detector efficiency which semi-empirical models cannot provide. The plot of detector efficiency with respect to incident energy is shown in Figure 7.

As observed from Figure 7, detector efficiency decreases as incident energy increases, which indicates that more X-rays are intended to escape from the detector as incident X-ray energy increases.

Figures 8 and 9 show the Monte Carlo simulated surface flux spectrum and the pulse-height spectrum after convolving with detector response functions for nickel and lead element respectively. The figures present the Monte Carlo simulated surface flux spectrum in blue dashed line and the red solid lines show the library pulse-height spectrum after detector response function convolution. It is worthwhile to point out that the detector efficiency with respect to X-ray energy is calculated by Monte Carlo simulation and shown in y-coordinate.
Figure 8 Monte Carlo simulated surface flux and convolution with detector response functions for Nickel.

Figure 9 Monte Carlo simulated surface flux and convolution with detector response functions for Lead.
Conclusions

An improved DRF model has been implemented on micro-focused XRF analyzer which uses Si detector crystal. The model parameters are obtained by linear and nonlinear regression and the fitted DRF spectrum was compared with the experimental spectrum and they indicate good agreement. And the Monte Carlo simulation method was used to predict the full response of the micro-focused X-ray analyzer system and predict the detector efficiency accurately.

References


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